IN THE UNITED STATES DISTRICT COURT
FOR THE DISTRICT OF MARYLAND
NORTHERN DIVISION

G.W. ARU, LLC, AND COCHISE TECHNOLOGY, LLC Plaintiffs, v. W.R. GRACE & CO.-Conn, Defendant.	Civ. No. 1:22-cv-02636-JKB

DEFENDANT W.R. GRACE & CO.-CONN'S
INITIAL DISCLOSURE OF PRELIMINARY INVALIDITY CONTENTIONS

TABLE OF CONTENTS

I. INTRODUCTION 1
II. SUMMARY OF INVALIDITY CONTENTIONS 1
III. BACKGROUND OF THE PURPORTED INVENTION / STATE OF THE ART 2
A. Eggshell Catalyst 4
B. Particle Size 7
IV. INVALIDITY UNDER AIA 35 U.S.C. §§ 102 AND 103 9
A. Applicant Admitted Prior Art (AAPA) 10
B. Prior Art Under AIA 35 U.S.C. § 102 10
C. Prior Art Under AIA 35 U.S.C. § 103 12
D. Motivation to Combine 15
E. Additional Systems and/or Products Used, Known of, and/or Offered for Sale that Alone or in Combination Anticipate or Render Obvious One or More of the Asserted Claims 18
V. INVALIDITY UNDER AIA 35 U.S.C. § 112(a) and/or (b) 19
VI. ACCOMPANYING DOCUMENT PRODUCTION 19
VII. DEFENDANT'S RESERVATION OF RIGHTS 20

Cochise Technology Ex. 2013
W.R. Grace v. Cochise Technology - IPR 2023-01063

I. INTRODUCTION

Pursuant to the Court's Scheduling Order (D.I. 24) and Local Rule 804.1, Defendant W.R. Grace & Co.-Conn ("Grace") hereby provides its Initial Disclosure of Preliminary Invalidity Contentions ("Invalidity Contentions") for U.S. Patent No. 11,224,864 ("the '864 Patent") asserted by Plaintiffs G.W. Aru, LLC ("Aru") and Cochise Technology, LLC's ("Cochise") (collectively, "Plaintiffs"). Grace also serves contemporaneously herewith the accompanying production of documents as required by the Scheduling Order and the Local Rules.

II. SUMMARY OF INVALIDITY CONTENTIONS

Plaintiffs have alleged that Grace infringes claims 1, 3, 6, 12, and 15 of the '864 Patent (the "Asserted Claims"). Subject to its Reservations of Right set forth in Section VII below, Grace contends the Asserted Claims are invalid under one or more of AIA 35 U.S.C. §§ 102, 103, and 112, for at least the following reasons:

- One or more of the Asserted Claims are invalid under 35 U.S.C. § 102 (AIA) because a single prior art reference, either alone or in combination with the knowledge of one of ordinary skill in the art, expressly or inherently discloses each and every limitation of the purported invention;
- One or more of the Asserted Claims are invalid under 35 U.S.C. § 103 (AIA) because single or multiple prior art references, in combination with the knowledge of one having ordinary skill in the art, renders them obvious;
- One or more of the Asserted Claims are invalid as indefinite under 35 U.S.C. § 112(b) (AIA) because they fail to particularly point out and distinctly claim the subject matter that the applicant regards as the invention;

Cochise Technology Ex. 2013
W.R. Grace v. Cochise Technology - IPR 2023-01063

- One or more of the Asserted Claims are invalid for failure to satisfy the written description requirement under 35 U.S.C. § 112(a) (AIA) because the specification of the ’864 Patent does not show that the inventor had possession of the claimed inventions as of the filing date of the Asserted Claims;
- One or more of the Asserted Claims are invalid for failure to satisfy the enablement requirement under 35 U.S.C. § 112(a) (AIA) because the specification of the ’864 Patent does not disclose the manner and process of making and using the invention as of the filing date of the Asserted Claims.

Grace’s invalidity contentions under AIA 35 U.S.C. §§ 102 and 103 are made in the alternative to its contentions that the Asserted Claims should be invalidated under AIA 35 U.S.C. § 112 (for indefiniteness, lack of written description, and/or lack of enablement). *See, e.g.*, Fed. R. Civ. P. 8(d).

III. BACKGROUND OF THE PURPORTED INVENTION / STATE OF THE ART

As described and/or admitted in the ’864 patent, CO to CO_2 combustion promoters were well known and used in a fluid catalytic cracking (FCC) unit as of the priority date for the ’864 patent. ’864 pat., at 1:26-27. FCC is a process that converts heavy fractions of petroleum oil to lighter products which are in greater demand. In FCC processes, fluidizable catalyst particles are contacted in a conversion zone of high temperatures with hydrocarbons in order to crack higher boiling hydrocarbons to lower boiling hydrocarbons. *Id.* at 1:27-31. Common components of catalyst particles are an aluminosilicate component and a porous inorganic oxide matrix. *Id.* at 31-34. This type of catalyst must be regenerated to low carbon levels to assure that the catalyst particles possess desired activity and selectivity before the particles are recycled to the conversion or cracking zone. *Id.* at 34-39.

CO to CO_2 combustion promoters are commonly made from impregnation of Group VIII noble metal or metals, commonly platinum and/or palladium, onto a porous alumina and/or other substrate microspheres of average particle size 60 to 90 microns. *Id.* at 1:62-67. According to the '864 patent, a disadvantage of noble metal-based CO to CO_2 combustion promoters is that relatively large amounts of noble metal are required to achieve the desired CO combustion in the FCC process. *Id.* at 2: 14-17. This is allegedly because impregnation of the noble metals results in a uniform distribution of the Group VIII noble metal throughout the internal and external surfaces of the porous microsphere. *Id.* at 1:67-3.

As of 2018, there were two known structures of catalytic particles—uniform distribution and non-uniform distribution. Uniform distribution results in impregnation of Group VIII noble metals in a homogeneous concentration throughout the catalyst particle. Non-uniform distribution, by contrast, results in a heterogeneous concentration. An "eggshell" catalyst is an example of non-uniform distribution, because there is a greater concentration of noble metal on the outer "shell" of the particle and decreased concentration of noble metal in the core (or "yolk") of the particle. The cost of noble metals necessary for activity of the catalyst would motivate a person of ordinary skill in the art to select an eggshell distribution, thereby reducing the amount of noble metal while retaining activity of the catalyst. Indeed, Gavriilidis[1] explains that the superiority of eggshell catalysts in conversion, selectivity, durability, and thermal sensitivity characteristics over uniform catalysts was known in the art as early as the 1960s. Harold[2] likewise touts the benefits of non-uniform catalyst over uniform catalysts and in particular the effect of diffusion of metal into the

[1] "*Optimal Distribution of Catalyst in Pellet*" ("Gavriilidis"), authored by Asterios Gavriilidis et al. and published in 1993.
[2] "*Impact of the Catalytic Activity Profile on Observed Multiplicity Features: CO Oxidation on* Pt/Al_2O_3" ("Harold") authored by Michael P. Harold et al. and published in 1987.

Cochise Technology Ex. 2013
W.R. Grace v. Cochise Technology - IPR 2023-01063

surface of the catalyst pellet in the characteristics and performance of the catalyst as early as the 1980s.

A. Eggshell Catalyst

Eggshell catalysts were known in the art prior to the priority filing date of the '864 patent. For example, Miura[3] discloses "a metal oxide particle comprising a core part relatively rich in a component originated in the population of first colloid particles and a surface layer relatively rich in a component originated in the population of second colloid particles can be obtained." Miura, [0014]. Miura therefore discloses a surface layer (i.e., outer region) of a catalyst that is relatively rich in particles "selected from the group consisting of alumina, ceria, zirconia and titania colloid particles group," and may be ceria. *Id.*, [0023]-[0026]. Miura further discloses that a "noble metal such as platinum, rhodium and palladium is supported on . . . the surface layer" wherein platinum is the most preferable. *Id.* [0048], [0072]. Miura depicts this structure—referred to in the art as an "eggshell"—in FIG. 2, showing a surface layer (12) of particulate support comprising CeO_2 which covers an entire core part (11) formed of ZrO_2, and a noble metal (13), such as platinum, supported on the particulate support. *Id.*, FIG. 2, [0068].

Similarly, Lin[4] discloses "an eggshell catalyst and process for preparation of the same," including "single- or multiple-metal eggshell of the finished catalyst." Lin, 1. Lin further teaches that the "eggshell" structure is known in the art and discloses an eggshell catalyst wherein the "[m]etal is distributed on the outer layer of the support (with eggshell shape), which is usually used in fast reactions and capable of improving the selectivity and yield of the desired products." *Id.*, 1-2. Lin teaches that the metal catalysts with eggshell active metal profile "preferably

[3] U.S. Patent Application No. 2007/0197379 A1 ("Miura"), filed on April 27, 2004 and published on August 23, 2007.
[4] UK Patent Application No. GB 2,310,384 ("Lin"), filed on February 22, 1996 and published on August 27, 1997.

comprises Pd, Ni, Co, Ho, Cu, Pt, Fe, Ag, Tr, Pb, Re, Ti, Sn, V, and Zn, etc." *Id.*, Abstract, 3. Lin shows an example catalyst having a "linear decrease of Pd metal distribution from outer layer toward spherical core." *Id.*, 10. Lin provides a comparison of a uniform particle in Figure 1 and an eggshell particle in Figure 2, and it demonstrates the linear decrease of Pd metal distribution in Figure 2 showing the Pd distribution in an eggshell catalyst from electron probe microanalyser:

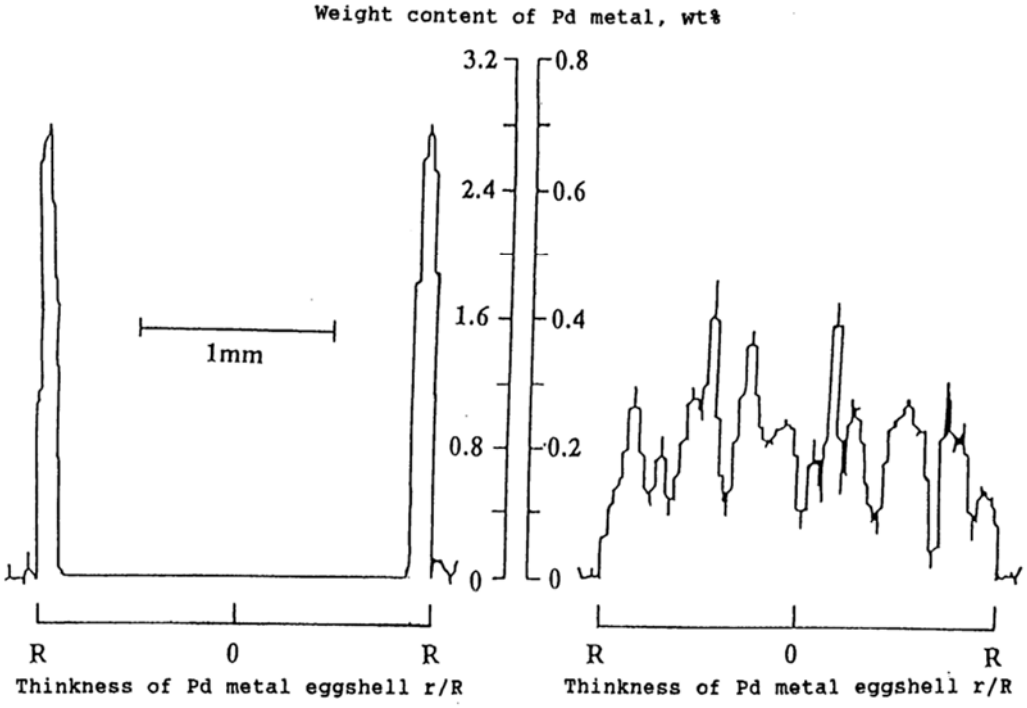


Fig.2 Fig.1

Id., 3-4, Fig. 1, Fig. 2.

Likewise, Schmidt[5] teaches an improved catalyst particle wherein the surface layer of the particle comprises platinum metal in a concentration at least 1.5 times the concentration of the core of each particle. Schmidt, 3:15-35. Similarly, Hagemeyer[6] teaches a method for producing a

[5] U.S. Patent Publication No. 5,017,541 ("Schmidt"), filed on June 27, 1990 and published on May 21, 1991.
[6] U.S. Patent Application No. 2010/0190638 ("Hagemeyer"), filed on May 31, 2007 and published on July 29, 2010.

Cochise Technology Ex. 2013
W.R. Grace v. Cochise Technology - IPR 2023-01063

"shell" catalyst wherein the outer shell contains metal in a higher concentration compared to the core. Hagemeyer, Abstract. Shyr[7] teaches a catalyst comprising a "shell" of Pt and examines the benefits of a catalyst particle having a "sharply defined shell of absorbed Pt." Shyr, 428-29.

The prior art also demonstrates that non-uniform catalysts were well-known in the art prior to the claimed invention wherein the active or noble metals are distributed in a heterogeneous or varied concentration throughout the catalyst particle. For example, Munnik[8] teaches a "heterogeneous" catalyst distinct from a uniform catalyst in the concentration of metal throughout the particle. Munnik, 6687. In particular, Munnik teaches an eggshell distribution wherein the concentration of Pd is higher in the "shell" of the catalyst. *Id.* at 6697-98. As another example, Papageorgiou[9] discloses a method of creating a catalyst where Group VIII noble metals are dispersed on the surface area of a support layer resulting in an eggshell catalyst with a higher concentration of platinum in the surface layer of the particle. Papageorgiou, 1, 11. Similarly, Carlsson[10] teaches an eggshell catalyst formed by impregnating an aluminum-based catalyst support with a solution containing metal compounds such that the metal is concentrated in the outer layer on the support. Carlsson, Abstract. Espinoza[11] likewise discloses a catalyst comprising an inner core and outer layer wherein the outer layer contains catalytically active metal, such as a Group VIII metal, which is *not* deposited in the core of the particle. Espinoza, 4:26-47. Witte[12]

[7] "*Preparation of Nonuniformly Active Catalysts*" ("Shyr"), authored by Yen-Shin Shyr et al. and published in 1980.
[8] "*Recent Developments in the Synthesis of Supported Catalysts*" ("Munnik"), authored by Peter Munnik et al. and published on June 19, 2015.
[9] "*Preparation of Pt/γ-Al_2O_3 Pellets with Internal Step-Distribution of Catalyst: Experiments and Theory*" ("Parageorgiou"), authored by Panayiotis Papageorgiou et al. and published in 1996.
[10] U.S. Patent No. 9,981,252 ("Carlsson"), filed on May 1, 2009 and published on May 29, 2018.
[11] U.S. Patent No. 7,230,035 ("Espinoza"), filed on December 30, 2002 and published on June 12, 2007.
[12] U.S. Patent No. 10,258,967 ("Witte"), filed on March 20, 2015 and published on April 16, 2019.

similarly teaches a method of depositing metal salt onto the surface of the support such that the metal is not deposited within the pores of the support and is only present on the edge or outer layer of the catalyst. Witte, 1:29-47. Each of Venuto,[13] Rainer,[14] and Tran[15] likewise teaches a non-uniform catalyst wherein the amount of metal is varied considerably to achieve a desired level of CO combustion promoter by distributing metals in separate areas of the particle. Venuto, 30-31; Rainer, [0028], [0050]; Tran, Abstract (disclosing a hollow porous metal oxide microsphere).

B. Particle Size

During prosecution, the applicant of the '864 patent submitted that the term "microsphere" is "a term of art, the metes and bound of which are readily understood by one of ordinary skill in the art," and supported this argument with a declaration by Dr. Natalie Herring, Ph.D. '864 Prosecution History, 94, 143-44. The applicant defined "microsphere sized porous particles" as those "***having a diameter of less than 1 mm***." *Id.*, 85. The '864 patent further acknowledges that "CO to CO_2 promoters are ***well known***" and "are ***commonly made*** from impregnation of Group VIII noble metal or metals, commonly platinum and/or palladium, onto a porous alumina and/or other substrate ***microspheres of average particle size 60 to 90 microns***." '864 patent, 1:26, 1:62-66; *see also* '864 Prosecution History, 94, 143-44.

As the applicant admitted during prosecution and in the '864 patent, microsphere particle sizes were known in the art before the priority date of the '864 patent. As to the average particle

[13] "*Fluid Catalytic Cracking with Zeolite Catalysts*" ("Venuto"), authored by Paul Venuto et al. and published in 1979.
[14] U.S. Patent Application No. 2009/0050529 ("Rainer"), filed on June 8, 2007 and published on February 26, 2009.
[15] U.S. Patent No. 9,358,533 ("Tran"), filed on December 20, 2012 and published on June 7, 2016.

size, Baker,[16] for example, teaches that the support particles are "microspheroidal." *Id.*, 1:60-67, 3:34-46. Baker specifically teaches that the catalyst has the following particle size distribution:

0 to 20 microns	0–30 wt %
20 to 44 microns	0–60 wt %
44 to 88 microns	10–80 wt %
88 to 106 microns	0–80 wt %
>106 microns	0–40 wt %
>300 microns	0–5 wt %

id., 3:16-33. Therefore, Baker teaches microsphere sized porous particles having an average diameter size of less than 1mm.

As additional examples, Espinoza teaches that particle size ranging from 0.5 mm to 0.25 mm are most useful in fluidized bed catalyst reactions. Espinoza, 7:51-67. Witte teaches a particle size ranging from 1 to 500 microns and a preferable size distribution of particle size between 10 and 200 microns. Witte, 5:60-6:2. Tran discloses a microsphere sized particle with a particle size ranging from about 5-100 microns. Tran, 1:13-29.

Miura also discloses a range of average particle diameters of the metal oxide particles as, "less than 10 μm, less than 5 μm, less than 1 μm, less than 500 nm, less than 200 nm, less than 100 nm or less than 50 nm." Miura, [0015], [0068]. Therefore, Miura discloses particles having an average diameter size of less than 1mm. Miura further teaches benefits of using small particle size in the disclosed combustion promoter. Miura, [0068].

Lin also discloses "an eggshell-shaped catalyst for use in selective reduction or oxidation," including, for example, oxidation of CO, wherein a catalyst was prepared using "spherical particles with diameter of 2mm." Lin, 1-2, 5, 13. Hagemeyer similarly teaches a preferred particle size ranging from 2mm to 9mm depending on the application of the catalyst material. Hagemeyer,

[16] U.S. Patent No. 7,053,024 ("Baker"), filed on January 9, 2003 and published on May 30, 2006.

[0130]. Likewise, Schmidt teaches a particle size ranging from 0.75 mm to 3.3 mm wherein the preferred size is 1.6 mm. Schmidt, 5:3-35.

IV. INVALIDITY UNDER AIA 35 U.S.C. §§ 102 AND 103

Subject to Grace's Reservation of Rights set forth herein, Grace identifies each item of prior art that anticipates the Asserted Claims and/or renders the Asserted Claims obvious under AIA 35 U.S.C. §§ 102 and 103. Charts identifying where specifically in each alleged item of prior art each limitation of each Asserted Claim is found are identified below and attached to Grace's Invalidity Contentions as Exhibits A-Q. For those prior art references that render the Asserted Claims obvious, an explanation of why the prior art renders the Asserted Claim obvious, including an identification of any combinations of prior art showing obviousness, is set forth in each reference's chart and in Section IV.B, below. Grace relies on each prior art reference as a whole, for all of the teachings or disclosures it may make to a person of ordinary skill in the art (POSITA) at the time the claimed invention was allegedly made. The exemplary teachings cited in the claim charts are thus representative only, and Grace reserves the right to rely on additional teachings from the asserted references—whether or not such teachings are explicitly cited in the attached claim charts. Grace may also rely on other documents and information, including cited references and prosecution histories for the '864 Patent (and related patents and/or patent applications) and witness testimony, including expert testimony, to explain, amplify, illustrate, demonstrate, provide context, or aid in understanding the cited portions of the references and/or the background knowledge or understanding of a POSITA.

Grace anticipates that additional prior art references will be identified as the case progresses based on information obtained during discovery from Plaintiffs, Grace's internal investigations, the Court's rulings including any further claim construction, and documents and

Cochise Technology Ex. 2013
W.R. Grace v. Cochise Technology - IPR 2023-01063

evidence obtained from third parties. Grace therefore reserves the right to supplement its Invalidity Contentions accordingly.

A. Applicant Admitted Prior Art (AAPA)

Admissions by an applicant in the specification and during prosecution constitute "an admission which can be relied upon for both anticipation and obviousness determinations, regardless of whether the admitted prior art would otherwise qualify as prior art under the statutory categories of 35 U.S.C. [§] 102." MPEP § 2129 (citing *Riverwood Int'l Corp. v. R.A. Jones & Co.*, 324 F.3d 1346, 1354 (Fed. Cir. 2003); *Constant v. Advanced Micro-Devices Inc.*, 848 F.2d 1560, 1570 (Fed. Cir. 1988)).

B. Prior Art Under AIA 35 U.S.C. § 102

The Asserted Claims are invalid under AIA 35 U.S.C. § 102 because "the claimed invention was patented, described in a printed publication, or in public use, on sale, or otherwise available to the public before the effective filing date of the claimed invention." AIA 35 U.S.C. § 102(a). Alternatively, the Asserted Claims are invalid because "the claimed invention was described in a patent issued under section 151, or in an application for patent published or deemed published under section 122(b), in which the patent or application, as the case may be, names another inventor and was effectively filed before the effective filing date of the claimed invention." *Id*. To be anticipatory, a reference must describe, either expressly or inherently, each and every claim limitation and enable one of skill in the art to practice an embodiment of the claimed invention without undue experimentation. *Iovate Health Sciences, Inc. v. Bio-Engineered Supplements & Nutrition Inc.*, 586 F.3d 1376, 1380 (Fed. Cir. 2009). A reference may anticipate inherently if a claim limitation that is not expressly disclosed "is necessarily present, or inherent, in the single anticipating reference." *In re Aoyama*, 656 F.3d 1293, 1337 (Fed. Cir. 2011) (quoting *Schering Corp. v. Geneva Pharm., Inc.*, 339 F.3d 1373, 1377 (Fed. Cir. 2003)); *Akamai*

Technologies, Inc. v. Cable & Wireless Internet Services, Inc., 344 F.3d 1186, 1195 (Fed. Cir. 2003). The following prior art patents, printed publications, and AAPA anticipate one or more of the Asserted Claims of the '864 Patent by expressly or inherently disclosing each and every limitation of those claims under AIA 35 U.S.C. § 102 for the reasons set forth below and in Exhibits A-Q:

- **AAPA**: As shown in Exhibit A attached hereto, claims 1, 3, 6, 12, and 15 of the '864 Patent are invalid under AIA 35 U.S.C. § 102.
- **Baker**: As shown in Exhibit B attached hereto, claims 1, 3, 6, 12, and 15 of the '864 Patent are invalid under AIA 35 U.S.C. § 102.
- **Carlsson**: As shown in Exhibit C attached hereto, claims 1, 3, 6, 12, and 15 of the '864 Patent are invalid under AIA 35 U.S.C. § 102.
- **Espinoza**: As shown in Exhibit D attached hereto, claims 1, 3, 6, 12, and 15 of the '864 Patent are invalid under AIA 35 U.S.C. § 102.
- **Gavriilidis**: As shown in Exhibit E attached hereto, claims 1, 3, 6, 12, and 15 of the '864 Patent are invalid under AIA 35 U.S.C. § 102.
- **Hagemeyer**: As shown in Exhibit F attached hereto, claims 1, 3, 6, 12, and 15 of the '864 Patent are invalid under AIA 35 U.S.C. § 102.
- **Harold**: As shown in Exhibit G attached hereto, claims 1, 3, 6, 12, and 15 of the '864 Patent are invalid under AIA 35 U.S.C. § 102.
- **Lin**: As shown in Exhibit H attached hereto, claims 1, 3, 6, 12, and 15 of the '864 Patent are invalid under AIA 35 U.S.C. § 102.
- **Miura**: As shown in Exhibit I attached hereto, claims 1, 3, 6, 12, and 15 of the '864 Patent are invalid under AIA 35 U.S.C. § 102.
- **Munnik**: As shown in Exhibit J attached hereto, claims 1, 3, 6, 12, and 15 of the '864 Patent are invalid under AIA 35 U.S.C. § 102.
- **Papageorgiou**: As shown in Exhibit K attached hereto, claims 1, 3, 6, 12, and 15 of the '864 Patent are invalid under AIA 35 U.S.C. § 102.
- **Rainer**: As shown in Exhibit L attached hereto, claims 1, 3, 6, 12, and 15 of the '864 Patent are invalid under AIA 35 U.S.C. § 102.

- **Schmidt**: As shown in Exhibit M attached hereto, claims 1, 3, 6, 12, and 15 of the ’864 Patent are invalid under AIA 35 U.S.C. § 102.

- **Shyr**: As shown in Exhibit N attached hereto, claims 1, 3, 6, 12, and 15 of the ’864 Patent are invalid under AIA 35 U.S.C. § 102.

- **Tran**: As shown in Exhibit O attached hereto, claims 1, 3, 6, 12, and 15 of the ’864 Patent are invalid under AIA 35 U.S.C. § 102.

- **Venuto**: As shown in Exhibit P attached hereto, claims 1, 3, 6, 12, and 15 of the ’864 Patent are invalid under AIA 35 U.S.C. § 102.

- **Witte**: As shown in Exhibit Q attached hereto, claims 1, 3, 6, 12, and 15 of the ’864 Patent are invalid under AIA 35 U.S.C. § 102.

Discovery is ongoing, and Grace reserves the right to identify prior art patents and printed publications that anticipate one or more of the Asserted Claims of the ’864 Patent by expressly or inherently disclosing each and every limitation of those claims under AIA 35 U.S.C. § 102.

C. Prior Art Under AIA 35 U.S.C. § 103

The Asserted Claims are also invalid under AIA 35 U.S.C. § 103 because “the differences between the claimed invention and the prior art are such that the claimed invention as a whole would have been obvious before the effective filing date of the claimed invention to a person having ordinary skill in the art to which the claimed invention pertains.” AIA 35 U.S.C. § 103(a). A claimed invention is obvious when a skilled artisan would have been motivated to combine the teaching of the prior art references to achieve the claimed invention, and the skilled artisan would have had a reasonable expectation of success in doing so. *See, e.g., Unigene Labs., Inc. v. Apotex, Inc.,* 655 F.3d 1352, 1360 (Fed. Cir. 2011). “Any need or problem known in the field of endeavor at the time of invention and addressed by the patent can provide a reason for combining the elements in the manner claimed.” *See KSR Int’l Co. v. Teleflex Inc.,* 550 U.S. 398, 420 (2007). While an analysis of any teaching, suggestion, or motivation to combine elements from different prior art references is useful in an obviousness analysis, the overall inquiry must be expansive and

flexible, and must include the common sense and ordinary creativity of a person having ordinary skill in the art as part of the analysis. *Id.* at 419-21.

To the extent the above references are found not to anticipate the Asserted Claims of the '864 patent, one or more of the Asserted Claims is invalid under AIA 35 U.S.C. § 103 in view of the prior art patents, printed publications, and AAPA described above either alone or in combination with other references and/or the knowledge of a person having ordinary skill in the art, as described therein. The following prior art patents, printed publications, and AAPA render obvious one or more of the Asserted Claims of the '864 Patent under AIA 35 U.S.C. § 103 for the reasons set forth below and in Exhibits A-Q:

- **AAPA**: As shown in Exhibit A attached hereto, claims 1, 3, 6, 12, and 15 of the '864 Patent are invalid under AIA 35 U.S.C. § 103(a) in view of AAPA alone or in combination with other references and/or the knowledge of one skilled in the art, as described therein.

- **Baker**: As shown in Exhibit B attached hereto, claims 1, 3, 6, 12, and 15 of the '864 Patent are invalid under AIA 35 U.S.C. § 103 in view of Baker alone or in combination with other references and/or the knowledge of one skilled in the art, as described therein.

- **Carlsson**: As shown in Exhibit C attached hereto, claims 1, 3, 6, 12, and 15 of the '864 Patent are invalid under AIA 35 U.S.C. § 103 in view of Carlsson alone or in combination with other references and/or the knowledge of one skilled in the art, as described therein.

- **Espinoza**: As shown in Exhibit D attached hereto, claims 1, 3, 6, 12, and 15 of the '864 Patent are invalid under AIA 35 U.S.C. § 103 in view of Espinoza alone or in combination with other references and/or the knowledge of one skilled in the art, as described therein.

- **Gavriilidis**: As shown in Exhibit E attached hereto, claims 1, 3, 6, 12, and 15 of the '864 Patent are invalid under AIA 35 U.S.C. § 103 in view of Gavriilidis alone or in combination with other references and/or the knowledge of one skilled in the art, as described therein.

- **Hagemeyer**: As shown in Exhibit F attached hereto, claims 1, 3, 6, 12, and 15 of the '864 Patent are invalid under AIA 35 U.S.C. § 103 in view of Hagemeyer alone or in combination with other references and/or the knowledge of one skilled in the art, as described therein.

Cochise Technology Ex. 2013
W.R. Grace v. Cochise Technology - IPR 2023-01063

- **Harold**: As shown in Exhibit G attached hereto, claims 1, 3, 6, 12, and 15 of the ’864 Patent are invalid under AIA 35 U.S.C. § 103 in view of Harold alone or in combination with other references and/or the knowledge of one skilled in the art, as described therein.

- **Lin**: As shown in Exhibit H attached hereto, claims 1, 3, 6, 12, and 15 of the ’864 Patent are invalid under AIA 35 U.S.C. § 103 in view of Lin alone or in combination with other references and/or the knowledge of one skilled in the art, as described therein.

- **Miura**: As shown in Exhibit I attached hereto, claims 1, 3, 6, 12, and 15 of the ’864 Patent are invalid under AIA 35 U.S.C. § 103 in view of Miura alone or in combination with other references and/or the knowledge of one skilled in the art, as described therein.

- **Munnik**: As shown in Exhibit J attached hereto, claims 1, 3, 6, 12, and 15 of the ’864 Patent are invalid under AIA 35 U.S.C. § 103 in view of Munnik alone or in combination with other references and/or the knowledge of one skilled in the art, as described therein.

- **Papageorgiou**: As shown in Exhibit K attached hereto, claims 1, 3, 6, 12, and 15 of the ’864 Patent are invalid under AIA 35 U.S.C. § 103 in view of Papageorgiou alone or in combination with other references and/or the knowledge of one skilled in the art, as described therein.

- **Rainer**: As shown in Exhibit L attached hereto, claims 1, 3, 6, 12, and 15 of the ’864 Patent are invalid under AIA 35 U.S.C. § 103 in view of Rainer alone or in combination with other references and/or the knowledge of one skilled in the art, as described therein.

- **Schmidt**: As shown in Exhibit M attached hereto, claims 1, 3, 6, 12, and 15 of the ’864 Patent are invalid under AIA 35 U.S.C. § 103 in view of Schmidt alone or in combination with other references and/or the knowledge of one skilled in the art, as described therein.

- **Shyr**: As shown in Exhibit N attached hereto, claims 1, 3, 6, 12, and 15 of the ’864 Patent are invalid under AIA 35 U.S.C. § 103 in view of Shyr alone or in combination with other references and/or the knowledge of one skilled in the art, as described therein.

- **Tran**: As shown in Exhibit O attached hereto, claims 1, 3, 6, 12, and 15 of the ’864 Patent are invalid under AIA 35 U.S.C. § 103 in view of Tran alone or in combination with other references and/or the knowledge of one skilled in the art, as described therein.

- **Venuto**: As shown in Exhibit P attached hereto, claims 1, 3, 6, 12, and 15 of the ’864 Patent are invalid under AIA 35 U.S.C. § 103 in view of Venuto alone or in

Cochise Technology Ex. 2013
W.R. Grace v. Cochise Technology - IPR 2023-01063

combination with other references and/or the knowledge of one skilled in the art, as described therein.

- **Witte**: As shown in Exhibit Q attached hereto, claims 1, 3, 6, 12, and 15 of the '864 Patent are invalid under AIA 35 U.S.C. § 103 in view of Witte alone or in combination with other references and/or the knowledge of one skilled in the art, as described therein.

A POSITA would have been motivated to combine these prior art references with one another and/or with the knowledge of a POSITA to achieve the claimed invention, and would have had a reasonable expectation of success in doing so.

D. Motivation to Combine

A POSITA would have appreciated the benefits of using small particle size in a combustion promoter. In particular, a POSITA would have considered the use of microsphere particles in an eggshell structure combustion promoter.

As of 2018, an eggshell catalyst structure was well-known in the art with particle sizes in the microsphere size ranging from microns to nanometers. The applicant admitted during prosecution that eggshell catalysts were known in the art. *See* Prosecution History of '864 pat. at 27. As described above, Miura, Lin, and Schmidt each teach an eggshell structure catalyst. Similarly, Hagemeyer teaches a method for producing a "shell" catalyst wherein the outer shell contains metal in a higher concentration compared to the core. Hagemeyer, Abstract. Shyr likewise teaches a catalyst comprising a "shell" of Pt and examines the benefits of a catalyst particle having a "sharply defined shell of absorbed Pt." Shyr, 428-29.

As admitted by applicant and further demonstrated in the prior art, in particular Gavriilidis and Harold, heterogeneous catalysts were well-known in the art prior the claimed invention. For example, Munnik teaches a "heterogeneous" catalyst distinct from a uniform catalyst in the concentration of metal throughout the particle. Munnik, 6687. That is, Munnik teaches that a heterogeneous catalyst has a higher concentration of metals in the outer region than the inner

core/region. As another example, Papageorgiou discloses a similar method of creating a catalyst wherein Group VIII noble metals are dispersed on the surface area of the support layer. Papageorgiou, 1. Similarly, Carlsson teaches an eggshell catalyst formed by impregnating an aluminum-based catalyst support with a solution containing metal compounds such that the metal is concentrated in the outer layer on the support. Carlsson, Abstract. Further, Espinoza teaches a similar eggshell structure wherein only the outer layer of the particle contains catalytically active metal, which is not deposited in the core of the particle. Espinoza, 4:26-47. Witte similarly teaches catalyst wherein the active metal is not deposited within the pores of the support and is only present on the edge or outer layer of the catalyst. Witte, 1:29-47. Each of Venuto, Rainer, and Tran likewise teaches a non-uniform catalyst wherein the amount of metal is varied considerably to achieve a desired level of CO combustion promoter. Venuto, 30-31; Rainer, [0050]; Tran, Abstract (disclosing a hollow porous metal oxide microsphere).

Further, the applicant admitted that CO to CO_2 combustion promoters are well known in the art and "commonly made from impregnation of Group VIII noble metal or metals, commonly platinum and/or palladium, onto a porous alumina and/or other substrate microspheres of average particle size 60 to 90 microns," '864 pat. at 1:62-2:3, and therefore particles having an average diameter size of less than 1mm was known for use in CO to CO_2 combustion promoters. Likewise, the applicant admitted during prosecution that a POSITA would be familiar with "microspheres" in the context of CO to CO_2 combustion promoters as "***spherical particles as small as 1 micron in diameter and as large as 1,000 microns (1 mm).***" Prosecution History of the '864 pat. at 94 (internal citations omitted). As described above, Baker teaches microsphere-sized porous particles having an average diameter size of less than 1mm. Further as described above, Espinoza teaches

that particle size ranging from 0.5 mm to 0.25 mm are most useful in fluidized bed catalyst reactions. Espinoza, 7:51-67.

A POSITA would have recognized that the particles with an average particle size less than 1mm may be used in conjunction with the heterogeneous catalyst particles disclosed in the prior art, including Miura, Hagemeyer, Munnik, Papageorgiou, Carlsson, Espinoza, Venuto, Rainer, and Tran. For example, a POSITA would have recognized that the prior art each teaches similar catalyst structures and components of the combustion promoters, including silica, aluminum, and Group VIII metals and, in particular, Miura teaches benefits of small particle diameter. Miura, [0068]. By incorporating the specific particle size range of Baker to the prior art combustion promoters, the properties of the prior art combustion promoter can be further improved through the specific particle size teachings of Baker. Moreover, the applicant admitted that such "microsphere-sized" particles were well-known by a POSITA at the time of the claimed invention. Prosecution History of the '864 pat. at 94 (internal citations omitted). Similarly, a POSITA would have recognized that the particles with an average particle size less than 1mm may be substituted for the 2mm particles disclosed in Lin and Hagemeyer or the 1.6mm particles disclosed in Schmidt. For example, Lin, Hagemeyer, Schmidt, and Baker each teaches similar catalyst structure and similar components of the combustion promoters, including silica, aluminum, and Group VIII metals and the improved selectivity and yield of the eggshell shaped catalyst. Lin, 1-2; Hagemeyer, [0130]; Schmidt, 3:15-35. By incorporating the specific particle size range of Baker to Lin, Hagemeyer, or Schmidt, a POSITA would have recognized that the properties of Lin's, Hagemeyer's, or Schmidt's combustion promoter can be further improved through the specific particle size teachings of Baker.

A POSITA would have been further motivated to pursue this implementation of the well-known microsphere-sized particles, Baker's, or Espinoza's specific particle size into the prior art combustion promoters as doing so is merely the application of known techniques (*e.g.*, using small particles) to a known structure (*e.g.*, combustion promoter material) to yield predictable results (*e.g.*, improved performance). A POSITA would have expected success in the combination given that Baker and Espinoza each describes using sub-1mm particles for improved catalytic performance.

Moreover, a POSITA would have appreciated that the size of the particle is determined by the application of the catalyst material. For example, a larger particle size may be more appropriate in the application of fluidized catalysts whereas a smaller particle size is appropriate in a fixed bed catalyst application. *See* Espinoza, 7:51-67. Therefore, a POSITA would have readily adjusted the size of the particle size for application in a CO to CO_2 combustion promoter.

E. Additional Systems and/or Products Used, Known of, and/or Offered for Sale that Alone or in Combination Anticipate or Render Obvious One or More of the Asserted Claims

Additionally, the following systems and/or products are invalidating prior art under at least AIA 35 U.S.C. § 102(a) and/or AIA 35 U.S.C. § 103. While Grace's investigation continues, information available to date indicates that each system and/or product was in public use, on sale, and/or otherwise known to the public in this country more than one year before the effective filing date of the claimed invention. As discovery is just beginning, Grace reserves the right to rely on testimony of witnesses (including third-party witnesses) knowledgeable about such systems and/or products, as well as documents that are currently in the possession of third parties describing these systems and products:

- Grace CP®3

- Grace CP®5

- Fluid catalytic cracking additives made, offered, and/or sold by Johnson Matthey Process Technologies, Inc., including but not limited to: Sulphur oxide (SOx) reduction FCC additives (e.g., Super SOxGetter™ and Lo-SOx™PB families and precursors), nitrogen oxide (NOx) reduction FCC additives (e.g., NONOX™ family and precursors), catalyst enhancement FCC additives (e.g., CAT-AID™ family and precursors), CO oxidation FCC additives (e.g., COP™-250, COP™-375, COP™-550, COP™-850, COP™-NP, and COP™-NP HD families and precursors), propylene butylene and octane improvement additives (e.g., Super Z™, ZMX™ B-HP, PropylMax™, IsoCat™ families and precursors), and bottoms cracking FCC additives (e.g., BCA-105™ families and precursors).

V. INVALIDITY UNDER AIA 35 U.S.C. § 112(a) and/or (b)

Subject to Grace's Reservation of Rights set forth herein, Grace identifies the following grounds for invalidity of the Asserted Claims based on indefiniteness, written description, and/or enablement:

- "each microsphere sized porous particle having a diameter of less than 1mm, and independently"

- "one or more Group VIII noble metals distributed in the particle as an eggshell such that a higher concentration of the one or more Group VIII noble metals is present in the outer region of the microsphere sized porous particle as compared to the concentration of the one or more Group VIII noble metals in the centre of the microsphere sized porous particle"

VI. ACCOMPANYING DOCUMENT PRODUCTION

Along with these contentions, Grace has produced prior art identified with the following Bates range: **GRACE_0058059 - GRACE_0058587**. Additional prior art can be found in the file histories for the '864 Patent, as well as related patents and/or patent applications, all of which are incorporated by reference fully herein.

Grace's search for prior art references, additional documentation, and/or corroborating evidence, including witness' testimony concerning prior art systems, is ongoing. Accordingly, Grace reserves the right to continue to supplement its production as Grace obtains additional prior

Cochise Technology Ex. 2013
W.R. Grace v. Cochise Technology - IPR 2023-01063

art references, documentation, testimony and/or corroborating evidence concerning invalidity during the course of discovery.

VII. DEFENDANT'S RESERVATION OF RIGHTS

Grace's Invalidity Contentions address only those claims of the '864 Patent identified as being asserted in Plaintiffs' Initial Disclosure of Infringement Contentions. Grace's contentions reflect its current knowledge and contentions as of this early date in this Action. Grace's contentions are based in whole or in part on its present understanding of the Asserted Claims and Plaintiffs' apparent position as to the scope of the Asserted Claims. Grace reserves the right to seek leave to revise, amend, modify and/or supplement its Invalidity Contentions. In addition, Grace reserves the right to raise additional prior art and invalidity defenses not included in these Invalidity Contentions based on additional discovery or other issues raised by Plaintiffs in this Action or any related action. Grace further reserves the right to seek leave to amend these Invalidity Contentions should, for example, Plaintiffs provide any information that they failed to provide in its disclosures or should any third party provide relevant discovery.

Grace's Invalidity Contentions should not be construed as any admission regarding the proper construction of any Asserted Claim or deemed to represent or limit claim constructions that Grace has proposed or advanced, or will propose or advance, in this Action. Further, Grace reserves the right to seek leave to amend the information provided herein, including identifying and relying on additional references based on Grace's ongoing discovery. Grace reserves the right to rely on witness testimony about the prior art references to supplement these contentions, where appropriate. Moreover, Grace reserves the right to revise its contentions concerning the invalidity of the Asserted Claims, which may change depending upon the Court's construction of the Asserted Claims, any findings as to the priority date of the Asserted Claims, and/or positions that

Cochise Technology Ex. 2013
W.R. Grace v. Cochise Technology - IPR 2023-01063

Plaintiffs or their fact or expert witnesses may take concerning claim construction, infringement, and/or invalidity issues.

Grace reserves the right to present additional items of prior art or further support for current prior art under 35 U.S.C. §§ 102, 103, and/or 112 should discovery or further investigation show that any Asserted Claims of the ’864 Patent are not entitled to the priority date of the earlier filed patent application as Plaintiffs or Grace contends.

Respectfully submitted,

FISH & RICHARDSON P.C.

Dated: May 15, 2023

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Cochise Technology Ex. 2013
W.R. Grace v. Cochise Technology - IPR 2023-01063

CERTIFICATE OF SERVICE

The undersigned hereby certifies that a true and correct copy of DEFENDANT W.R. GRACE & CO.-CONN'S INITIAL DISCLOSURE OF PRELIMINARY INVALIDITY CONTENTIONS has been served on May 15, 2023, to all counsel of record who are deemed to have consented to electronic service via the Court's CM/ECF system.

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Cochise Technology Ex. 2013
W.R. Grace v. Cochise Technology - IPR 2023-01063

EXHIBIT A

EXHIBIT A

Invalidity of U.S. Patent No. 11,224,864 in View of Applicant Admitted Prior Art ("*AAPA*")

The Applicant Admitted Prior Art ("*AAPA*") described in U.S. Patent No. 11,224,864 ("the '864 patent") qualifies as prior art under at least 35 U.S.C. §§ 102 and/or 103 for the asserted claims of the '864 patent, as discussed in detail in the claim chart below.

The Court has not yet construed the claims and therefore the meaning of the terms in the claims has yet to be resolved by the Court. The support identified here for these limitations is responsive to Plaintiffs' infringement contentions, which Defendant disagrees with. As such, nothing in Defendant's claim charts should be construed as an admission regarding infringement, either literally or under the doctrine of equivalents, or as an admission regarding Defendant's understanding of the proper scope of the asserted claims. Defendant reserves the right to rely on additional citations or sources of evidence that also may be applicable, or that may become applicable in light of claim constructions, changes in Plaintiffs' infringement contentions, and/or information obtained during discovery as the case progresses.

To the extent Plaintiffs allege that *AAPA* does not disclose any particular limitation of the asserted claims of the '864 patent, either expressly or inherently, it would have been obvious to a person of ordinary skill in the art as of the priority date of the '864 patent to modify *AAPA* and/or to combine the teachings of *AAPA* with other prior art references, including but not limited to the present prior art references cited in the Cover Pleading and the relevant section(s) of claim charts for other prior art references for the '864 patent in a manner that would have rendered the asserted claims invalid as obvious.

Defendant reserves the right to amend or supplement this claim chart at a later date as more fully set forth in the Cover Pleading.

EXHIBIT A

Invalidity of U.S. Patent No. 11,224,864 in View of Applicant Admitted Prior Art ("*AAPA*")

'864 patent	**Applicant Admitted Prior Art ("*AAPA*")**
1[a] A CO to CO_2 combustion promoter comprising microsphere sized porous particles, each microsphere sized porous particle having a diameter of less than 1mm, and independently comprising	*AAPA* discloses a CO to CO_2 combustion promoter comprising microsphere sized porous particles, each microsphere sized porous particle having a diameter of less than 1mm, and independently comprising the limitations discussed in the following sections. *See, e.g.*, ['864 Patent, 1:25-2:20] "Such CO to CO_2 combustion promoters are well known and are used in a fluid catalytic cracking (FCC) unit. In such a FCC process fluidizable catalyst particles are contacted in a conversion zone at high temperatures with hydrocarbons such to crack higher boiling hydrocarbons to lower boiling hydrocarbons, typically boiling in the gasoline range. The catalyst may be a crystalline zeolitic aluminosilicate component, usually an ion-exchanged form of a synthetic crystalline Faujasite, and a porous inorganic oxide matrix. This type of catalyst must be regenerated to low carbon levels, typically 0.5% or less, to assure that the catalyst particles possess desired activity and selectivity before the particles are recycled to the conversion zone, also referred to as cracking zone. In most regenerators the combustible solids deposited on the spent solid catalyst particles from the cracking zone are burned in a confined regeneration zone in the form of a fluidized bed which has a relatively high concentration of catalyst particles (dense phase). A region of lower solids concentration (dilute phase) is maintained above the dense phase. High residual concentration of carbon monoxide in flue gases from regenerators has been a problem. The oxidation of carbon monoxide is highly exothermic and can result in so-called 'carbon monoxide afterburning' which can take place in the dilute catalyst phase, in the cyclones or in the flue gas lines. Afterburning has caused significant damage to plant equipment. CO to CO_2 combustion promoters are added to the FCC catalyst inventory of the regenerator with the objective to achieve a complete combustion of carbon monoxide in the regenerator dense phase and thereby avoiding this afterburning. The combustion promoters are separate particles from the cracking catalyst particles. The combustion promoters will be circulated

EXHIBIT A

Invalidity of U.S. Patent No. 11,224,864 in View of Applicant Admitted Prior Art ("*AAPA*")

'864 patent	Applicant Admitted Prior Art ("*AAPA*")
	with the rest of the FCC catalyst inventory from the regenerator to the riser, to the stripper and back to the regenerator. Such CO to CO_2 combustion promoters are commonly made from impregnation of Group VIII noble metal or metals, commonly platinum and/or palladium, onto a porous alumina and/or other substrate microspheres of average particle size 60 to 90 microns, with physical properties very similar to the base FCCU catalysts. Such impregnation results in a uniform distribution of the Group VIII noble metal throughout the internal and external surfaces of the porous microsphere. EP1879982 describes a CO to CO_2 combustion promoter for use in FCC containing platinum or palladium predominately present in the core of the particle and a metal active for catalysing NOx decomposition in a shell around the platinum or palladium. US2005/0042158 describes a CO to CO_2 combustion promoter for use in FCC containing cerium oxide and a noble metal, like platinum and/or rhodium. Other background publications are U.S. Pat. No. 6,165, 933, US2005042158, U.S. Pat. Nos. 4,812,431, 4,300,997. A disadvantage of all noble metal based CO to CO_2 combustion promoters is that relatively large amounts of noble metal are required to achieve the desired CO combustion in the FCC process. It is an objective of the present invention to provide a CO to CO_2 combustion promoter which requires less noble metal to achieve the same level of CO combustion in the FCC process." ['864 Patent, 3:14-22] "A further advantage with the eggshell of noble metal is that less NOx formation is achieved. This may be explained as follows. NOx forming reactions are slower and thus less diffusion limited than the CO to CO_2 reactions. Thus, the prior art

EXHIBIT A

Invalidity of U.S. Patent No. 11,224,864 in View of Applicant Admitted Prior Art ("*AAPA*")

'864 patent	Applicant Admitted Prior Art ("*AAPA*")
	combustion promoters having a uniform distribution and higher loading of the noble metal will generate more NOx as this additional noble metal will catalyse NOx forming reactions." [Prosecution History of '864 Patent, Page 27] "As an initial matter, in the remarks dated 07/07/2021 ('Remarks'), the Applicant noted that the terms 'microsphere' and 'eggshell' are not interchangeable. See Remarks at p8. It appears that the Applicant is distinguishing between the two terms in that 'microsphere' relates to the shape of the claimed particles while 'eggshell' relates to the distribution of the claimed 'one or more Group VIII noble metals' in the claimed microsphere. Along with other references, the Applicant points to pages 6688-99 of Munnik et al. ('Recent Developments in the Synthesis of Supported Catalysts') to support this distinction. See Remarks at p9. Munnik teaches that an eggshell distribution is a 'macroscopic distribution' that is resulted from the adsorption of a metal precursor in the outer shell of the support body.' See Munnik at section 4.2.2. The Office will adopt this interpretation of both terms." [Prosecution History of '864 Patent, Page 51] "Applicant submits that the term 'microsphere' is a term of art, the metes and bounds of which are readily understood by one of ordinary skill in the art. For example, Microspheres Online, a collection of technical information written by industry experts, defines microspheres as spherical particles 'as small as 1 micron in diameter and as large as 1,000 microns (1 mm).' *See* Exhibit 1 submitted herewith. This term is particularly well-established within the relevant field of FCC catalysts, appearing without the need for definition or elaboration in U.S. Patents even before WWII (See U.S. Patents 2,283,172; 2,933,456; and 3,243,262) and technical books from at least 40 years ago (See Fig. 13 of Venuto and Habib 'Fluid catalytic cracking with zeolite catalysts.' Marcel Dekker, Inc. New York (1979); Section VII of which is provided herewith as Exhibit 2)."

EXHIBIT A

Invalidity of U.S. Patent No. 11,224,864 in View of Applicant Admitted Prior Art ("*AAPA*")

'864 patent	**Applicant Admitted Prior Art ("*AAPA*")**
	[Prosecution History of '864 Patent, Page 65] "'Microsphere' is a well understood term within the field. 'Microsphere' is defined as spherical particles having a diameter of 1 micron to 1,000 microns (1mm). In my professional experience, this term is well-understood to have this meaning by those of ordinary skill in the art." [Prosecution History of '864 Patent, Page 65] "Porous particles generally are known for use in FCC processes and as combustion promoters." To the extent that this limitation is not disclosed, either explicitly or inherently, by *AAPA*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
1[b] silica, alumina, or mixtures thereof; and	*AAPA* discloses a combustion promoter comprising silica, alumina, or mixtures thereof. *See, e.g.*, ['864 Patent, 1:25-2:20] "Such CO to CO_2 combustion promoters are well known and are used in a fluid catalytic cracking (FCC) unit. In such a FCC process fluidizable catalyst particles are contacted in a conversion zone at high temperatures with hydrocarbons such to crack higher boiling hydrocarbons to lower boiling hydrocarbons, typically boiling in the gasoline range. The catalyst may be a crystalline zeolitic aluminosilicate component, usually an ion-exchanged form of a synthetic crystalline Faujasite, and a porous inorganic oxide matrix. This type of catalyst must be regenerated to low carbon levels, typically 0.5% or less, to assure that the catalyst particles possess

EXHIBIT A

Invalidity of U.S. Patent No. 11,224,864 in View of Applicant Admitted Prior Art (“*AAPA*”)

’864 patent	Applicant Admitted Prior Art (“*AAPA*”)
	desired activity and selectivity before the particles are recycled to the conversion zone, also referred to as cracking zone. In most regenerators the combustible solids deposited on the spent solid catalyst particles from the cracking zone are burned in a confined regeneration zone in the form of a fluidized bed which has a relatively high concentration of catalyst particles (dense phase). A region of lower solids concentration (dilute phase) is maintained above the dense phase. High residual concentration of carbon monoxide in flue gases from regenerators has been a problem. The oxidation of carbon monoxide is highly exothermic and can result in so-called ‘carbon monoxide afterburning’ which can take place in the dilute catalyst phase, in the cyclones or in the flue gas lines. Afterburning has caused significant damage to plant equipment. CO to CO_2 combustion promoters are added to the FCC catalyst inventory of the regenerator with the objective to achieve a complete combustion of carbon monoxide in the regenerator dense phase and thereby avoiding this afterburning. The combustion promoters are separate particles from the cracking catalyst particles. The combustion promoters will be circulated with the rest of the FCC catalyst inventory from the regenerator to the riser, to the stripper and back to the regenerator. Such CO to CO_2 combustion promoters are commonly made from impregnation of Group VIII noble metal or metals, commonly platinum and/or palladium, onto a porous alumina and/or other substrate microspheres of average particle size 60 to 90 microns, with physical properties very similar to the base FCCU catalysts. Such impregnation results in a uniform distribution of the Group VIII noble metal throughout the internal and external surfaces of the porous microsphere. EP1879982 describes a CO to CO_2 combustion promoter for use in FCC containing platinum or palladium predominately present in the core of the particle and a metal active for catalysing NOx decomposition in a shell around the platinum or palladium.

EXHIBIT A

Invalidity of U.S. Patent No. 11,224,864 in View of Applicant Admitted Prior Art ("*AAPA*")

'864 patent	**Applicant Admitted Prior Art ("*AAPA*")**
	US2005/0042158 describes a CO to CO_2 combustion promoter for use in FCC containing cerium oxide and a noble metal, like platinum and/or rhodium. Other background publications are U.S. Pat. No. 6,165, 933, US2005042158, U.S. Pat. Nos. 4,812,431, 4,300,997. A disadvantage of all noble metal based CO to CO_2 combustion promoters is that relatively large amounts of noble metal are required to achieve the desired CO combustion in the FCC process. It is an objective of the present invention to provide a CO to CO_2 combustion promoter which requires less noble metal to achieve the same level of CO combustion in the FCC process." To the extent that this limitation is not disclosed, either explicitly or inherently, by *AAPA*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
1[c] one or more Group VIII noble metals distributed in the particle as an eggshell such that a higher concentration of the one or more Group VIII noble metals is present in the outer region of the microsphere sized porous particle as compared to the concentration of the one or more Group VIII noble metals in the centre of the microsphere sized porous particle.	*AAPA* discloses a combustion promoter comprising one or more Group VIII noble metals, the metals distributed in the particle as an eggshell such that a higher concentration of the one or more Group VIII noble metals is present in the outer region of the microsphere sized porous particle as compared to the concentration of the one or more Group VIII noble metals in the centre of the microsphere sized porous particle. *See, e.g.*, ['864 Patent, 1:25-2:20] "Such CO to CO_2 combustion promoters are well known and are used in a fluid catalytic cracking (FCC) unit. In such a FCC process fluidizable catalyst particles

EXHIBIT A

Invalidity of U.S. Patent No. 11,224,864 in View of Applicant Admitted Prior Art ("*AAPA*")

'864 patent	Applicant Admitted Prior Art ("*AAPA*")
	are contacted in a conversion zone at high temperatures with hydrocarbons such to crack higher boiling hydrocarbons to lower boiling hydrocarbons, typically boiling in the gasoline range. The catalyst may be a crystalline zeolitic aluminosilicate component, usually an ion-exchanged form of a synthetic crystalline Faujasite, and a porous inorganic oxide matrix. This type of catalyst must be regenerated to low carbon levels, typically 0.5% or less, to assure that the catalyst particles possess desired activity and selectivity before the particles are recycled to the conversion zone, also referred to as cracking zone. In most regenerators the combustible solids deposited on the spent solid catalyst particles from the cracking zone are burned in a confined regeneration zone in the form of a fluidized bed which has a relatively high concentration of catalyst particles (dense phase). A region of lower solids concentration (dilute phase) is maintained above the dense phase. High residual concentration of carbon monoxide in flue gases from regenerators has been a problem. The oxidation of carbon monoxide is highly exothermic and can result in so-called 'carbon monoxide afterburning' which can take place in the dilute catalyst phase, in the cyclones or in the flue gas lines. Afterburning has caused significant damage to plant equipment. CO to CO_2 combustion promoters are added to the FCC catalyst inventory of the regenerator with the objective to achieve a complete combustion of carbon monoxide in the regenerator dense phase and thereby avoiding this afterburning. The combustion promoters are separate particles from the cracking catalyst particles. The combustion promoters will be circulated with the rest of the FCC catalyst inventory from the regenerator to the riser, to the stripper and back to the regenerator. Such CO to CO_2 combustion promoters are commonly made from impregnation of Group VIII noble metal or metals, commonly platinum and/or palladium, onto a porous alumina and/or other substrate microspheres of average particle size 60 to 90 microns, with physical properties very similar to the base FCCU catalysts. Such

EXHIBIT A

Invalidity of U.S. Patent No. 11,224,864 in View of Applicant Admitted Prior Art ("*AAPA*")

'864 patent	**Applicant Admitted Prior Art ("*AAPA*")**
	impregnation results in a uniform distribution of the Group VIII noble metal throughout the internal and external surfaces of the porous microsphere. EP1879982 describes a CO to CO_2 combustion promoter for use in FCC containing platinum or palladium predominately present in the core of the particle and a metal active for catalysing NOx decomposition in a shell around the platinum or palladium. US2005/0042158 describes a CO to CO_2 combustion promoter for use in FCC containing cerium oxide and a noble metal, like platinum and/or rhodium. Other background publications are U.S. Pat. No. 6,165, 933, US2005042158, U.S. Pat. Nos. 4,812,431, 4,300,997. A disadvantage of all noble metal based CO to CO_2 combustion promoters is that relatively large amounts of noble metal are required to achieve the desired CO combustion in the FCC process. It is an objective of the present invention to provide a CO to CO_2 combustion promoter which requires less noble metal to achieve the same level of CO combustion in the FCC process." ['864 Patent, 3:14-22] "A further advantage with the eggshell of noble metal is that less NOx formation is achieved. This may be explained as follows. NOx forming reactions are slower and thus less diffusion limited than the CO to CO_2 reactions. Thus, the prior art combustion promoters having a uniform distribution and higher loading of the noble metal will generate more NOx as this additional noble metal will catalyse NOx forming reactions." [Prosecution History of '864 Patent, Page 27] "As an initial matter, in the remarks dated 07/07/2021 ('Remarks'), the Applicant noted that the terms 'microsphere' and 'eggshell' are not interchangeable. See

EXHIBIT A

Invalidity of U.S. Patent No. 11,224,864 in View of Applicant Admitted Prior Art (“*AAPA*”)

’864 patent	Applicant Admitted Prior Art (“*AAPA*”)
	Remarks at p8. It appears that the Applicant is distinguishing between the two terms in that ‘microsphere’ relates to the shape of the claimed particles while ‘eggshell’ relates to the distribution of the claimed ‘one or more Group VIII noble metals’ in the claimed microsphere. Along with other references, the Applicant points to pages 6688-99 of Munnik et al. (‘Recent Developments in the Synthesis of Supported Catalysts’) to support this distinction. See Remarks at p9. Munnik teaches that an eggshell distribution is a ‘macroscopic distribution’ that is resulted from the adsorption of a metal precursor ‘in the outer shell of the support body.’ See Munnik at section 4.2.2. The Office will adopt this interpretation of both terms.” [Prosecution history of ’864 Patent, Page 53] “To the contrary, Applicant submits that ‘microspheroidal’ and ‘eggshell’ are terms of art which are in no way interchangeable. As demonstrated above, ‘microspheroidal’ refers to spherical particles having a diameter of 1 micron to 1,000 microns (1 mm). In contrast, ‘eggshell,’ at least within the fields of nanoparticles and in particular catalyst particles, refers to a structure with a thin layer or ‘shell’ surrounding an inner layer that is chemically distinct from the outer layer. In particular, an ‘eggshell’ particle that comprises a catalyst comprises higher levels of catalyst in the outer layer. In this particular instance, the use of ‘eggshell’ of instant claim 1 would be understood by one of ordinary skill in the art to indicate that the particle has an outer layer (e.g., a shell) with a relatively high concentration of the one or more Group VIII noble metals and an inner layer (e.g., a core) with a relatively lower concentration of the one or more Group VIII noble metals.” [Prosecution history of ’864 Patent, Page 54] “The foregoing is substantiated by the accompanying Exhibits 3-5, which demonstrate that ‘eggshell’ is a term of art with the meaning articulated above. In particular, please see Exhibit 3 page 1, left column, third paragraph; Exhibit 3 Fig. 5; Exhibit 4 pages 1-2 including Fig. l; and Exhibit 5 pages 6688-6699 including Fig. 1 Additionally, the accompanying Declaration of Dr. Herring further

EXHIBIT A

Invalidity of U.S. Patent No. 11,224,864 in View of Applicant Admitted Prior Art ("*AAPA*")

'864 patent	**Applicant Admitted Prior Art ("*AAPA*")**
	substantiates that these two terms are well understood by one of ordinary skill in the art and are not synonymous. *See*, paragraphs 6 and 7 of the Declaration of Dr. Herring." [Prosecution History of '864 Patent, Page 65] "'Eggshell' and 'microsphere' are not synonymous. As detailed in the accompanying Exhibits 3-5, 'eggshell' is defined as a particle structure having chemically distinct inner and outer layers, and when a catalyst is present, having a higher concentration of the catalyst in the outer layer. In my professional experience, this term is well-understood to have this meaning by those of ordinary skill in the art. While a single particle, such as those described in the '222 application could certainly be both a microsphere and an eggshell particle, these two terms describe different physical aspects of a particle and are in no way interchangeable or equivalent." To the extent that this limitation is not disclosed, either explicitly or inherently, by *AAPA*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[3] The combustion promoter according to claim 1, comprising a plurality of microsphere sized porous particles comprising alumina.	*AAPA* discloses a combustion promoter comprising a plurality of microsphere sized porous particles comprising alumina. *See, e.g.*, The support provided above for Claim 1[b]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *AAPA*, this limitation is obvious in combination with the knowledge of one of skill

EXHIBIT A

Invalidity of U.S. Patent No. 11,224,864 in View of Applicant Admitted Prior Art ("*AAPA*")

'864 patent	**Applicant Admitted Prior Art ("*AAPA*")**
	in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[6] The combustion promoter according to claim 1, wherein the microsphere sized porous particles collectively have an average (D50) size of between 60 and 90 microns as measured by laser diffraction.	*AAPA* discloses a combustion promoter comprising microsphere sized porous particles, wherein the microsphere sized porous particles collectively have an average (D50) size of between 60 and 90 microns as measured by laser diffraction. *See, e.g.*, The support provided above for Claim 1[a]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *AAPA*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[12] The combustion promoter according to claim 1, wherein the one or more Group VIII noble metals comprises one or more of platinum, palladium, iridium, ruthenium, and rhodium.	*AAPA* discloses a combustion promoter comprising one or more Group VIII noble metals, wherein the one or more Group VIII noble metals comprises one or more of platinum, palladium, iridium, ruthenium, and rhodium. *See, e.g.*, The support provided above for Claim 1[c]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *AAPA*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of

EXHIBIT A

Invalidity of U.S. Patent No. 11,224,864 in View of Applicant Admitted Prior Art ("*AAPA*")

'864 patent	**Applicant Admitted Prior Art ("*AAPA*")**
	the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[15] The combustion promoter according to claim 12, wherein the one or more Group VIII noble metals comprises palladium.	*AAPA* discloses a combustion promoter comprising one or more Group VIII noble metals, wherein the one or more Group VIII noble metals comprises palladium. *See, e.g.*, The support provided above for Claim 1[c]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *AAPA*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.

EXHIBIT B

EXHIBIT B

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,053,024 ("*Baker*")

U.S. Patent No. 7,053,024 ("*Baker*"), filed on January 9, 2003 and published on May 30, 2006, qualifies as prior art under at least 35 U.S.C. §§ 102 and/or 103 for the asserted claims of U.S. Patent No. 11,224,864 ("the '864 patent"), as discussed in detail in the claim chart below.

The Court has not yet construed the claims and therefore the meaning of the terms in the claims has yet to be resolved by the Court. The support identified here for these limitations is responsive to Plaintiffs' infringement contentions, which Defendant disagrees with. As such, nothing in Defendant's claim charts should be construed as an admission regarding infringement, either literally or under the doctrine of equivalents, or as an admission regarding Defendant's understanding of the proper scope of the asserted claims. Defendant reserves the right to rely on additional citations or sources of evidence that also may be applicable, or that may become applicable in light of claim constructions, changes in Plaintiffs' infringement contentions, and/or information obtained during discovery as the case progresses.

To the extent Plaintiffs allege that *Baker* does not disclose any particular limitation of the asserted claims of the '864 patent, either expressly or inherently, it would have been obvious to a person of ordinary skill in the art as of the priority date of the '864 patent to modify *Baker* and/or to combine the teachings of *Baker* with other prior art references, including but not limited to the present prior art references cited in the Cover Pleading and the relevant section(s) of claim charts for other prior art references for the '864 patent in a manner that would have rendered the asserted claims invalid as obvious.

Defendant reserves the right to amend or supplement this claim chart at a later date as more fully set forth in the Cover Pleading.

EXHIBIT B

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,053,024 ("*Baker*")

'864 patent	**U.S. Patent No. 7,053,024 ("*Baker*")**
1[a] A CO to CO_2 combustion promoter comprising microsphere sized porous particles, each microsphere sized porous particle having a diameter of less than 1mm, and independently comprising	*Baker* discloses a CO to CO_2 combustion promoter comprising microsphere sized porous particles, each microsphere sized porous particle having a diameter of less than 1mm, and independently comprising the limitations discussed in the following sections. *See, e.g.*, [Abstract] "Process for preparing a supported metal catalyst composition comprising impregnating support particles having a mean diameter of greater than 300 microns with a solution of least one catalytically active metal, or precursor thereof, such that the metal, or its precursor, is in a mobile state in the support particles; and drying the impregnated support particles. The mobile metal, or its precursor, in the support particles is then treated with a liquid comprising at least one reducing agent to deposit and immobilize the metal, or its precursor, in the support particles such that the metal, or its precursor, is distributed in the support particle in a layer below the surface of the support particle, the layer being between an inner and an outer region, each of the inner and outer regions having a lower concentration of the metal or precursor than the layer."

EXHIBIT B

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,053,024 ("*Baker*")

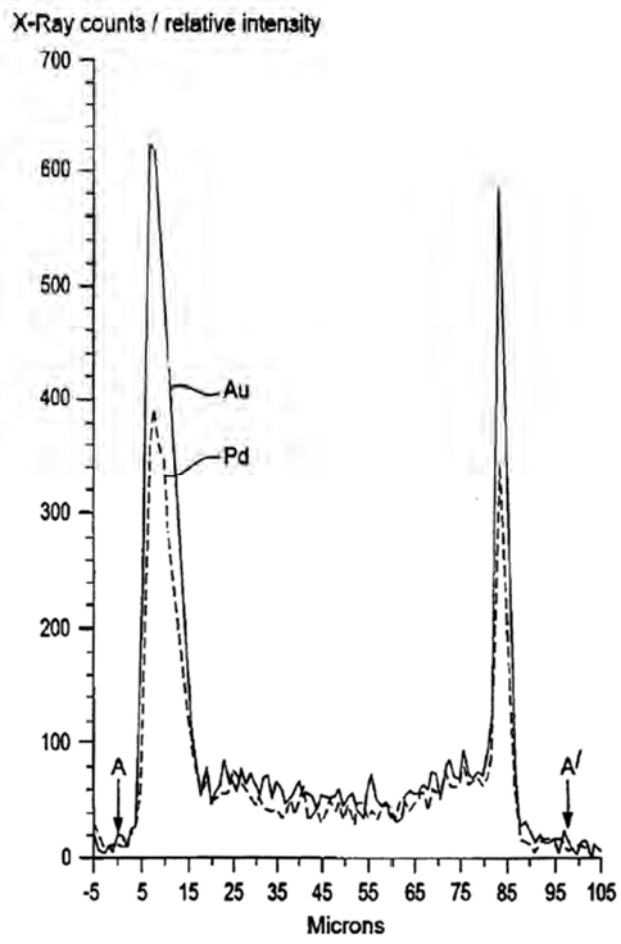
'864 patent	**U.S. Patent No. 7,053,024 ("*Baker*")**
	[Fig. 2] 

EXHIBIT B

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,053,024 ("*Baker*")

'864 patent	U.S. Patent No. 7,053,024 ("*Baker*")
	[1: 46-55] "U.S. Pat. No. 4,677,084 describes a process for preparing attrition resistant catalyst, catalyst precursor and catalyst support particles and in particular silica-containing vanadium/phosphorus oxide catalysts. The catalyst, catalyst precursor or catalyst support particles are slurried in a solution of an oxide such as silica. The slurry is then spray-dried and calcined to produce microspheres. The process results in the formation of an oxide-rich layer at the periphery of each calcined microsphere." [1:60-67] "Thus, according to the present invention there is provided a catalyst composition comprising support particles having at least one catalytically active metal distributed therein, in which the metal is distributed in the support particle in a layer below the surface of said particle, said layer being between an inner and an outer region of said support particle, and each of said inner and outer regions having a lower concentration of said metal than said layer." [2:41-58] "In a preferred embodiment, the layer containing the catalytically active metal, or its precursor, has an outer edge which is at least 3% and no more than 75% of the particle radius from the surface of the support particle and preferably, at least 5%, and more preferably at least 10% of the particle radius from the surface of the support particle. Depending upon the size of the support particles, alternatively or additionally, the layer containing the catalytically active metal, or its precursor, preferably has an outer edge which is at least 3 microns and no more than 20 microns below the surface of each support particle, and is more preferably 4 to 20 microns, below the surface of each particle, and yet more preferably is 5 to 15 microns below the surface of each particle.

EXHIBIT B

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,053,024 ("*Baker*")

'864 patent	U.S. Patent No. 7,053,024 ("*Baker*")
	Typically, the layer has an average thickness which is less than half the radius of the particle, for example less than 25 microns. Preferably, the layer has an average thickness of greater than 0.1 microns." [2:63-3:57] "A suitable support material for use in a fluid bed process is a microspheroidal particulate material. When the catalyst composition is to be used in a fluid bed process, as is well known in the fluid bed art, the support particles must be small enough to be maintained in a fluid bed state under reaction conditions while keeping sufficient attrition resistance such that excessive amounts of catalyst composition need not be replenished during the process. Further, although typical particle sizes (as measured by mean particle diameters) should not be so large as to be difficult to keep in 5 a fluid bed state, there should not be an excessive amount of very small particles (fines) which are difficult to remove from the system and may plug gas recycle lines. Thus, typically suitable fluid bed support particles have a distribution of larger to smaller particle sizes. For example, in the fluid bed manufacture of vinyl acetate from ethylene, acetic acid and oxygen-containing gas, typically, at least 80% and preferably at least 90% of the support particles have mean diameters of less than about 300 microns. A typical catalyst useful in this invention may have the following particle size distribution. 0 to 20 microns 0–30 wt % 20 to 44 microns 0–60 wt % 44 to 88 microns 10–80 wt % 88 to 106 microns 0–80 wt % >106 microns 0–40 wt % >300 microns 0–5 wt %

EXHIBIT B

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,053,024 ("*Baker*")

'864 patent	U.S. Patent No. 7,053,024 ("*Baker*")
	Persons skilled in the art will recognize that support particles sizes of 44, 88, and 300 microns are arbitrary measures in that they are based on standard sieve sizes. Particle sizes and particle size distributions may be measured by an automated laser device such as a Microtrac X100. Microspheroidal support particles useful in the present invention are sufficiently porous to permit gaseous reactants to diffuse into the particle and contact catalytic sites incorporated within the particle. Thus, the pore volume should be high enough to permit gaseous diffusion. However, a support particle with an exceedingly high pore volume typically will not have sufficient attrition resistance or will not have sufficient surface area for catalytic activity. A typically suitable microspheroidal support particle has a pore volume (measured by nitrogen sorption) between about 0.2 and 0.7 cc/g. A preferable support particle has a pore volume between about 0.3 and 0.65 cc/g and more preferably between about 0.4 and 0.55 cc/g. Surface areas (measured by nitrogen BET) for fluid bed support particles with mean diameters and pore volumes useful in the present invention typically are above about 50 m^2/g and may range up to about 200 m^2/g. A typical measured surface area is about 60 to about 125 m^2/g. Typically useful support particles, especially silica support particles are described in U.S. Pat. No. 5,591,688, incorporated by reference herein. In these supports microspheroidal particles are produced by spray drying a mixture of a silica sol with silica particles followed by drying and calcining." [4:8-21] "An advantageous silica sol for preparing a catalyst support useful in the present invention contains silica particles in the sol typically more than 20 nanometers in mean diameter and may be up to about 100 nanometers or more Preferable sols contain silica particles of about 40 to 80 nanometers. Nalco silica sol 1060

EXHIBIT B

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,053,024 ("*Baker*")

'864 patent	U.S. Patent No. 7,053,024 ("*Baker*")
	particularly is advantageous because of the relatively large mean silica particle sizes of 60 nm pack less efficiently than smaller sol particles such as Nalco 2327 at about 20 nm. The larger particle size sol yields a final support with higher mesopore volume and less micropore volume. A suitable support material for use in a fixed bed vinyl acetate process may be spherical. The support particles typically may have a diameter of 3 to 9mm. Surface areas (measured by nitrogen BET) for fixed bed support particles with mean diameters and pore volumes useful in this invention typically are above about 5 m^2/g and may range up to about 800 m^2/g. Suitably, a fixed bed support particle has a pore volume (measured by nitrogen sorption) between about 0.2 and 3.5 ml/gram. Suitably, fixed bed support particles have an apparent bulk density of 0.3 to 1.5 gram/ml. Although silica-based support particles are the most preferred in this invention for use in fluid or fixed bed processes, other oxides may be used as long as a particle of appropriate size and with sufficient pore volume is produced in which may be deposited the required catalytic materials. Possible other oxides include alumina, silica-alumina, ceria, magnesia, titania, zirconia and mixed oxides and mixtures thereof. The support may be impregnated with organic or inorganic bases for example Group I or Group II hydroxides and ammonium hydroxide." [4:48-65] "The catalytically active metal(s) may be impregnated in one or more steps onto the support particles in the form of precursor salt solutions. In a preferred aspect of the present invention, microspheroidal support particles are preferably impregnated with a palladium compound in a suitable solvent. Suitable solvents may be water, carboxylic acids such as acetic acid, benzene, toluene, alcohols such as methanol or ethanol, nitriles such as acetonitrile or benzonitrile, tetrahydrofuran or chlorinated solvents such as dichloromethane. Preferably, the solvent is water and/or acetic acid.

EXHIBIT B

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,053,024 ("*Baker*")

'864 patent	U.S. Patent No. 7,053,024 ("*Baker*")
	Suitably, the support particles are impregnated with palladium acetate, sulphate, nitrate, chloride or halogen-containing palladium compounds such as H_2PdC1_4, which is sometimes also represented as $[PdCl_2]2HCl$, and Group I or Group II salts thereof such as Na_2PdCl_4 and K_2 $PdCl_4$. A preferred water soluble compound is Na_2PdCl_4. A preferred acetic acid-soluble palladium compound is palladium acetate." [4:66-5:12] "The catalyst composition suitable for the manufacture of vinyl acetate may also comprise, as promoters, other metals such as gold, copper, cerium and mixtures thereof preferably gold. These other metals may also be more concentrated in the layer than in the inner and outer regions, that is the inner and outer regions may have a lower concentration of said promoter metal than said layer. Typically, the weight percent of gold is at least about 0.1 wt %, preferably, at least 0.2 wt % gold to about 3 wt % and preferably up to 1 wt % gold. Typically, the weight percent of cerium is at least about 0.1 wt%, preferably at least 0.2 wt% to about 10 wt% or more, preferably up to 5 wt % of cerium. Typically, the weight percent of copper is at least 0.1 to about 10 wt%, preferably 10 up to 5 wt % copper." [7:35-53] "In FIG. 1, a catalyst particle (I) is provided with a layer (2) of a Group VIII noble metal such as palladium. The layer (2) may optionally contain other metals such as gold. The layer (2) is located with an outer edge (7) below the particle surface (3) and between an inner region (4) and an outer region (5) of the catalyst particle having lower concentrations of the Group VIII noble metal and/or other metals than the layer (2). FIG. 2 illustrates an X-ray profile from beyond and across a section of a catalyst particle containing palladium and gold using an Electron Microprobe Analyser, for example along line X- X' of FIG. 1. FIG. 2 clearly illustrates that the palladium and

'864 patent	U.S. Patent No. 7,053,024 ("*Baker*")
	gold are both mainly distributed in two specific locations along the diameter of the particle (that is, as a layer below the surface of the particle) with lower concentrations of palladium and gold distributed elsewhere. In FIG. 2 the outer edges of the particle are at positions labeled A and A' at 0 and 97 microns." [7:59-8:10] "Preformed microspheroidal support particles comprising 100% silica were used for all the Examples described below. The support particles were prepared by spray-drying a mixture of Nalco silica sol 1060 (Nalco Chemical Company) and Degussa Aerosil® silica (Degussa Chemical Company). In the dried support particles, 80% of the silica came from the sol and 20% of the silica came from the Aerosil. The spray-dried microspheroidal support particles were calcined in air at 640° C. for 4 hours. Prior to use the support particles were sieved and a specific particle size distribution was used in the preparation of the catalyst compositions as follows: Particle size % >300 microns 2 88–300 microns 30 44–88 microns 38 <44 microns 30 " To the extent that this limitation is not disclosed, either explicitly or inherently, by *Baker*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of

EXHIBIT B

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,053,024 ("*Baker*")

'864 patent	U.S. Patent No. 7,053,024 ("*Baker*")
	the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
1[b] silica, alumina, or mixtures thereof; and	*Baker* discloses a combustion promoter comprising silica, alumina, or mixtures thereof. *See, e.g.*, [1:12-54] "Supported metal catalysts are typically made by impregnating a suitable support material with a catalytically active metal or with its precursor. For example, catalysts for use in the production vinyl acetate monomer (VAM) by the reaction of ethylene, acetic acid and oxygen are made by impregnating a support such as silica or alumina with a compound of a Group VIII noble metal such as palladium together with a gold compound and an alkali metal salt, typically in the form of an acetate, the palladium and gold compounds being converted to catalytically active state. In early examples of fixed-bed catalysts for use in the production of VAM, palladium and gold were distributed more or less uniformly throughout the support, for example, U.S. Pat. No. 3,743,607. Since gaseous reactants do not diffuse significantly into the large fixed-bed catalyst particles, much of the expensive catalytic metal components in the interior of the catalyst were not useful. Subsequently, shell-impregnated, fixed-bed catalysts were developed in which most of the catalytic metals were deposited onto an outer shell of the support particle. For example, Great Britain Patent No. 1,500,167 describes a catalyst in which at least ninety percent of the palladium and gold is distributed in that part of the support particle which is not more than thirty percent of the particle radius from the surface. The palladium and gold being at/near the surface are susceptible to loss through attrition.

EXHIBIT B

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,053,024 ("*Baker*")

'864 patent	U.S. Patent No. 7,053,024 ("*Baker*")
	In the preparation of shell-impregnated, fixed bed catalysts such as that described in GB 1,500,167 and EP-A-0 569624, after impregnation of a support with a Group VIII noble metal solution, the noble metal is subsequently precipitated to the support by, for example, treatment with an aqueous solution of an alkali metal salt Such precipitated noble metal has limited mobility. U.S. Pat. No. 4,677,084 describes a process for preparing attrition resistant catalyst, catalyst precursor and catalyst support particles and in particular silica-containing vanadium/phosphorus oxide catalysts. The catalyst, catalyst precursor or catalyst support particles are slurried in a solution of an oxide such as silica. The slurry is then spray-dried and calcined to produce microspheres. The process results in the formation of an oxide-rich layer at the periphery of each calcined microsphere." [1:60-67] "Thus, according to the present invention there is provided a catalyst composition comprising support particles having at least one catalytically active metal distributed therein, in which the metal is distributed in the support particle in a layer below the surface of said particle, said layer being between an inner and an outer region of said support particle, and each of said inner and outer regions having a lower concentration of said metal than said layer." [3:52- 4:18] "Typically useful support particles, especially silica support particles are described in U.S. Pat. No. 5,591,688, incorporated by reference herein. In these supports microspheroidal particles are produced by spray drying a mixture of a silica sol with silica particles followed by drying and calcining. In the preparation, at least 10 wt. %, preferably at least 50 wt. %, of a silica sol is mixed with particulate silica. A useful particulate silica is a fumed silica such as Aerosil® (Degussa Chemical Company). A typical silica particulate material has a high surface area (about 200 m^2/g) with essentially no micropores, and, typically, are aggregates (with mean

EXHIBIT B

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,053,024 ("*Baker*")

'864 patent	U.S. Patent No. 7,053,024 ("*Baker*")
	diameters of several hundred nm) of individual particles with average diameters of about 10 nm (above 7 nm). Preferably, the silica is sodium free. Sufficient particulate silica is added to the mixture to obtain a desired pore volume in the resulting support particle. The amount of particulate silica may range up to 90 wt. % and typically ranges up to 10 to 50 wt. % of the silica in the mixture. Typically, the silica sol/particulate silica mixture is spray dried at an elevated temperature such as between 115° to 280° C., preferably 130° to 240° C., followed by calcining at temperature typically ranging from between 550° to 700° and, preferably 630° to 660° C. An advantageous silica sol for preparing a catalyst support useful in the present invention contains silica particles in the sol typically more than 20 nanometers in mean diameter and may be up to about 100 nanometers or more Preferable sols contain silica particles of about 40 to 80 nanometers. Nalco silica sol 1060 particularly is advantageous because of the relatively large mean silica particle sizes of 60 nm pack less efficiently than smaller sol particles such as Nalco 2327 at about 20 nm. The larger particle size sol yields a final support with higher mesopore volume and less micropore volume." [4:29-38] "Although silica-based support particles are the most preferred in this invention for use in fluid or fixed bed processes, other oxides may be used as long as a particle of appropriate size and with sufficient pore volume is produced in which may be deposited the required catalytic materials. Possible other oxides include alumina, silica-alumina, ceria, magnesia, titania, zirconia and mixed oxides and mixtures thereof. The support may be impregnated with organic or inorganic bases for example Group I or Group II hydroxides and ammonium hydroxide."

EXHIBIT B

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,053,024 ("*Baker*")

'864 patent	**U.S. Patent No. 7,053,024 ("*Baker*")**
	[7:59-60] "Preformed microspheroidal support particles comprising 100% silica were used for all the Examples described below." [8:19-23] "Silica support particles (163 g) were impregnated with a solution of $Na_2PdC1_4.xH_2O$ (containing 2.9 g palladium) and $HAuC1_4.xH_2O$ (containing 1.2 g gold) in distilled water by incipient wetness. The resulting mixture was mixed thoroughly, left to stand for 1 hour and dried overnight." To the extent that this limitation is not disclosed, either explicitly or inherently, by *Baker*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
1[c] one or more Group VIII noble metals distributed in the particle as an eggshell such that a higher concentration of the one or more Group VIII noble metals is present in the outer region of the microsphere sized porous particle as compared to the concentration of the one or more Group VIII noble metals in the centre of the microsphere sized porous particle.	*Baker* discloses a combustion promoter comprising one or more Group VIII noble metals, the metals distributed in the particle as an eggshell such that a higher concentration of the one or more Group VIII noble metals is present in the outer region of the microsphere sized porous particle as compared to the concentration of the one or more Group VIII noble metals in the centre of the microsphere sized porous particle. *See, e.g.*, [Abstract] "Process for preparing a supported metal catalyst composition comprising impregnating support particles having a mean diameter of greater than 300 microns with a solution of least one catalytically active metal, or precursor thereof, such that the metal, or its precursor, is in a mobile state in the support particles; and drying the

EXHIBIT B

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,053,024 ("*Baker*")

'864 patent	U.S. Patent No. 7,053,024 ("*Baker*")
	impregnated support particles. The mobile metal, or its precursor, in the support particles is then treated with a liquid comprising at least one reducing agent to deposit and immobilize the metal, or its precursor, in the support particles such that the metal, or its precursor, is distributed in the support particle in a layer below the surface of the support particle, the layer being between an inner and an outer region, each of the inner and outer regions having a lower concentration of the metal or precursor than the layer." [1:12-54] "Supported metal catalysts are typically made by impregnating a suitable support material with a catalytically active metal or with its precursor. For example, catalysts for use in the production vinyl acetate monomer (VAM) by the reaction of ethylene, acetic acid and oxygen are made by impregnating a support such as silica or alumina with a compound of a Group VIII noble metal such as palladium together with a gold compound and an alkali metal salt, typically in the form of an acetate, the palladium and gold compounds being converted to catalytically active state. In early examples of fixed-bed catalysts for use in the production of VAM, palladium and gold were distributed more or less uniformly throughout the support, for example, U.S. Pat. No. 3,743,607. Since gaseous reactants do not diffuse significantly into the large fixed-bed catalyst particles, much of the expensive catalytic metal components in the interior of the catalyst were not useful. Subsequently, shell-impregnated, fixed-bed catalysts were developed in which most of the catalytic metals were deposited onto an outer shell of the support particle. For example, Great Britain Patent No. 1,500,167 describes a catalyst in which at least ninety percent of the palladium and gold is distributed in that part of the support particle which is not more than thirty percent of the particle radius from the surface. The palladium and gold being at/near the surface are susceptible to loss through attrition.

EXHIBIT B

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,053,024 ("*Baker*")

'864 patent	U.S. Patent No. 7,053,024 ("*Baker*")
	In the preparation of shell-impregnated, fixed bed catalysts such as that described in GB 1,500,167 and EP-A-0 569624, after impregnation of a support with a Group VIII noble metal solution, the noble metal is subsequently precipitated to the support by, for example, treatment with an aqueous solution of an alkali metal salt Such precipitated noble metal has limited mobility. U.S. Pat. No. 4,677,084 describes a process for preparing attrition resistant catalyst, catalyst precursor and catalyst support particles and in particular silica-containing vanadium/phosphorus oxide catalysts. The catalyst, catalyst precursor or catalyst support particles are slurried in a solution of an oxide such as silica. The slurry is then spray-dried and calcined to produce microspheres. The process results in the formation of an oxide-rich layer at the periphery of each calcined microsphere." [1:60-67] "Thus, according to the present invention there is provided a catalyst composition comprising support particles having at least one catalytically active metal distributed therein, in which the metal is distributed in the support particle in a layer below the surface of said particle, said layer being between an inner and an outer region of said support particle, and each of said inner and outer regions having a lower concentration of said metal than said layer." [2:5-58] "The present invention also provides a process for preparing a supported metal catalyst composition which process comprises impregnating support particles with a solution of least one catalytically active metal, or precursor thereof, such that the metal, or its precursor, is in a mobile state in the support particles and then treating the mobile metal, or precursor, in the support particles with at least one chemical reagent to deposit and immobilize the metal, or its precursor, in the support particles such that the metal, or its precursor, is distributed in the support particle in a layer below the surface of said support particle, said layer being between an inner and an

EXHIBIT B

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,053,024 ("*Baker*")

'864 patent	U.S. Patent No. 7,053,024 ("*Baker*")
	outer region, each of said inner and outer regions having a lower concentration of said metal or precursor than said layer. Also, according to the present invention there is provided a composition comprising support particles having at least one precursor of a catalytically active metal distributed therein, in which the precursor is distributed in the support particle in a layer below the surface of said particle, said layer being between an inner and an outer region of said support particle, and each of said inner and outer regions having a lower concentration of said precursor than said layer. An advantage of the process of the present invention is that by treating a catalytically active metal, or its precursor, which is in a mobile state in the support particle with at least one chemical reagent which deposits and immobilizes it, the metal, or its precursor, is distributed predominantly in a layer below the surface of the particle such that the catalyst composition so produced has high attrition resistance as well as high activity. Preferably, the concentration of catalytically active metal or of its precursor in each of the inner and outer regions is less than half the concentration of the catalytically active metal or of its precursor in the layer. In a preferred embodiment, the layer containing the catalytically active metal, or its precursor, has an outer edge which is at least 3% and no more than 75% of the particle radius from the surface of the support particle and preferably, at least 5%, and more preferably at least 10% of the particle radius from the surface of the support particle. Depending upon the size of the support particles, alternatively or additionally, the layer containing the catalytically active metal, or its precursor, preferably has an outer edge which is at least 3 microns and no more than 20 microns below the

EXHIBIT B

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,053,024 ("*Baker*")

'864 patent	U.S. Patent No. 7,053,024 ("*Baker*")
	surface of each support particle, and is more preferably 4 to 20 microns, below the surface of each particle, and yet more preferably is 5 to 15 microns below the surface of each particle. Typically, the layer has an average thickness which is less than half the radius of the particle, for example less than 25 microns. Preferably, the layer has an average thickness of greater than 0.1 microns." [4:39-47] "Preferably, the catalytically active metal comprises at least one Group VIII noble metal. The noble metals of Group VIII of the Periodic Table of the Elements (IUPAC) are palladium, platinum, rhodium, ruthenium, osmium and iridium. Typically, the noble metal used in a catalyst composition for the manufacture of vinyl acetate comprises palladium. Such a catalyst composition typically contains at least about 0.1%, preferably at least 0.2 wt % palladium to about 5 wt % and preferably up to 4 wt % palladium. The catalytically active metal(s) may be impregnated in one or more steps onto the support particles in the form of precursor salt solutions. In a preferred aspect of the present invention, microspheroidal support particles are preferably impregnated with a palladium compound in a suitable solvent. Suitable solvents may be water, carboxylic acids such as acetic acid, benzene, toluene, alcohols such as methanol or ethanol, nitriles such as acetonitrile or benzonitrile, tetrahydrofuran or chlorinated solvents such as dichloromethane. Preferably, the solvent is water and/or acetic acid. Suitably, the support particles are impregnated with palladium acetate, sulphate, nitrate, chloride or halogen-containing palladium compounds such as H_2PdC1_4, which is sometimes also represented as $[PdCl_2]2HCl$, and Group I or Group II salts thereof such as $Na_2 PdCl_4$ and K_2PdCl_4. A preferred water soluble compound is Na_2PdCl_4. A preferred acetic acid-soluble palladium compound is palladium acetate."

EXHIBIT B

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,053,024 ("*Baker*")

'864 patent	U.S. Patent No. 7,053,024 ("*Baker*")
	[4:66-5:12] "The catalyst composition suitable for the manufacture of vinyl acetate may also comprise, as promoters, other metals such as gold, copper, cerium and mixtures thereof preferably gold. These other metals may also be more concentrated in the layer than in the inner and outer regions, that is the inner and outer regions may have a lower concentration of said promoter metal than said layer. Typically, the weight percent of gold is at least about 0.1 wt %, preferably, at least 0.2 wt % gold to about 3 wt % and preferably up to 1 wt % gold. Typically, the weight percent of cerium is at least about 0.1 wt%, preferably at least 0.2 wt% to about 10 wt% or more, preferably up to 5 wt % of cerium. Typically, the weight percent of copper is at least 0.1 to about 10 wt%, preferably 10 up to 5 wt % copper." [5:12-54] "Impregnation of the support particles with the gold, copper, cerium or mixtures thereof may be carried out together with or separately from the impregnation of the support particles with the Group VII noble metal compounds such as palladium compound(s). Suitable gold compounds include gold chloride, dimethyl gold acetate, barium acetoaurate, gold acetate, tetrachloroauric acid ($HAuCl_4$, sometimes represented as $AuCl_3HCl$) and Group I and Group II salts of tetrachloroauric acid such as $NaAuCl_4$ and $KAuCl_4$. Preferably, the gold compound is $HAuCl_4$. The gold compounds may be prepared in situ from suitable reagents. These promoters may be used in an amount of 0.1 to 10% by weight of each promoter metal present in the finished catalyst composition. In catalyst compositions suitable for the production of vinyl acetate, in addition to Group VIII noble metals such as palladium and optional promoter selected from gold, copper and cerium, the support particles may also be impregnated at any suitable stage during the preparation process with one or more salts of Group I, Group II, lanthanide and transition metals promoters, preferably of cadmium,

EXHIBIT B

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,053,024 ("*Baker*")

'864 patent	U.S. Patent No. 7,053,024 ("*Baker*")
	barium, potassium, sodium, manganese, antimony, lanthanum or matures thereof, which are present in the finished catalyst composition as salts, typically acetates. Generally, potassium will be present. Suitable salts of these compounds are acetates but any soluble salt may be used. These promoters may be used in an amount of 0.1 to 15%, preferably 3 to 9%, by weight of each promoter salt present in the finished catalyst composition. The impregnation of the support particles may be performed using any suitable technique. A preferable method to impregnate salt solutions is an incipient wetness technique in which there is used a salt solution in an amount up to the volume of the pores of the support particles without excess solution being used. Thus, a desired level of metal compounds such as palladium and other metal species may be incorporated into the support particles by calculating the amount of metals and the volume of solution needed. The impregnation is typically performed at ambient temperature. Elevated temperatures may be used for example, with palladium acetate in acetic acid, greater than 60° C. and up to 120° C." [6:31-44] "It has been found that the amount of chemical reagent needed to give a layered structure depends on the amount of metal e.g. Group VIII noble metal such as palladium metal which is present in the catalyst composition. Generally, higher concentrations of chemical reagent than have hitherto been used are used. Thus, for example, if palladium is present in the catalyst composition in amounts of 0.5- 2 wt %, a hydrazine concentration in water in excess of 2 wt %, for example, at least 3 wt %, such as 4- 20 wt % and preferably 4- 8 wt % has been found to produce a layered structure of the present invention. It has also been found that the more concentrated the solution of hydrazine the greater the distance the layer will be below the surface of the particle."

EXHIBIT B

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,053,024 ("*Baker*")

'864 patent	U.S. Patent No. 7,053,024 ("*Baker*")
	[6:51-7:7] "Preferably hydrazine at a concentration in water of at least 2 wt%, preferably in excess of 2 wt %, for example at least 4 wt%, is used in the preparation of the catalyst compositions. Thus, according to another embodiment of the present invention there is provided a process for preparing a catalyst composition wherein said process comprises impregnating support particles with a solution of least one Group VIII noble metal and then contacting the impregnated support with hydrazine at a concentration in water of at least 2 wt%. Contacting the chemical reagent with the mobile metal- or precursor-impregnated support particles deposits and immobilizes the metal or its precursor such that the metal or its precursor is distributed as a layer below the surface of each support particle. Preferably, at least 50% of the metal is distributed as a layer in each support particle. The distribution of the metal may be determined by suitable techniques such as Electron Microscopy. Base may be used to influence the mobility of the metal or its precursor and to affect the size and location of the layer. The base may be added before or during the impregnation of the support with the metal or its precursor. There should not be used so much base as to completely immobilize the metal or its precursor before addition of the chemical reagent." [7:35-53] "In FIG. 1, a catalyst particle (I) is provided with a layer (2) of a Group VIII noble metal such as palladium. The layer (2) may optionally contain other metals such as gold. The layer (2) is located with an outer edge (7) below the particle surface (3) and between an inner region (4) and an outer region (5) of the catalyst particle having lower concentrations of the Group VIII noble metal and/or other metals than the layer (2).

EXHIBIT B

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,053,024 ("*Baker*")

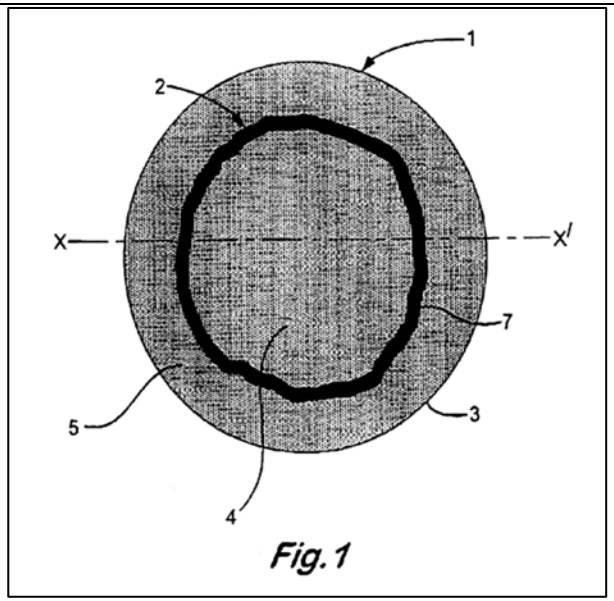
'864 patent	U.S. Patent No. 7,053,024 ("*Baker*")
	 *Fig. 1* FIG. 2 illustrates an X-ray profile from beyond and across a section of a catalyst particle containing palladium and gold using an Electron Microprobe Analyser, for example along line X-X′ of FIG. 1. FIG. 2 clearly illustrates that the palladium and gold are both mainly distributed in two specific locations along the diameter of the particle (that is, as a layer below the surface of the particle) with lower concentrations of palladium and gold distributed elsewhere. In FIG. 2 the outer edges of the particle are at positions labeled A and A′ at 0 and 97 microns.

EXHIBIT B

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,053,024 ("*Baker*")

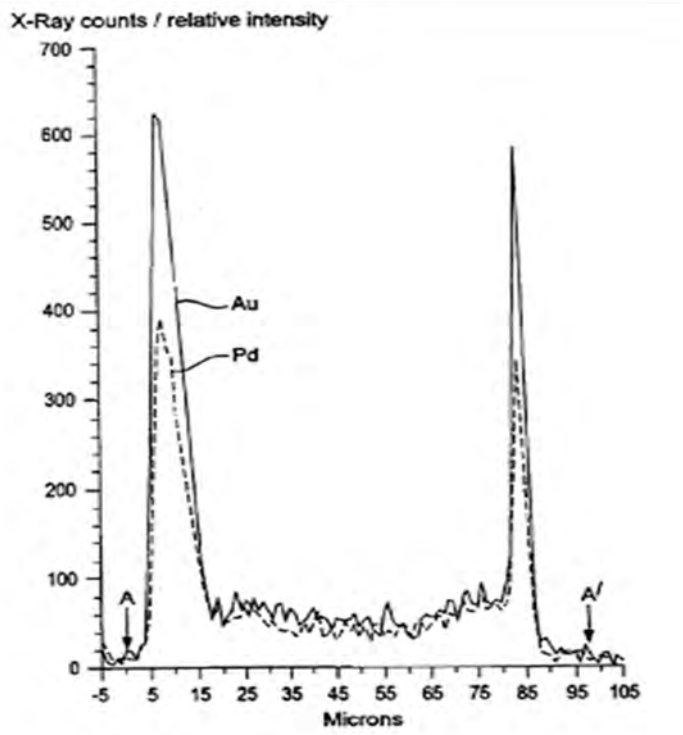
'864 patent	U.S. Patent No. 7,053,024 ("*Baker*")
	 *Fig.2* " [9:11-42] "Measurement of Noble Metal Layer Depth A sample (approx. 20 particles) from each of Comparative Examples 1 and 5 and Example 2-4 and 6-7 was set in Araldite® resin overnight at 60° C. Thin sections (<100 nm) were cut with a diamond knife from prepared ' mesas'. The images were recorded by Transmission Electron Microscopy (TEM) using a JEOL 2000FX

EXHIBIT B

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,053,024 ("*Baker*")

'864 patent	U.S. Patent No. 7,053,024 ("*Baker*")
	instrument. Photographic plates of the catalyst composition particles were then studied and measurements of the depth of the major layer of metals from the particle surface were made. No measurements were made on any particle with a diameter of 25 microns or less. The results area given in Table 1.

TABLE 1

Example	Hydrazine concentration (wt %)	Reduction temperature	Average layer depth below surface (microns)	Average layer thickness (microns)
Comparative 1	1	room temp.	1	1
2	2	room temp.	3	0.75
3	4	room temp.	6	0.25
4	8	room temp.	8	0.25
Comparative 5	1	80° C.	1	1.5
6	8	80° C.	10	0.5
7	4	room temp.	8	0.25

The data illustrates that as the hydrazine concentration increases the layer depth below the surface increases. This effect is observed at both room temperature and at 80° C. The greater the layer depth the more protected the noble metal catalyst component will be from loss by abrasion or attrition."

[10:11-16]
"Catalyst A was a shell type catalyst whereas catalyst B had been prepared by a process according to the present invention. A significant proportion of the palladium and gold in catalyst B was located in a layer with an outer edge 8 microns below the surface of the particles.

EXHIBIT B

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,053,024 ("*Baker*")

<table>
<tr><th>'864 patent</th><th>U.S. Patent No. 7,053,024 ("Baker")</th></tr>
<tr><td></td><td>Table 2 shows the amounts of palladium and gold lost by the two catalysts during the attrition test.

TABLE 2
<table><tr><th>Catalyst</th><th>palladium loss</th><th>gold loss</th></tr><tr><td>A</td><td>56.0%</td><td>52.2%</td></tr><tr><td>B</td><td>4.0%</td><td>5.9%</td></tr></table>
The results in table 2 show that the catalyst according to the present invention loses less of the catalytically active metals palladium and gold than the shell type catalyst."

To the extent that this limitation is not disclosed, either explicitly or inherently, by Baker, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.</td></tr>
<tr><td>[3] The combustion promoter according to claim 1, comprising a plurality of microsphere sized porous particles comprising alumina.</td><td>Baker discloses a combustion promoter comprising a plurality of microsphere sized porous particles comprising alumina. See, e.g.,

The support provided above for Claim 1[b].

To the extent that this limitation is not disclosed, either explicitly or inherently, by Baker, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of</td></tr>
</table>

EXHIBIT B

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,053,024 ("*Baker*")

'864 patent	**U.S. Patent No. 7,053,024 ("*Baker*")**
	the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[6] The combustion promoter according to claim 1, wherein the microsphere sized porous particles collectively have an average (D50) size of between 60 and 90 microns as measured by laser diffraction.	*Baker* discloses a combustion promoter comprising microsphere sized porous particles, wherein the microsphere sized porous particles collectively have an average (D50) size of between 60 and 90 microns as measured by laser diffraction. *See, e.g.*, The support provided above for Claim 1[a]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Baker*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[12] The combustion promoter according to claim 1, wherein the one or more Group VIII noble metals comprises one or more of platinum, palladium, iridium, ruthenium, and rhodium.	*Baker* discloses a combustion promoter comprising one or more Group VIII noble metals, wherein the one or more Group VIII noble metals comprises one or more of platinum, palladium, iridium, ruthenium, and rhodium. *See, e.g.*, The support provided above for Claim 1[c]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Baker*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted

EXHIBIT B

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,053,024 ("*Baker*")

'864 patent	**U.S. Patent No. 7,053,024 ("*Baker*")**
	for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[15] The combustion promoter according to claim 12, wherein the one or more Group VIII noble metals comprises palladium.	*Baker* discloses a combustion promoter comprising one or more Group VIII noble metals, wherein the one or more Group VIII noble metals comprises palladium. *See, e.g.*, The support provided above for Claim 1[c]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Baker*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.

EXHIBIT C

EXHIBIT C

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,981,252 ("*Carlsson*")

U.S. Patent No. 9,981,252 ("*Carlsson*"), filed on May 1, 2009 and published on May 29, 2018, qualifies as prior art under at least 35 U.S.C. §§ 102 and/or 103 for the asserted claims of U.S. Patent No. 11,224,864 ("the '864 patent"), as discussed in detail in the claim chart below.

The Court has not yet construed the claims and therefore the meaning of the terms in the claims has yet to be resolved by the Court. The support identified here for these limitations is responsive to Plaintiffs' infringement contentions, which Defendant disagrees with. As such, nothing in Defendant's claim charts should be construed as an admission regarding infringement, either literally or under the doctrine of equivalents, or as an admission regarding Defendant's understanding of the proper scope of the asserted claims. Defendant reserves the right to rely on additional citations or sources of evidence that also may be applicable, or that may become applicable in light of claim construction, changes in Plaintiffs' infringement contentions, and/or information obtained during discovery as the case progresses.

To the extent Plaintiffs allege that *Carlsson* does not disclose any particular limitation of the asserted claims of the '864 patent, either expressly or inherently, it would have been obvious to a person of ordinary skill in the art as of the priority date of the '864 patent to modify *Carlsson* and/or to combine the teachings of *Carlsson* with other prior art references, including but not limited to the present prior art references cited in the Cover Pleading and the relevant section(s) of claim charts for other prior art references for the '864 patent in a manner that would have rendered the asserted claims invalid as obvious.

Defendant reserves the right to amend or supplement this claim chart at a later date as more fully set forth in the Cover Pleading.

EXHIBIT C

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,981,252 (“*Carlsson*”)

’864 patent	**U.S. Patent No. 9,981,252 (“*Carlsson*”)**
1[a] A CO to CO_2 combustion promoter comprising microsphere sized porous particles, each microsphere sized porous particle having a diameter of less than 1mm, and independently comprising	*Carlsson* discloses a CO to CO_2 combustion promoter comprising microsphere sized porous particles, each microsphere sized porous particle having a diameter of less than 1mm, and independently comprising the limitations discussed in the following sections. *See, e.g.*, [Abstract] “A method for preparing a catalyst comprising (i) preparing a calcined shaped calcium aluminate catalyst support, (ii) treating the calcined shaped calcium aluminate support with water, and then drying the support, (iii) impregnating the dried support with a solution containing one or more metal compounds and drying the impregnated support, (iv) calcining the dried impregnated support, to form metal oxide on the surface of the support and (v) optionally repeating steps (ii), (iii) and (iv) on the metal oxide coated support. The method provides an eggshell catalyst in which the metal oxide is concentrated in an outer layer on the support.” [1:16-23] “This invention relates to a method of preparing catalysts supported on calcium aluminate. Calcium aluminate-supported catalysts are used in a number of industrial processes, including methanation and steam reforming processes, such as pre-reforming, primary reforming and secondary reforming. In such cases the catalytically-active metal is typically nickel, but other transition metals or precious metals, may also be used.” [1:49-56] “We have found that by re-hydrating the surface of the calcined shaped calcium aluminate catalyst support and then drying it, the support, once impregnated with a metal compound provides an egg-shell catalyst precursor in which the metal oxide

EXHIBIT C

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,981,252 ("*Carlsson*")

'864 patent	U.S. Patent No. 9,981,252 ("*Carlsson*")
	formed upon calcination is concentrated as an outer surface layer on the support and is not uniformly distributed. Moreover, the properties of such catalysts are enhanced in comparison to the known catalysts." [3:17-42] "The composition is desirably shaped into pellets using known techniques, but may also be prepared as extrudates or granules. The length, width and height of such shaped units may be in the range 3-50 mm. The support may be in the form of saddles as described in the aforesaid U.S. Pat. No. 4,707,351 but preferably the support is pressed into pellets in the form of cylinders, which may have one or more through holes, for example as described in WO 2004/014549. More preferably the shaped support is in the form of a cylindrical pellet having between 1 and 12 holes extending there-through, especially 3-10 holes of circular cross section, and optionally between 2 and 20 flutes or lobes running along the length of the pellet. Suitable diameters for such pellets are in the range 4-40 mm and the aspect ratio (length/diameter) is preferably $\leq$2. A particularly preferred shape is a highly domed cylindrical pellet in the form of a cylinder having a length C and diameter D, which has one or more holes extending there through, wherein the cylinder has domed ends of lengths A and B, such that (A+B+C)/D is in the range 0.50 to 2.00, and (A+B)/C is in the range 0.40 to 5.00. Such shapes are described in co- pending WO 2010/029323 Al and WO 2010/029324 Al. C is preferably in the range 1 to 25 mm and D is preferably in the range 4 to 40 mm." [3:59-62] "The shaped calcined catalyst support preferably has a total surface area, as measured by nitrogen absorption, of 0.5 to 40, particularly 1 to 15, m^2g^{-1}, and a pore volume of 0.1 to 0.3 $cm^3 \cdot g^{-1}$, as determined by mercury porosimetry."

EXHIBIT C

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,981,252 ("*Carlsson*")

'864 patent	U.S. Patent No. 9,981,252 ("*Carlsson*")
	[3:43-58] "After shaping, the cement in the shaped catalyst support should be cured and the support dried, typically at under 200° C., and then calcined. Curing of the calcium aluminate cement may take place before or during a drying step, e.g. by spraying or immersing the shaped catalyst support with water prior to drying or by heating the shaped catalyst support under conditions of controlled relative humidity prior to volatilising residual water. Calcination is typically carried out by heating the shaped units to between 500 and 1400° C. in air for between 1 and 16 hours. The catalyst support strength increases, while the porosity and surface area decrease, as the calcination temperature increases. Hence the support calcination should be effected at sufficient temperature to obtain the required mechanical strength but should not be so high that the surface area and porosity are unduly reduced." [4:31-38] "Without wishing to be bound by theory, it is believed that the re-hydration and drying steps change the surface chemistry of the calcium aluminate support. Evidence indicates that the pores in the surface of the support are not blocked; rather that the surface basicity of the support is affected so that upon impregnation, insoluble compounds of the metal impregnate precipitate at/near the surface of the support thereby generating the eggshell catalyst." [4:58-5:5] "Impregnation may be performed at ambient or elevated temperature and at atmospheric or elevated pressure using known techniques, including immersion of the re-hydrated and dried catalyst support in a metal-containing solution or by so-called 'incipient wetness' impregnation where the volume of solution used equates approximately to the pore volume of the support material. Impregnation of the metal compound at ambient temperature (i.e. 10 to 25° C.), and at atmospheric pressure (about 1 bar abs) may be used, however it has been found that by impregnating the

EXHIBIT C

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,981,252 ("*Carlsson*")

'864 patent	U.S. Patent No. 9,981,252 ("*Carlsson*")
	support at temperatures in the range 50-90° C., improved control over the thickness of the eggshell layer may be obtained. For example, the eggshell layer thickness on pelleted materials may be ≤800 μm at 20-30° C., but impregnation at 50-90°C. can produce thicknesses of ≤300 μm." [6:15-21] "The eggshell catalysts prepared according to the invention may be used in steam reforming processes such as primary steam reforming, secondary reforming of a primary reformed gas mixture and pre-reforming. The catalysts may also be used for methanation reactions, hydrogenation reactions and, in oxidic unreduced form, for the decomposition of hypochlorite in aqueous solutions." [7:33-43] "Where the catalyst is to be used for methanation, in order to remove low concentrations of CO and CO_2 (0.1-0.5% vol) from a hydrogen-containing gas, the hydrogen-containing gas is typically passed through a particulate fixed bed of a nickel-containing catalyst at a temperature in the range 230-450° C. and pressures up to about 50 bar abs or higher up to about 250 bar abs. Unlike steam reforming the catalyst are preferably simple cylindrical pellets without through holes, although such pellets may be used if desired. Typical pellet diameters are in the range 2.5-6 mm, with lengths in the same range. The catalysts may be provided in oxidic form or pre-reduced and passivated form." [7:64-8:8] "Example 1. Preparation of a Catalyst Support a) Calcium aluminate cement was blended with alumina trihydrate and lime to obtain a mixture with a Ca:Al ratio of 10:43. Graphite (4 wt %) was added, and the resulting mixture pelleted using a hydraulic tabletting machine to give cylinders of

EXHIBIT C

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,981,252 ("*Carlsson*")

'864 patent	U.S. Patent No. 9,981,252 ("*Carlsson*")
	diameter 3.3 mm and length 3.3 mm. The pellets were subjected to water-curing and calcination to obtain a calcined shaped support with the following properties. BET (Nitrogen): 5.7 m^2/g Pore volume: 0.28 cm^3/g Density: 1.66 g/cc (b) The method of Example l(a) was repeated to produce pellets of diameter 5.4 mm and length 3.0 mm. (c) The method of Example 1(a) was repeated except that calcium aluminate cement was blended with alumina trihydrate to obtain a mixture with a Ca:Al ratio of 10:74." [9:49-59] "Example 4. Preparation of Catalysts a) Catalyst Support. Calcium aluminate cement was blended with alumina trihydrate and lime to obtain a mixture with a Ca:Al ratio of 10:43. Graphite (4 wt%) was added, and the resulting mixture pelleted using a hydraulic tabletting machine to give cylinders of diameter 5.4 mm and length 5.4 mm. The pellets were subjected to water-curing and calcination to obtain a calcined shaped support with the same properties as example 1(a)." To the extent that this limitation is not disclosed, either explicitly or inherently, by *Carlsson*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and

EXHIBIT C

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,981,252 ("*Carlsson*")

'864 patent	**U.S. Patent No. 9,981,252 ("*Carlsson*")**
	predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
1[b] silica, alumina, or mixtures thereof; and	*Carlsson* discloses a combustion promoter comprising silica, alumina, or mixtures thereof. *See, e.g.*, [Abstract] "A method for preparing a catalyst comprising (i) preparing a calcined shaped calcium aluminate catalyst support, (ii) treating the calcined shaped calcium aluminate support with water, and then drying the support, (iii) impregnating the dried support with a solution containing one or more metal compounds and drying the impregnated support, (iv) calcining the dried impregnated support, to form metal oxide on the surface of the support and (v) optionally repeating steps (ii), (iii) and (iv) on the metal oxide coated support. The method provides an eggshell catalyst in which the metal oxide is concentrated in an outer layer on the support." [1:16-45] "This invention relates to a method of preparing catalysts supported on calcium aluminate. Calcium aluminate-supported catalysts are used in a number of industrial processes, including methanation and steam reforming processes, such as pre-reforming, primary reforming and secondary reforming. In such cases the catalytically-active metal is typically nickel, but other transition metals or precious metals, may also be used. In methanation and steam reforming processes, pellets comprising nickel oxide on alumina or calcium aluminate are typically installed and the reduction of the nickel oxide to the active elemental nickel carried out in-situ.

EXHIBIT C

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,981,252 ("*Carlsson*")

'864 patent	U.S. Patent No. 9,981,252 ("*Carlsson*")
	U.S. Pat. No. 4,707,351 describes steam-reforming catalysts made of a low-silica calcium aluminate cement composition in a saddle configuration. The catalysts were prepared by mixing calcium aluminate with water and polyvinylacetate, stamping shapes from the resulting material, drying and calcining the saddles at up to 1400° C. before impregnation with nickel nitrate. The impregnated saddles were further dried and calcined to generate the catalyst precursor. In this process, the calcium aluminate support was hydrated during the shaping process and then calcined to increase its strength and define the micromimetric properties prior to impregnation with nickel nitrate. Heretofore it has been seen as necessary to impregnate calcium aluminate supports to achieve a uniform dispersion of the metal compound within the pellets such that upon calcination the metal oxide is uniformly dispersed within the pellet, thereby maximising the metal surface area and hence catalyst activity." [1:49-2:4] "We have found that by re-hydrating the surface of the calcined shaped calcium aluminate catalyst support and then drying it, the support, once impregnated with a metal compound provides an egg-shell catalyst precursor in which the metal oxide formed upon calcination is concentrated as an outer surface layer on the support and is not uniformly distributed. Moreover, the properties of such catalysts are enhanced in comparison to the known catalysts. Accordingly, the invention provides a method for preparing a catalyst comprising the steps of: (i) preparing a calcined shaped calcium aluminate catalyst support, (ii) treating the calcined shaped calcium aluminate support with water, and then drying the support,

EXHIBIT C

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,981,252 ("*Carlsson*")

'864 patent	U.S. Patent No. 9,981,252 ("*Carlsson*")
	(iii) impregnating the dried support with a solution containing one or more metal compounds and drying the impregnated support, (iv) calcining the dried impregnated support, to form metal oxide on the surface of the support and (v) optionally repeating steps (ii), (iii) and (iv) on the metal oxide coated support. The invention further provides an eggshell catalyst obtainable by the method." [2:19-3:4] "The catalyst support is prepared from a calcium aluminate cement. By the term calcium aluminate cement we include those hydraulic cements containing one or more calcium aluminate compounds of the formula $nCaO.mAl_2O_3$ where n and m are integers. Example of such calcium aluminate compounds include calcium monoaluminate ($CaO.Al_2O_3$), tricalcium aluminate ($3CaO.Al_2O_3$), penta calcium trialuminate ($5CaO.3Al_2O_3$), tricalcium penta aluminate ($3CaO.5Al_2O_3$), and dodeca calcium hepta aluminate ($12CaO.7Al_2O_3$). Some calcium aluminate cements, e.g. the so-called 'high alumina' cements, may contain alumina in admixture with, dissolved in, or combined with, such calcium aluminate compounds. For example, a well known commercial high alumina cement has a composition corresponding to about 18% calcium oxide, 79% alumina and 3% water and other oxides. This material has a calcium:aluminium atomic ratio of about 1:5, i.e. $2CaO.5Al_2O_3$. Calcium aluminates are often contaminated with iron compounds, but these are not believed to be detrimental to the present invention. Suitable cements include the commercially available Ciment Fondu, and Secar 50, Secar 71, Secar 80 available from Kemeos and CA-25, CA-14, CA-270 available from Almatis. The support composition employed in the present invention preferably has a calcium:aluminium atomic ratio within the range 1:3 to 1:12, more preferably 1:3 to

EXHIBIT C

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,981,252 ("*Carlsson*")

'864 patent	U.S. Patent No. 9,981,252 ("*Carlsson*")
	1:10, most preferably 1:4 to 1:8. Where the calcium aluminate cement is a 'high alumina' cement, no additional alumina may be necessary but, in general, the support is desirably made from a calcium aluminate cement to which an additional amount of alumina, which may be in the form of a transition alumina, monohydrate or trihydrate, has been added. To accelerate setting, an amount of lime (CaO), e.g. up to 15% by weight of the composition, may also be incorporated into the support composition. Hence the support will usually be a refractory composition consisting of a calcined mixture of alumina, one or more of said calcium aluminate compounds and optionally lime. Other oxidic materials, e.g. titania, zirconia or lanthana, may be present in the calcium aluminate support composition. While silica may in some cases be incorporated, for use as a steam reforming support, a low silica content, i.e. less than 1% by weight, preferably less than 0.5% by weight, based on the weight of the oxidic material in the support composition is desirable, as silica has an appreciable volatility under steam reforming conditions. The support composition preferably contains ≤25% by weight, more preferably ≤15% by weight, most preferably ≤10% by weight of oxidic material other than calcium aluminate and alumina. The shaped catalyst support may be made by forming a calcium aluminate cement powder, optionally with additional alumina and/or lime, into the desired shape, curing the cement and subsequently calcining the shaped support." [3:11-16] "A typical composition suitable for pellet formation comprises 30 to 70% by weight of a calcium aluminate cement (comprising 65 to 85% by weight of alumina and 15 to 35% by weight of CaO) mixed with 24 to 48% by weight of alumina, 0 to 15% by weight of lime, and 2 to 5% by weight of graphite."

EXHIBIT C

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,981,252 (“*Carlsson*”)

’864 patent	U.S. Patent No. 9,981,252 (“*Carlsson*”)
	[7:64-8:15] “Example 1. Preparation of a Catalyst Support a) Calcium aluminate cement was blended with alumina trihydrate and lime to obtain a mixture with a Ca:Al ratio of 10:43. Graphite (4 wt %) was added, and the resulting mixture pelleted using a hydraulic tabletting machine to give cylinders of diameter 3.3 mm and length 3.3 mm. The pellets were subjected to water-curing and calcination to obtain a calcined shaped support with the following properties. BET (Nitrogen): 5.7 m^2/g Pore volume: 0.28 cm^3/g Density: 1.66 g/cc (b) The method of Example l(a) was repeated to produce pellets of diameter 5.4 mm and length 3.0 mm. (c) The method of Example 1(a) was repeated except that calcium aluminate cement was blended with alumina trihydrate to obtain a mixture with a Ca:Al ratio of 10:74.” [9:49-59] “Example 4. Preparation of Catalysts a) Catalyst Support. Calcium aluminate cement was blended with alumina trihydrate and lime to obtain a mixture with a Ca:Al ratio of 10:43. Graphite (4 wt%) was added, and the resulting mixture pelleted using a hydraulic tabletting machine to give cylinders of diameter 5.4 mm and length 5.4 mm. The pellets were subjected to water-curing and calcination to obtain a calcined shaped support with the same properties as example 1(a).”

EXHIBIT C

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,981,252 ("*Carlsson*")

'864 patent	**U.S. Patent No. 9,981,252 ("*Carlsson*")**
	To the extent that this limitation is not disclosed, either explicitly or inherently, by *Carlsson*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
1[c] one or more Group VIII noble metals distributed in the particle as an eggshell such that a higher concentration of the one or more Group VIII noble metals is present in the outer region of the microsphere sized porous particle as compared to the concentration of the one or more Group VIII noble metals in the centre of the microsphere sized porous particle.	*Carlsson* discloses a combustion promoter comprising one or more Group VIII noble metals, the metals distributed in the particle as an eggshell such that a higher concentration of the one or more Group VIII noble metals is present in the outer region of the microsphere sized porous particle as compared to the concentration of the one or more Group VIII noble metals in the centre of the microsphere sized porous particle. *See, e.g.*, [Abstract] "A method for preparing a catalyst comprising (i) preparing a calcined shaped calcium aluminate catalyst support, (ii) treating the calcined shaped calcium aluminate support with water, and then drying the support, (iii) impregnating the dried support with a solution containing one or more metal compounds and drying the impregnated support, (iv) calcining the dried impregnated support, to form metal oxide on the surface of the support and (v) optionally repeating steps (ii), (iii) and (iv) on the metal oxide coated support. The method provides an eggshell catalyst in which the metal oxide is concentrated in an outer layer on the support." [1:16-23] "Calcium aluminate-supported catalysts are used in a number of industrial processes, including methanation and steam reforming processes, such as pre-

EXHIBIT C

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,981,252 ("*Carlsson*")

'864 patent	U.S. Patent No. 9,981,252 ("*Carlsson*")
	reforming, primary reforming and secondary reforming. In such cases the catalytically-active metal is typically nickel, but other transition metals or precious metals, may also be used." [1:49-2:8] "We have found that by re-hydrating the surface of the calcined shaped calcium aluminate catalyst support and then drying it, the support, once impregnated with a metal compound provides an egg-shell catalyst precursor in which the metal oxide formed upon calcination is concentrated as an outer surface layer on the support and is not uniformly distributed. Moreover, the properties of such catalysts are enhanced in comparison to the known catalysts. Accordingly, the invention provides a method for preparing a catalyst comprising the steps of: (i) preparing a calcined shaped calcium aluminate catalyst support, (ii) treating the calcined shaped calcium aluminate support with water, and then drying the support, (iii) impregnating the dried support with a solution containing one or more metal compounds and drying the impregnated support, (iv) calcining the dried impregnated support, to form metal oxide on the surface of the support and (v) optionally repeating steps (ii), (iii) and (iv) on the metal oxide coated support. The invention further provides an eggshell catalyst obtainable by the method.

EXHIBIT C

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,981,252 ("*Carlsson*")

'864 patent	U.S. Patent No. 9,981,252 ("*Carlsson*")
	The invention further provides a process for the steam reforming of hydrocarbons comprising the step of contacting a mixture of hydrocarbon and steam at elevated temperature and pressure with the eggshell catalyst." [2:12-18] "By the term 'eggshell catalyst' we mean that the catalytically active metal or metals are not uniformly distributed within the catalyst support but are concentrated at the surface and therefore form a thin layer, with the metal or metals being absent beneath this layer. The thickness of the eggshell layer is preferably ≤ 1000 μm, more preferably ≤ 800 μm, most preferably ≤ 300 μm." [4:31-38] "Without wishing to be bound by theory, it is believed that the re-hydration and drying steps change the surface chemistry of the calcium aluminate support. Evidence indicates that the pores in the surface of the support are not blocked; rather that the surface basicity of the support is affected so that upon impregnation, insoluble compounds of the metal impregnate precipitate at/near the surface of the support thereby generating the eggshell catalyst." [4:58-5:16] "Impregnation may be performed at ambient or elevated temperature and at atmospheric or elevated pressure using known techniques, including immersion of the re-hydrated and dried catalyst support in a metal-containing solution or by so-called 'incipient wetness' impregnation where the volume of solution used equates approximately to the pore volume of the support material. Impregnation of the metal compound at ambient temperature (i.e. 10 to 25° C.), and at atmospheric pressure (about 1 bar abs) may be used, however it has been found that by impregnating the support at temperatures in the range 50-90° C., improved control over the thickness of the eggshell layer may be obtained. For example, the eggshell layer thickness on

EXHIBIT C

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,981,252 (“*Carlsson*”)

’864 patent	U.S. Patent No. 9,981,252 (“*Carlsson*”)
	pelleted materials may be ≤ 800 μm at 20-30° C., but impregnation at 50-90° C. can produce thicknesses of ≤ 300 μm. Following impregnation, the impregnated support is dried and calcined. Drying conditions are preferably the same as those used following the re-hydration step. The calcination step to convert the impregnated metal compound to its corresponding metal oxide is preferably performed in air at a temperature in the range 250-850° C. An advantage of the present invention, by virtue of the lower metal content and the increased metal concentration at the surface of the catalysts, is that the amount of nitrogen oxides evolved during calcination of metal nitrate-based precursors can be reduced compared with current catalyst materials. The catalytic metal content of the resulting catalyst may be determined by a number of factors such as the metal content of the solution and the impregnation conditions. Steam reforming catalysts formed by impregnation typically have a NiO content in the range 10-35% wt. Precipitated pre-reforming catalysts can have NiO contents of 40-80% wt or more. Methanation catalysts typically have a NiO content in the region of 30-35% wt. In the present invention, because the catalytic metal oxide is concentrated at the surface of the 25 support it is possible to achieve improved catalyst activity with reduced metal loadings. This has clear commercial benefits. The catalytic metal oxide content of the calcined catalyst is preferably in the range 2-25% wt, preferably 4-15% wt. Thus one impregnation may be sufficient to generate the desired catalyst. However if desired, the re-hydration, drying and impregnation steps may be repeated until the metal oxide content of the calcined material is above 2.5% wt, preferably above 5% wt, more preferably above 7.5% wt, most preferably above 10% wt. Multiple impregnations may be performed using the same or different catalytically active metal. In order to maintain the eggshell catalyst, the metal oxide containing support should be re- hydrated, dried and calcined prior to each metal impregnation.”

EXHIBIT C

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,981,252 ("*Carlsson*")

'864 patent	U.S. Patent No. 9,981,252 ("*Carlsson*")
	[5:35-40] "Multiple impregnations may be performed using the same or different catalytically active metal. In order to maintain the eggshell catalyst, the metal oxide containing support should be re-hydrated, dried and calcined prior to each metal impregnation." [5:44-58] "One or more promoter compounds may be impregnated into the dried support and/or the metal oxide coated support. Hence one or more promoter compounds may be included in the metal impregnating solution or the promoter may be added subsequently by a separate impregnation. The promoter may be confined to the eggshell layer or may be distributed throughout the catalyst support. Promoters include precious metals such as platinum, palladium, iridium, ruthenium, rhodium and gold. Lanthanide metals such as lanthanum and cerium may also be included as promoters. Water-soluble salts, particularly nitrates, may be used as sources of the metal promoters. More than one promoter may be present and additional alkali may also be added. The amount of promoter metal will typically be in the range 0.1-5% wt on the calcined catalyst material." [6:15-21] "The eggshell catalysts prepared according to the invention may be used in steam reforming processes such as primary steam reforming, secondary reforming of a primary reformed gas mixture and pre-reforming. The catalysts may also be used for methanation reactions, hydrogenation reactions and, in oxidic unreduced form, for the decomposition of hypochlorite in aqueous solutions." [7:49-57] "FIG. 1 depicts an image of a cylindrical catalyst pellet cut in two to show an eggshell layer of catalyst prepared according to the present invention;

EXHIBIT C

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,981,252 (“*Carlsson*”)

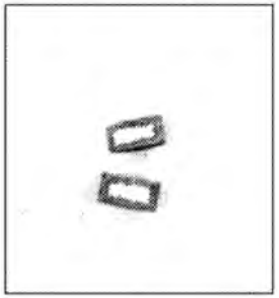
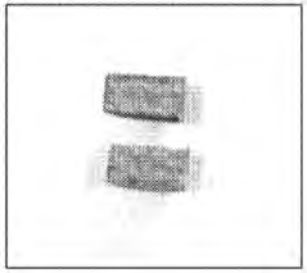
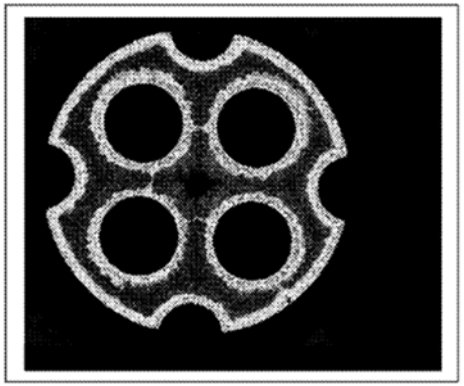
’864 patent	U.S. Patent No. 9,981,252 (“*Carlsson*”)
	FIG. 2 depicts an image of a similar catalyst pellet prepared according to the prior art, and FIG. 3 depicts an electron-probe micro-analyser (EMPA) image of a cross section of a lobed 4-hole cylindrical catalyst pellet with an eggshell layer of catalyst prepared according to the present invention.”  Figure 1  Figure 2 (Comparative)  Figure 3

EXHIBIT C

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,981,252 ("*Carlsson*")

'864 patent	**U.S. Patent No. 9,981,252 ("*Carlsson*")**
	[8:23-35] "(b) Incorporation of Ni. The catalyst support pellets were then immersed in a solution of nickel nitrate in de-mineralised water (200 g Ni/liter) for 5 minutes at 25° C. The impregnated pellets were then removed and allowed to drain for 10 minutes and dried at 110° C. for 6 hours. The dried impregnated pellets were then heated at 100° C./hour to 650° C. and then held at 650° C. for 4 hours to convert the nickel nitrate to nickel oxide. The re-hydrating, drying, impregnating, drying and calcining procedure was repeated on the nickel oxide containing pellets a further two times. The Ni was concentrated in a thin layer around the edge of the catalyst pellet as shown in FIG. 1. The thickness of the eggshell layer is about 800 μm." [10:10-14] "The pellets were analysed by optical microscopy and in each case it was found that the metal compound was concentrated around the edge of the pellet, i.e. that an eggshell material had been formed." To the extent that this limitation is not disclosed, either explicitly or inherently, by *Carlsson*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[3] The combustion promoter according to claim 1, comprising a plurality of microsphere sized porous particles comprising alumina.	*Carlsson* discloses a combustion promoter comprising a plurality of microsphere sized porous particles comprising alumina. *See, e.g.*, The support provided above for Claim 1[b].

EXHIBIT C

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,981,252 ("*Carlsson*")

'864 patent	**U.S. Patent No. 9,981,252 ("*Carlsson*")**
	To the extent that this limitation is not disclosed, either explicitly or inherently, by *Carlsson*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[6] The combustion promoter according to claim 1, wherein the microsphere sized porous particles collectively have an average (D50) size of between 60 and 90 microns as measured by laser diffraction.	*Carlsson* discloses a combustion promoter comprising microsphere sized porous particles, wherein the microsphere sized porous particles collectively have an average (D50) size of between 60 and 90 microns as measured by laser diffraction. *See, e.g.*, The support provided above for Claim 1[a]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Carlsson*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[12] The combustion promoter according to claim 1, wherein the one or more Group VIII noble metals comprises one or more of platinum, palladium, iridium, ruthenium, and rhodium.	*Carlsson* discloses a combustion promoter comprising one or more Group VIII noble metals, wherein the one or more Group VIII noble metals comprises one or more of platinum, palladium, iridium, ruthenium, and rhodium. *See, e.g.*, The support provided above for Claim 1[c].

EXHIBIT C

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,981,252 ("*Carlsson*")

'864 patent	**U.S. Patent No. 9,981,252 ("*Carlsson*")**
	To the extent that this limitation is not disclosed, either explicitly or inherently, by *Carlsson*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[15] The combustion promoter according to claim 12, wherein the one or more Group VIII noble metals comprises palladium.	*Carlsson* discloses a combustion promoter comprising one or more Group VIII noble metals, wherein the one or more Group VIII noble metals comprises palladium. *See, e.g.*, The support provided above for Claim 1[c]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Carlsson*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.

EXHIBIT D

EXHIBIT D

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,230,035 ("*Espinoza*")

U.S. Patent No. 7,230,035 ("*Espinoza*"), filed on December 30, 2002 and published on June 12, 2007, qualifies as prior art under at least 35 U.S.C. §§ 102 and/or 103 for the asserted claims of U.S. Patent No. 11,224,864 ("the '864 patent"), as discussed in detail in the claim chart below.

The Court has not yet construed the claims and therefore the meaning of the terms in the claims has yet to be resolved by the Court. The support identified here for these limitations is responsive to Plaintiffs' infringement contentions, which Defendant disagrees with. As such, nothing in Defendant's claim charts should be construed as an admission regarding infringement, either literally or under the doctrine of equivalents, or as an admission regarding Defendant's understanding of the proper scope of the asserted claims. Defendant reserves the right to rely on additional citations or sources of evidence that also may be applicable, or that may become applicable in light of claim construction, changes in Plaintiffs' infringement contentions, and/or information obtained during discovery as the case progresses.

To the extent Plaintiffs allege that *Espinoza* does not disclose any particular limitation of the asserted claims of the '864 patent, either expressly or inherently, it would have been obvious to a person of ordinary skill in the art as of the priority date of the '864 patent to modify *Espinoza* and/or to combine the teachings of *Espinoza* with other prior art references, including but not limited to the present prior art references cited in the Cover Pleading and the relevant section(s) of claim charts for other prior art references for the '864 patent in a manner that would have rendered the asserted claims invalid as obvious.

Defendant reserves the right to amend or supplement this claim chart at a later date as more fully set forth in the Cover Pleading.

EXHIBIT D

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,230,035 ("*Espinoza*")

'864 patent	**U.S. Patent No. 7,230,035 ("*Espinoza*")**
1[a] A CO to CO_2 combustion promoter comprising microsphere sized porous particles, each microsphere sized porous particle having a diameter of less than 1mm, and independently comprising	*Espinoza* discloses a CO to CO_2 combustion promoter comprising microsphere sized porous particles, each microsphere sized porous particle having a diameter of less than 1mm, and independently comprising the limitations discussed in the following sections. *See, e.g.*, [Abstract] "The present invention relates to improved catalyst compositions, as well as methods of making and using such compositions to prepare synthesis gas and ultimately C_{5+} hydrocarbons. In particular, preferred embodiments of the present invention comprise catalyst systems comprising a core and an outer region disposed on said core, wherein a substantial amount of the catalytic metal is located in the outer region of the catalyst support matrix. In addition, the catalyst systems are able to maintain high conversion and selectivity values with very low catalytically active metal loadings. The catalyst systems are appropriate for improved syngas, oxidative dehydrogenation and other partial oxidation reactions, including improved reaction schemes for the conversion of hydrocarbon gas to C_{5+} hydrocarbons." [1:25-30] "The present invention generally relates to catalysts and processes for the catalytic conversion of hydrocarbons (e.g., natural gas) to produce carbon monoxide and hydrogen (synthesis gas). More particularly, the invention relates to such catalysts and their manner of making, and to processes employing the catalysts for production of synthesis gas." [2:50-53] "It should be appreciated that volume here and further herein is contemplated to include both the solid and pore volumes that make up a particle structure."

EXHIBIT D

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,230,035 ("*Espinoza*")

'864 patent	U.S. Patent No. 7,230,035 ("*Espinoza*")
	[3:21-30] "Non-porous material (defined as a material with a surface area of less than 1 m^2/g) would typically provide a catalyst core without significant diffusion of reactants/products and thus would overcome the production of undesirable by-products. However, the use of a non-porous support material for the catalyst core is not an acceptable solution, because non-porous materials do not have a sufficient amount of surface area on which to disperse the catalytic material. In addition, lack of sufficient surface area can prevent the reactions from igniting." [3:49-4:4] "The present invention is directed towards improved catalyst systems, as well as methods of making and using such systems. The present invention provides a syngas catalyst system comprising a core and an outer layer comprising a catalytically active metal, in which effective use of the catalyst metal is enhanced and the occurrence of undesirable secondary reactions is inhibited. The invention achieves this result by minimizing diffusion of the reactant gases into the core of the catalyst particle. Thus, the reactant gases are not allowed to linger in the particle core and react unfavorably, and the expensive catalytically active materials are only placed where their presence is most economically beneficial. In particular, preferred embodiments of the present invention comprise new partial oxidation catalysts and methods of preparing and using said catalysts. Accordingly, the present invention also encompasses an improved method for converting a hydrocarbon containing gas and an oxygen containing gas to a gas mixture comprising hydrogen and carbon monoxide, i.e., syngas, using catalyst systems in accordance with the present invention. In addition, the present invention contemplates an improved method for converting hydrocarbon gas to liquid hydrocarbons using the novel syngas catalysts described herein."

EXHIBIT D

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,230,035 ("*Espinoza*")

'864 patent	U.S. Patent No. 7,230,035 ("*Espinoza*")
	[4:26-47] "The present invention further relates to methods for preparing catalyst compositions in accordance with the present invention. For example, a method for preparing nonmicroporous catalyst particles, each having a core comprising a refractory support like alumina or zirconia and an outer layer, includes applying a pore blocking material such as silicic acid or sodium carbonate to the support material, so as to substantially block the micropores of the support material and obtain a nonmicroporous core of the catalyst particle. The particles are then provided a catalytically active material for catalyzing reactions, wherein the catalytically active material preferably comprises a Group VIII metal like rhodium and optionally a promoter metal such as a lanthanide metal or rhenium, dispersed in the outer layer, wherein the catalytically active material does not substantially penetrate and deposit into the core because of the blocked pores. Another embodiment involves preparing catalysts using an impregnation technique employing a fast drying or low vapor pressure solvent, which allows preferential deposition of the active material in an outer layer, preferably deposited on the outer surface of the support." [5:36-51] "The present invention relates to improved catalyst systems. The present invention also comprises methods of preparing and using the catalyst systems. The present invention is more preferably directed towards syngas catalysts used in partial oxidation reactions and even more preferably used in syngas catalysts that contain Group VIII or noble metals. However, it should be appreciated that the catalyst systems according to the present invention are useful for other partial oxidation reactions, which are intended to be within the scope of the present invention. In addition, the present invention contemplates an improved method for converting hydrocarbon gas to C_{5+} hydrocarbons using the novel syngas catalyst systems described herein. Thus, the invention also relates to processes for converting

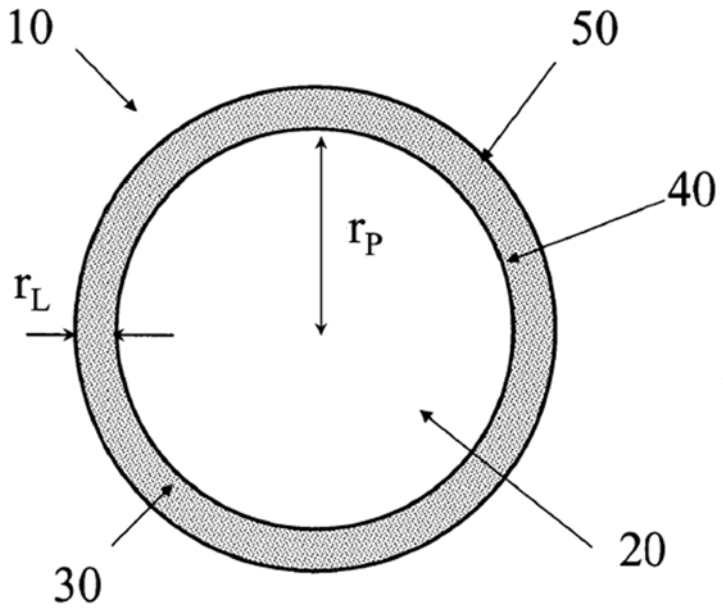
EXHIBIT D

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,230,035 ("*Espinoza*")

'864 patent	U.S. Patent No. 7,230,035 ("*Espinoza*")
	hydrocarbon-containing gas to C_{5+} hydrocarbons via an integrated syngas to Fischer-Tropsch process." [5:52-6:23] "One embodiment of the present invention is for a catalyst system in which the majority of the catalytic metal is near the surface of the support matrix. In other words, the catalyst is not uniformly dispersed throughout the catalyst matrix, but instead has regions of differing catalyst concentration. For example, as shown in FIG. 1, the support could have more than one discrete region comprising differing catalyst concentrations. As shown, the spherical catalyst system 10 has two regions 20 and 30. Region 20 is the inner core, and region 30, also called outer layer, is the area at the periphery of the core. Region 30 is delimited by exterior surface 50 of catalyst system 10 and outer surface 40 of the region 20. In the broadest and simplest terms and in accordance with the present invention, region 20 should have a lower amount of catalyst than region 30. In other words, the catalyst material should be distributed such that if region 20 and 30 were divided into two sections of equal volume, i.e., 50-50, more than 50% of the catalytically active material would be located in region 30. In a preferred embodiment, region 20 would comprise substantially lower amounts of catalyst than region 30. The general objective being that more than 50% of the catalytically active material is located in a contiguous region of the support that comprises 50% or less of the total support volume. In a more preferred embodiment, region 20 would have substantially zero catalytic material.

EXHIBIT D

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,230,035 ("*Espinoza*")

'864 patent	U.S. Patent No. 7,230,035 ("*Espinoza*")
	 Figure 1 In another embodiment of the present invention shown in FIG. 2, a catalyst particle 110 has a core 120 with an outer surface 140, a catalytic outer layer 130 with an exterior surface 150, and a substantially blocked-micropore layer 135. The substantially blocked-micropore layer 135 substantially prevents access to deposition of catalytic material in internal surface of core 120 of the catalyst particle 110. Thus, catalyst material are deposited primarily only on the outer surface 140 of

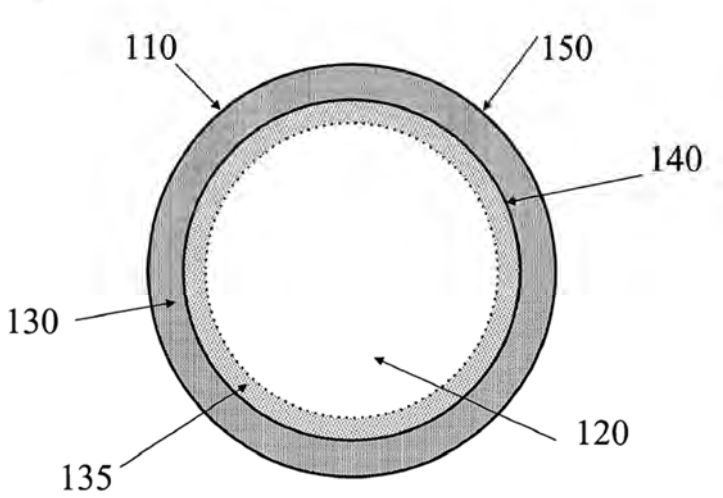
'864 patent	U.S. Patent No. 7,230,035 ("*Espinoza*")
	the core 120 and form the outer layer 130. The substantially blocked-micropore layer 135 can be of any thickness, so long as it substantially blocks access of the catalytic material to the core 120 of the catalyst particle 110. It is preferred that the catalytic layer 130 be not more than 300 micron thick, and greater than 0.5 micron thick." [Fig. 2]  Figure 2 " [6:44-7:7] "It should also be appreciated that although a spherical-shape structure for the support is illustrated in FIGS. 1-2 and a trilobe-shape structure for the support is illustrated in FIG. 3, the present invention is not intended to be so limited and is

EXHIBIT D

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,230,035 ("*Espinoza*")

'864 patent	U.S. Patent No. 7,230,035 ("*Espinoza*")
	equally applicable to any shape so long as the catalytic material is predominately in the region that is delimited by the outer surface of the core and the exterior surface of the catalyst. In addition, some combination of discrete and graduated regions could be used. In another embodiment, catalyst systems that have less than 60% of their total volume within 300 μm of the catalyst support outer surface should have at least 60% of the catalytically active metal located within 300 μm of the catalyst exterior surface, preferably wherein about 75% of the catalytically active metal is located within 250 μm of the catalyst exterior surface, and still more preferably 80% of the catalytically active metal is located within 200 μm of the catalyst exterior surface. By locating the catalytic metal near the outer surface of the support, a larger percentage of expensive catalytic metal can be more efficiently used because less of the metal will be exiled to the core and fewer side reactions are possible because fewer undesirable products will be formed in the core. It should be understood that for the present disclosure depositing at or near the outer surface of a support particle may include (1) on its outer surface causing an 'extension' of the surface with continued buildup of catalyst material, (2) on the outer surface and inside the pores and/or pore surfaces such that little buildup, if any, occurs, or (3) some combination of the two, e.g., some deposition within the pores with an average buildup on the outer surface." [7:19-41] "By combining the above-identified features, the present invention becomes a significant improvement over catalysts with more uniform dispersions of catalytic metal and higher catalyst loadings on the internal surface of a support material. Accordingly, in another preferred embodiment, the catalyst systems of the present invention comprise catalytically active metal loadings of preferably 1 wt % or less, more preferably 0.75 wt% or less and still more preferably 0.5 wt% or less, wherein

EXHIBIT D

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,230,035 ("*Espinoza*")

'864 patent	U.S. Patent No. 7,230,035 ("*Espinoza*")
	more than 50% of the catalytically active material is located in a contiguous region of the support that comprises 50% or less of the total support volume and wherein that region contains all of the outer surface of the support. In another embodiment, it is preferred that catalyst systems that have less than 60% of their volume within 300 μm of the catalyst exterior surface comprise catalytically active metal loadings of preferably 1 wt % or less, more preferably 0.75 wt % or less and still more preferable 0.5 wt % or less, wherein at least 60% of the catalytically active metal is located within 300 μm of the catalyst exterior surface, preferably wherein about 75% of the catalytically active metal is located within 250 μm of the catalyst exterior surface, and still more preferably 80% within 200 μm of the catalyst exterior surface." [7:51-67] "The support material may comprise any well known in the art, such as refractory support materials like alumina, titania, zirconia, and the like, preferably alumina. The term porous support material as used herein is meant to include all supports that have a preferred initial surface area of at least 2 m^2/g and preferably at least 5 m^2/g. The support structures may be monoliths, particulates or any other shape or form that can be prepared from the compositions of the present invention. Often, the shape or form selected will dictate the type of catalyst bed that must be used. For example, fixed beds are comprised of monoliths and large particle sized supports (greater than 0.5 mm, preferably greater than about 1 mm). Small-size support particles (less than 0.5 mm, preferably less than 0.25 mm) tend to be more useful in fluidized beds. The more preferred embodiments of the present invention comprise fixed bed reactors having large to medium sized particulate type supports." [8:39-59] "Methods for the preparation of the catalyst systems described herein are not critical. Any method by which the catalytic metal can be dispersed within the

EXHIBIT D

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,230,035 ("*Espinoza*")

'864 patent	U.S. Patent No. 7,230,035 ("*Espinoza*")
	catalyst matrix in accordance with the present invention will be sufficient. For example, the following known techniques have been found to be useful in either blocking the pores of a porous support or preferentially depositing catalytic material near the outer surface of the porous catalyst support: (i) applying a solution of a pore-blocking agent such as silicic acid or sodium carbonate, to the support particle, and calcining the catalyst particle such that a substantial portion of micropores of the particle are filled or blocked; (ii) immersion in water followed by drying at a temperature ranging from about 150°F. to about 300° F. in a microwave oven, or the like, and then calcining in air; (iii) using a fast drying or low vapor pressure solvent during an impregnation step; and (iv) impregnation of 'egg yolks' by selecting an appropriate combination of a metal compound, redox reaction and type of support, e.g., hydroxyl ion-generating redox reaction with an acidic reacting support or a hydrogen ion-generating redox reaction with a basic support."

EXHIBIT D

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,230,035 ("*Espinoza*")

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[Table 1]

TABLE 1

Catalyst Compositions

Cat-a-lyst Ex.	Support	Sodium carbonate used	Loading: Rhodium	Loading: Promoter(s)
A	Alpha-alumina (Sasol); 5.1 m^2/g; 0.3–0.6 mm granules	3%	0.5%	0.5% Sm
B	Alpha-alumina (Norpor-Saint-Gobain); 4.4. m^2/g; 1 mm granules;	4%	1.0%	0.05% Re, 0.05% Zr
C	Alpha-alumina (Norpor-Saint-Gobain); 4.4. m^2/g; 1 mm granules	4%	0.75%	0.5% Sm
D	Alpha-alumina; 5.1 m^2/g; 1.6 mm extrudate; trilobes	5%	0.5%	0.5% Sm
E	Alpha-alumina (Sasol); 5.1 m^2/g; 0.3–0.6 mm granules	2%	0.5%	0.5% Sm
F	Alpha-alumina (Sasol); 5.1 m^2/g; 0.3–0.6 mm granules	1%	0.5%	0.5% Sm
G	Alpha-alumina (Sasol); 5.1 m^2/g; 0.3–0.6 mm granules	2.5%	0.5%	0.5% Sm
H	Alpha-alumina (Sasol); 5.1 m^2/g; 0.3–0.6 mm granules	1.75%	0.5%	0.5% Sm

EXHIBIT D

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,230,035 ("*Espinoza*")

'864 patent	**U.S. Patent No. 7,230,035 ("*Espinoza*")**
	To the extent that this limitation is not disclosed, either explicitly or inherently, by *Espinoza*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
1[b] silica, alumina, or mixtures thereof; and	*Espinoza* discloses a combustion promoter comprising silica, alumina, or mixtures thereof. *See, e.g.*, [2:28-36] "As stated above, these reactions are catalytic reactions and the literature is replete with varying catalyst compositions. The more preferred catalytic metals are Group VIII or noble metals, such as rhodium, platinum, or palladium. These metals are sometimes combined with secondary metals (also called promoter metals) to enhance their activity. Further, the catalyst compositions may include a support material such as alumina, silica, titania, zirconia, and the like." [4:26-47] "The present invention further relates to methods for preparing catalyst compositions in accordance with the present invention. For example, a method for preparing nonmicroporous catalyst particles, each having a core comprising a refractory support like alumina or zirconia and an outer layer, includes applying a pore blocking material such as silicic acid or sodium carbonate to the support material, so as to substantially block the micropores of the support material and obtain a nonmicroporous core of the catalyst particle. The particles are then provided a catalytically active material for catalyzing reactions, wherein the

EXHIBIT D

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,230,035 ("*Espinoza*")

'864 patent	U.S. Patent No. 7,230,035 ("*Espinoza*")
	catalytically active material preferably comprises a Group VIII metal like rhodium and optionally a promoter metal such as a lanthanide metal or rhenium, dispersed in the outer layer, wherein the catalytically active material does not substantially penetrate and deposit into the core because of the blocked pores. Another embodiment involves preparing catalysts using an impregnation technique employing a fast drying or low vapor pressure solvent, which allows preferential deposition of the active material in an outer layer, preferably deposited on the outer surface of the support." [7:42-67] "The preferred catalytic metals are Group VIII, noble metals and/or mixtures thereof, or rhenium, more preferably iridium, rhenium, or rhodium, still more preferably rhodium. In addition, the catalyst composition may contain promoters or other secondary metals. Suitable promoters or other secondary metals are well known in the art and are not critical to the invention. Without being limited to any particular promoter or secondary metal, some suitable ones include lanthanides, Group VIII metals and/or any mixtures thereof. The support material may comprise any well known in the art, such as refractory support materials like alumina, titania, zirconia, and the like, preferably alumina. The term porous support material as used herein is meant to include all supports that have a preferred initial surface area of at least 2 m^2/g and preferably at least 5 m^2/g. The support structures may be monoliths, particulates or any other shape or form that can be prepared from the compositions of the present invention. Often, the shape or form selected will dictate the type of catalyst bed that must be used. For example, fixed beds are comprised of monoliths and large particle sized supports (greater than 0.5 mm, preferably greater than about 1 mm). Small-size support particles (less than 0.5 mm, preferably less than 0.25 mm) tend to be more useful in fluidized beds. The more preferred embodiments of the present invention comprise fixed bed reactors having large to medium sized particulate type supports."

EXHIBIT D

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,230,035 ("*Espinoza*")

'864 patent	U.S. Patent No. 7,230,035 ("*Espinoza*")
	[10:22-24] "When the Fischer-Tropsch catalyst is supported, the support comprises generally alumina, titania, silica, zirconia or mixtures thereof." [10:48-56] "The dried supports were then impregnated with an aqueous solution of $RhCl_3 \cdot xH_2O$ and $SmCl_3.6H_2O$ using an appropriate quantity to achieve incipient wetness with the desired loading of Rh and Sm, except for Example B, where the dried alumina was next impregnated with an aqueous solution of $Rh(NO_3)_3$, $ReCl_3$, $ZrO(NO_3)_2...xH_2O$ using an appropriate quantity to achieve incipient wetness with the desired loading of Rh, Re and Zr." [8:39-59] "Methods for the preparation of the catalyst systems described herein are not critical. Any method by which the catalytic metal can be dispersed within the catalyst matrix in accordance with the present invention will be sufficient. For example, the following known techniques have been found to be useful in either blocking the pores of a porous support or preferentially depositing catalytic material near the outer surface of the porous catalyst support: (i) applying a solution of a pore-blocking agent such as silicic acid or sodium carbonate, to the support particle, and calcining the catalyst particle such that a substantial portion of micropores of the particle are filled or blocked; (ii) immersion in water followed by drying at a temperature ranging from about 150°F. to about 300° F. in a microwave oven, or the like, and then calcining in air; (iii) using a fast drying or low vapor pressure solvent during an impregnation step; and (iv) impregnation of 'egg yolks' by selecting an appropriate combination of a metal compound, redox reaction and type of support, e.g., hydroxyl ion-generating redox reaction with an acidic reacting support or a hydrogen ion-generating redox reaction with a basic support."

EXHIBIT D

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,230,035 ("*Espinoza*")

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	[Table 1]

TABLE 1

		Catalyst Compositions		
Cat-a-lyst Ex.	Support	Sodium carbonate used	Loading Rhodium	Promoter(s)
A	Alpha-alumina (Sasol); 5.1 m^2/g; 0.3–0.6 mm granules	3%	0.5%	0.5% Sm
B	Alpha-alumina (Norpor-Saint-Gobain); 4.4. m^2/g; 1 mm granules;	4%	1.0%	0.05% Re, 0.05% Zr
C	Alpha-alumina (Norpor-Saint-Gobain); 4.4. m^2/g; 1 mm granules	4%	0.75%	0.5% Sm
D	Alpha-alumina; 5.1 m^2/g; 1.6 mm extrudate; trilobes	5%	0.5%	0.5% Sm
E	Alpha-alumina (Sasol); 5.1 m^2/g; 0.3–0.6 mm granules	2%	0.5%	0.5% Sm
F	Alpha-alumina (Sasol); 5.1 m^2/g; 0.3–0.6 mm granules	1%	0.5%	0.5% Sm
G	Alpha-alumina (Sasol); 5.1 m^2/g; 0.3–0.6 mm granules	2.5%	0.5%	0.5% Sm
H	Alpha-alumina (Sasol); 5.1 m^2/g; 0.3–0.6 mm granules	1.75%	0.5%	0.5% Sm

EXHIBIT D

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,230,035 ("*Espinoza*")

'864 patent	**U.S. Patent No. 7,230,035 ("*Espinoza*")**
	To the extent that this limitation is not disclosed, either explicitly or inherently, by *Espinoza*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
1[c] one or more Group VIII noble metals distributed in the particle as an eggshell such that a higher concentration of the one or more Group VIII noble metals is present in the outer region of the microsphere sized porous particle as compared to the concentration of the one or more Group VIII noble metals in the centre of the microsphere sized porous particle.	*Espinoza* discloses a combustion promoter comprising one or more Group VIII noble metals, the metals distributed in the particle as an eggshell such that a higher concentration of the one or more Group VIII noble metals is present in the outer region of the microsphere sized porous particle as compared to the concentration of the one or more Group VIII noble metals in the centre of the microsphere sized porous particle. *See, e.g.*,

EXHIBIT D

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,230,035 ("*Espinoza*")

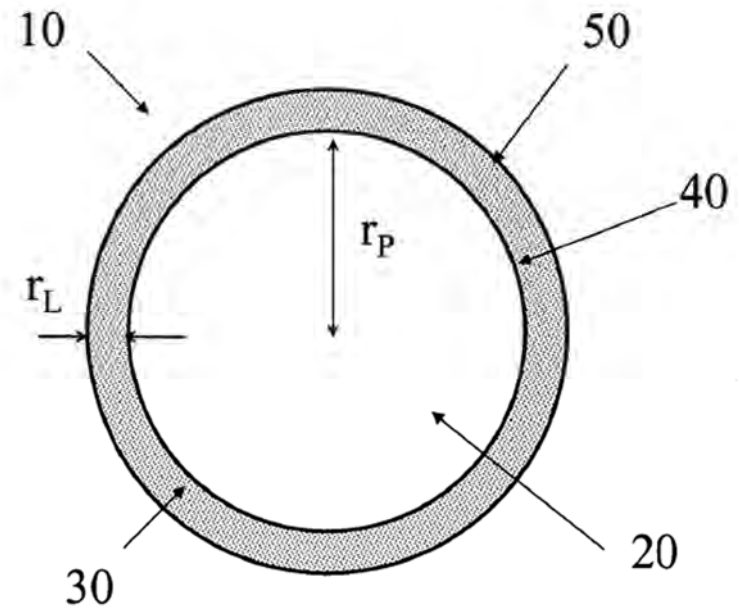
'864 patent	**U.S. Patent No. 7,230,035 ("*Espinoza*")**
	[Fig. 1]  Figure 1

EXHIBIT D

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,230,035 ("*Espinoza*")

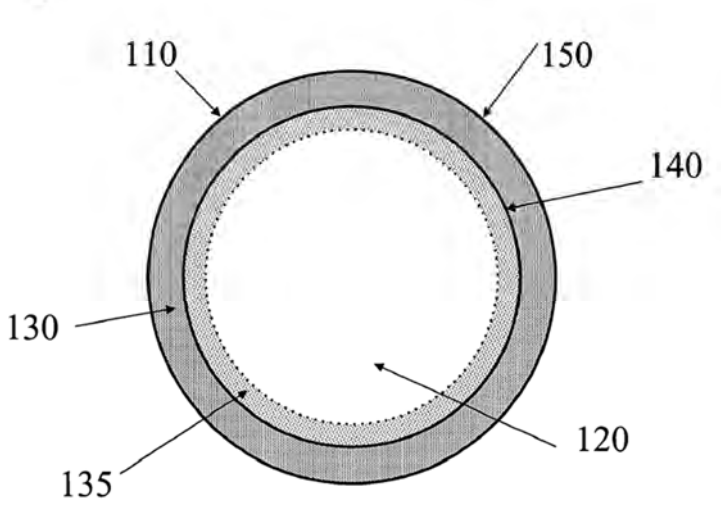
'864 patent	U.S. Patent No. 7,230,035 ("*Espinoza*")
	[Fig. 2]  Figure 2 [Abstract] "The present invention relates to improved catalyst compositions, as well as methods of making and using such compositions to prepare synthesis gas and ultimately C_{5+} hydrocarbons. In particular, preferred embodiments of the present invention comprise catalyst systems comprising a core and an outer region disposed on said core, wherein a substantial amount of the catalytic metal is located in the outer region of the catalyst support matrix. In addition, the catalyst systems are able to maintain high conversion and selectivity values with very low catalytically active metal loadings. The catalyst systems are appropriate for improved syngas, oxidative

EXHIBIT D

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,230,035 ("*Espinoza*")

'864 patent	U.S. Patent No. 7,230,035 ("*Espinoza*")
	dehydrogenation and other partial oxidation reactions, including improved reaction schemes for the conversion of hydrocarbon gas to C_{5+} hydrocarbons." [2:29-36-3:45] "As stated above, these reactions are catalytic reactions and the literature is replete with varying catalyst compositions. The more preferred catalytic metals are Group VIII or noble metals, such as rhodium, platinum, or palladium. These metals are sometimes combined with secondary metals (also called promoter metals) to enhance their activity. Further, the catalyst compositions may include a support material such as alumina, silica, titania, zirconia, and the like. Partial oxidation catalysts are generally prepared by uniformly dispersing one or more catalytic metal throughout a catalyst support matrix. One problem with this type preparation is that some preferred catalytic metals, e.g., Rh, Pt, Pd, are typically expensive. Regardless of how expensive or how high the loading, uniform dispersion inherently exiles a substantial amount of catalytic metal to the deep internal surfaces or 'core' of a support. For purposes of this disclosure 'deep internal surfaces' or 'core' is intended to mean the region of the catalyst structure that is deeper than the smaller of (1) 300 μm as measured from the catalyst exterior surface or (2) the outer 30% of the catalyst volume as measured from the catalyst exterior surface directly inwards toward the center of the structure. It should be appreciated that volume here and further herein is contemplated to include both the solid and pore volumes that make up a particle structure. The catalyst metal exiled in this manner creates at least two disadvantages. First, catalytic metal inside the core region of the catalyst support structure will be underutilized. This is especially true in high space velocity reactors where only a relatively small fraction of the reactant gases are able to reach the core region. A second disadvantage is that the any products formed inside the core region will have to diffuse out of the core and the outer layer surrounding the core. The time it takes for the products formed inside the

EXHIBIT D

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,230,035 ("*Espinoza*")

'864 patent	U.S. Patent No. 7,230,035 ("*Espinoza*")
	core region to leave the catalyst particle is significantly greater relative to the products formed at or near the exterior surface of the catalyst. More specifically, the difference is equal to the added time it takes for the products to diffuse out of the core. Thus, a further disadvantage is that the possibility for undesired side reactions becomes greater because the products and the reactants are exposed to active catalyst material for a longer period of time. Applicants theorize, without wishing to be bound by the theory, that in processes having very high reactant space velocities, such as, for example, the catalytic partial oxidation of methane, the inner part of the catalyst particles (e.g., core) do not significantly contribute to the overall reaction productivity. In fact, Applicants postulate that the undesired secondary reactions are predominant in the inner region. Therefore, the chemical activity in the core of the catalyst particle will have the undesirable effects set forth above such as affecting the H_2:CO molar ratio, lowering the H_2 and CO selectivities, and increasing the reaction temperature. Additionally, the catalytic metal inside the inner core of the catalyst is underutilized with respect to the desired reactions. Therefore, the catalyst metal dispersed in the inner core of the particles contributes significantly to the cost without contributing significantly to the productivity, and indeed potentially decreasing the productivity, of the reactor. Non-porous material (defined as a material with a surface area of less than 1 m^2/g) would typically provide a catalyst core without significant diffusion of reactants/products and thus would overcome the production of undesirable by-products. However, the use of a non-porous support material for the catalyst core is not an acceptable solution, because non-porous materials do not have a sufficient amount of surface area on which to disperse the catalytic material. In addition, lack of sufficient surface area can prevent the reactions from igniting.

EXHIBIT D

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,230,035 ("*Espinoza*")

'864 patent	U.S. Patent No. 7,230,035 ("*Espinoza*")
	Thus, there is a desire to have a syngas production process, which minimizes the amount of expensive catalytic material in the core of catalyst particles and also limits access of the core to the reactants, thus limiting the extent of the occurrence of the undesirable secondary reactions and minimizing the effects of those undesirable reactions. Accordingly, research has focused on developing new catalysts that can reduce or eliminate the problems associated with the prior art catalysts. The approach taken is to more effectively using smaller amounts of catalytic metal or using the catalytic metals more efficiently while maintaining the required minimum amount of surface area. The present invention has been developed with these considerations in mind and is believed to be an improvement over the catalyst systems in the prior art." [3:49-60] "The present invention is directed towards improved catalyst systems, as well as methods of making and using such systems. The present invention provides a syngas catalyst system comprising a core and an outer layer comprising a catalytically active metal, in which effective use of the catalyst metal is enhanced and the occurrence of undesirable secondary reactions is inhibited. The invention achieves this result by minimizing diffusion of the reactant gases into the core of the catalyst particle. Thus, the reactant gases are not allowed to linger in the particle core and react unfavorably, and the expensive catalytically active materials are only placed where their presence is most economically beneficial." [4:5-47] "The preferred embodiments include a partial oxidation catalyst in which wherein at least 60% of the catalytically active metal is located within 300 μm from the catalyst exterior surface. In a more preferred embodiment, the present invention comprises a catalyst having a catalytically active metal load of about 1 wt % or less, preferably of about 0.75 wt% or less, more preferably of about 0.5 wt% or less, on a support

EXHIBIT D

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,230,035 ("*Espinoza*")

'864 patent	U.S. Patent No. 7,230,035 ("*Espinoza*")
	wherein at least 60% of the catalytically active metal is located within 300 μm from the catalyst exterior surface. A particular preferred embodiment comprises a partial oxidation catalyst compositions capable of high methane and carbon monoxide selectivities comprising rhodium as the catalytic metal, with a rhodium loading of about 1 wt % or less, preferably of about 0.75 wt% or less, more preferably of about 0.5 wt % or less. The preferred reaction is partial oxidation of light hydrocarbons, particularly natural gas or methane to form synthesis gas, but the support is equally useful in any oxidative reactions such as conversion of a gaseous alkane to its corresponding olefin. The present invention further relates to methods for preparing catalyst compositions in accordance with the present invention. For example, a method for preparing nonmicroporous catalyst particles, each having a core comprising a refractory support like alumina or zirconia and an outer layer, includes applying a pore blocking material such as silicic acid or sodium carbonate to the support material, so as to substantially block the micropores of the support material and obtain a nonmicroporous core of the catalyst particle. The particles are then provided a catalytically active material for catalyzing reactions, wherein the catalytically active material preferably comprises a Group VIII metal like rhodium and optionally a promoter metal such as a lanthanide metal or rhenium, dispersed in the outer layer, wherein the catalytically active material does not substantially penetrate and deposit into the core because of the blocked pores. Another embodiment involves preparing catalysts using an impregnation technique employing a fast drying or low vapor pressure solvent, which allows preferential deposition of the active material in an outer layer, preferably deposited on the outer surface of the support." [5:35-6:9]

EXHIBIT D

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,230,035 ("*Espinoza*")

'864 patent	U.S. Patent No. 7,230,035 ("*Espinoza*")
	"The present invention relates to improved catalyst systems. The present invention also comprises methods of preparing and using the catalyst systems. The present invention is more preferably directed towards syngas catalysts used in partial oxidation reactions and even more preferably used in syngas catalysts that contain Group VIII or noble metals. However, it should be appreciated that the catalyst systems according to the present invention are useful for other partial oxidation reactions, which are intended to be within the scope of the present invention. In addition, the present invention contemplates an improved method for converting hydrocarbon gas to C_{5+} hydrocarbons using the novel syngas catalyst systems described herein. Thus, the invention also relates to processes for converting hydrocarbon-containing gas to C_{5+} hydrocarbons via an integrated syngas to Fischer-Tropsch process. One embodiment of the present invention is for a catalyst system in which the majority of the catalytic metal is near the surface of the support matrix. In other words, the catalyst is not uniformly dispersed throughout the catalyst matrix, but instead has regions of differing catalyst concentration. For example, as show in FIG. 1, the support could have more than one discrete region comprising differing catalyst concentrations. As shown, the spherical catalyst system 10 has two regions 20 and 30. Region 20 is the inner core, and region 30, also called outer layer, is the area at the periphery of the core. Region 30 is delimited by exterior surface 50 of catalyst system 10 and outer surface 40 of the region 20. In the broadest and simplest terms and in accordance with the present invention, region 20 should have a lower amount of catalyst than region 30. In other words, the catalyst material should be distributed such that if region 20 and 30 were divided into two sections of equal volume, i.e., 50-50, more than 50% of the catalytically active material would be located in region 30. In a preferred embodiment, region 20 would comprise substantially lower amounts of catalyst than region 30. The general objective being that more than 50% of the catalytically active material is located in a contiguous

EXHIBIT D

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,230,035 ("*Espinoza*")

'864 patent	U.S. Patent No. 7,230,035 ("*Espinoza*")
	region of the support that comprises 50% or less of the total support volume. In a more preferred embodiment, region 20 would have substantially zero catalytic material." [6:10-7:67] "In another embodiment of the present invention shown in FIG. 2, a catalyst particle 110 has a core 120 with an outer surface 140, a catalytic outer layer 130 with an exterior surface 150, and a substantially blocked-micropore layer 135. The substantially blocked-micropore layer 135 substantially prevents access to deposition of catalytic material in internal surface of core 120 of the catalyst particle 110. Thus, catalyst material are deposited primarily only on the outer surface 140 of the core 120 and form the outer layer 130. The substantially blocked-micropore layer 135 can be of any thickness, so long as it substantially blocks access of the catalytic material to the core 120 of the catalyst particle 110. It is preferred that the catalytic layer 130 be not more than 300 micron thick, and greater than 0.5 micron thick. Referring now back to FIG. 1, a catalyst particle 10 in accordance with the first embodiment of the present invention includes a catalytic outer layer 30. The particle 10 of FIG. 1 differs from particle 110 of FIG. 2 in that the substantially blocked-micropore layer of particle 10 extends substantially throughout the core 20 of the particle 10. Referring now to FIG. 3, another embodiment of a catalyst particle 210 has a trilobe configuration with a core 220 and an outer layer 230 comprising most of the catalytic material. As in the particle of FIG. 1, the blocked-micropore layer extends through substantially the entire core 220 of the particle 210, such that the catalytic material does not substantially penetrate into the core 220 of the particle 210 during the preparation of the catalyst.

EXHIBIT D

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,230,035 ("*Espinoza*")

'864 patent	U.S. Patent No. 7,230,035 ("*Espinoza*")
	It should be appreciated that discrete regions are difficult to produce in practice and a more practical approach may be to create a gradient of catalyst concentration with the highest concentration near the exterior surface and essentially zero near the center of the core. Again, producing a catalyst in which the majority of active catalyst material is near the exterior surface. It should also be appreciated that although a spherical-shape structure for the support is illustrated in FIGS. 1-2 and a trilobe-shape structure for the support is illustrated in FIG. 3, the present invention is not intended to be so limited and is equally applicable to any shape so long as the catalytic material is predominately in the region that is delimited by the outer surface of the core and the exterior surface of the catalyst. In addition, some combination of discrete and graduated regions could be used. In another embodiment, catalyst systems that have less than 60% of their total volume within 300 μm of the catalyst support outer surface should have at least 60% of the catalytically active metal located within 300 μm of the catalyst exterior surface, preferably wherein about 75% of the catalytically active metal is located within 250 μm of the catalyst exterior surface, and still more preferably 80% of the catalytically active metal is located within 200 μm of the catalyst exterior surface. By locating the catalytic metal near the outer surface of the support, a larger percentage of expensive catalytic metal can be more efficiently used because less of the metal will be exiled to the core and fewer side reactions are possible because fewer undesirable products will be formed in the core. It should be understood that for the present disclosure depositing at or near the outer surface of a support particle may include (1) on its outer surface causing an 'extension' of the surface with continued buildup of catalyst material, (2) on the outer surface and inside the pores and/or pore surfaces such that little buildup, if any, occurs, or (3) some combination of the two, e.g., some deposition within the pores with an average buildup on the outer surface.

EXHIBIT D

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,230,035 ("*Espinoza*")

'864 patent	U.S. Patent No. 7,230,035 ("*Espinoza*")
	In addition, an increase in the amount of catalytic metal that is located near the exterior surface of the catalyst may decrease the overall amount of catalytic metal that is needed to prepare an efficient catalyst system. Thus, another preferred embodiment of the present invention comprises a catalyst system wherein the catalytically active metal, e.g., rhodium, loading is preferably 1 wt % or less, more preferably 0.75 wt% or less and still more preferably 0.5 wt% or less. By using a smaller loading of catalytic metal, the present invention has the advantage of greatly reducing the cost associated with the expensive metals. By combining the above-identified features, the present invention becomes a significant improvement over catalysts with more uniform dispersions of catalytic metal and higher catalyst loadings on the internal surface of a support material. Accordingly, in another preferred embodiment, the catalyst systems of the present invention comprise catalytically active metal loadings of preferably 1 wt % or less, more preferably 0.75 wt% or less and still more preferably 0.5 wt% or less, wherein more than 50% of the catalytically active material is located in a contiguous region of the support that comprises 50% or less of the total support volume and wherein that region contains all of the outer surface of the support. In another embodiment, it is preferred that catalyst systems that have less than 60% of their volume within 300 μm of the catalyst exterior surface comprise catalytically active metal loadings of preferably 1 wt % or less, more preferably 0.75 wt % or less and still more preferable 0.5 wt % or less, wherein at least 60% of the catalytically active metal is located within 300 μm of the catalyst exterior surface, preferably wherein about 75% of the catalytically active metal is located within 250 μm of the catalyst exterior surface, and still more preferably 80% within 200 μm of the catalyst exterior surface.

EXHIBIT D

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,230,035 ("*Espinoza*")

'864 patent	U.S. Patent No. 7,230,035 ("*Espinoza*")
	The preferred catalytic metals are Group VIII, noble metals and/or mixtures thereof, or rhenium, more preferably iridium, rhenium, or rhodium, still more preferably rhodium. In addition, the catalyst composition may contain promoters or other secondary metals. Suitable promoters or other secondary metals are well known in the art and are not critical to the invention. Without being limited to any particular promoter or secondary metal, some suitable ones include lanthanides, Group VIII metals and/or any mixtures thereof. The support material may comprise any well known in the art, such as refractory support materials like alumina, titania, zirconia, and the like, preferably alumina. The term porous support material as used herein is meant to include all supports that have a preferred initial surface area of at least 2 m^2/g and preferably at least 5 m^2/g. The support structures may be monoliths, particulates or any other shape or form that can be prepared from the compositions of the present invention. Often, the shape or form selected will dictate the type of catalyst bed that must be used. For example, fixed beds are comprised of monoliths and large particle sized supports (greater than 0.5 mm, preferably greater than about 1 mm). Small-size support particles (less than 0.5 mm, preferably less than 0.25 mm) tend to be more useful in fluidized beds. The more preferred embodiments of the present invention comprise fixed bed reactors having large to medium sized particulate type supports." [8:9-14] "Syngas reactor 301 houses a plurality of catalyst particles such as those of FIG. 1, FIG. 2, or FIG. 3, wherein the catalytic material does not substantially penetrate into the core of the catalyst particle." [8:39-9:7] "Methods for the preparation of the catalyst systems described herein are not critical. Any method by which the catalytic metal can be dispersed within the catalyst matrix in accordance with the present invention will be sufficient. For

EXHIBIT D

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,230,035 ("*Espinoza*")

'864 patent	U.S. Patent No. 7,230,035 ("*Espinoza*")
	example, the following known techniques have been found to be useful in either blocking the pores of a porous support or preferentially depositing catalytic material near the outer surface of the porous catalyst support: (i) applying a solution of a pore-blocking agent such as silicic acid or sodium carbonate, to the support particle, and calcining the catalyst particle such that a substantial portion of micropores of the particle are filled or blocked; (ii) immersion in water followed by drying at a temperature ranging from about 150° F. to about 300° F. in a microwave oven, or the like, and then calcining in air; (iii) using a fast drying or low vapor pressure solvent during an impregnation step; and (iv) impregnation of 'egg yolks' by selecting an appropriate combination of a metal compound, redox reaction and type of support, e.g., hydroxyl ion-generating redox reaction with an acidic reacting support or a hydrogen ion-generating redox reaction with a basic support. A fast drying solvent may include any solvent with a low vapor pressure (at normal temperature and pressure) provided that the catalytic metal precursors are solvable in that solvent. For example, suitable solvents may include, but are not limited to, alcohols, such as methanol, ethyl alcohol, iso- or n-propanol, butanol and the like, acetone, or toluene. Using a fast drying solvent preferentially deposits catalytic metal precursors on and/or near the outer surface of the support because of the rapid drying of the solvent. The techniques described herein are not exhaustive of techniques useful to the present invention and are merely listed as examples of the preferred techniques. Many others exist, are known or can be created by those of ordinary skill in the art to perform the same function and get the same or similar result." [10:17-26] "Fischer-Tropsch catalysts are well known in the art and generally comprise a catalytically active metal, a promoter and a support structure. The most common catalytic metals used in Fischer-Tropsch catalysts are Group VIII metals, such as

EXHIBIT D

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,230,035 ("*Espinoza*")

'864 patent	U.S. Patent No. 7,230,035 ("*Espinoza*")
	cobalt, nickel, ruthenium, iron or mixtures thereof. When the Fischer-Tropsch catalyst is supported, the support comprises generally alumina, titania, silica, zirconia or mixtures thereof. Fischer-Tropsch reactors use fixed and fluid type conventional catalyst beds as well as slurry bubble columns." [10:40-67] "Catalyst Examples A-H were prepared in accordance with the present invention. Each was prepared using the following general procedure. A selected support material having an appropriate surface area, i.e., at least 2 m^2/g, and preferably at about 4-5 m^2/g, was impregnated with a sodium carbonate solution by incipient wetness. The sodium carbonate solution ranged from 1-5 wt %. The impregnated support materials were then dried in an oven for 3 hours at 150° C. The dried supports were then impregnated with an aqueous solution of $RhCl_3.xH_2O$ and $SmCl_3$. $6H_2O$ using an appropriate quantity to achieve incipient wetness with the desired loading of Rh and Sm, except for Example B, where the dried alumina was next impregnated with an aqueous solution of $Rh(NO_3)_3$, $ReCl_3$, $ZrO(NO_3)_2$... xH_2O using an appropriate quantity to achieve incipient wetness with the desired loading of Rh, Re and Zr. Other promoters can be added in like manner as desired. Because of the presence of the sodium carbonate, the Rh and promoter metals were deposited in the outer radial portion of the support structure. The catalyst compositions were then washed with distilled water to remove sodium. The catalyst samples were then dried in an oven for 2 hours at 120° C. and calcined in air for 2 hours at 800° C. Table 1 lists the type of support, surface area and particle size, as well as the concentration of the sodium carbonate solution used in the preparation of these Examples, and the catalytic material (Rhodium, promoters) loadings."

EXHIBIT D

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,230,035 ("*Espinoza*")

'864 patent	U.S. Patent No. 7,230,035 ("*Espinoza*")
	[Table 1]

TABLE 1

Catalyst Ex.	Support	Sodium carbonate used	Loading: Rhodium	Loading: Promoter(s)
A	Alpha-alumina (Sasol); 5.1 m^2/g; 0.3–0.6 mm granules	3%	0.5%	0.5% Sm
B	Alpha-alumina (Norpor-Saint-Gobain); 4.4. m^2/g; 1 mm granules;	4%	1.0%	0.05% Re, 0.05% Zr
C	Alpha-alumina (Norpor-Saint-Gobain); 4.4. m^2/g; 1 mm granules	4%	0.75%	0.5% Sm
D	Alpha-alumina; 5.1 m^2/g; 1.6 mm extrudate; trilobes	5%	0.5%	0.5% Sm
E	Alpha-alumina (Sasol); 5.1 m^2/g; 0.3–0.6 mm granules	2%	0.5%	0.5% Sm
F	Alpha-alumina (Sasol); 5.1 m^2/g; 0.3–0.6 mm granules	1%	0.5%	0.5% Sm
G	Alpha-alumina (Sasol); 5.1 m^2/g; 0.3–0.6 mm granules	2.5%	0.5%	0.5% Sm
H	Alpha-alumina (Sasol); 5.1 m^2/g; 0.3–0.6 mm granules	1.75%	0.5%	0.5% Sm

EXHIBIT D

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,230,035 ("*Espinoza*")

'864 patent	U.S. Patent No. 7,230,035 ("*Espinoza*")
	[12:1-7] "The data in Table 2 shows that the catalytic performance of the rhodium catalyst systems prepared in accordance with the present invention are comparable to other catalysts in the art with much higher catalytic metal loadings, such as those disclosed in U.S. published patent application 2002/0115730, which is incorporated herein by reference in its entirety." TABLE 2 Reactivity Data of Rhodium Catalysts Catalyst / Flow Rate / Exit Temp (° C.) / CH_4 Conversion (%) / H_2 Selectivity (%) / CO Selectivity (%) / H_2/CO in syngas product A 3500 772 94.9 88.3 96.3 1.83 5000 789 95.2 87.4 96.7 1.81 B 3500 730 93.2 87.0 95.8 1.82 5000 764 93.3 87.8 96.1 1.83 C 3500 778 91.6 94.4 95.8 1.97 5000 817 91.4 90.6 96.3 1.88 D 3500 669 95.0 93.0 93.0 1.93 5000 702 94.9 92.1 92.1 1.91 To the extent that this limitation is not disclosed, either explicitly or inherently, by *Espinoza*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.

EXHIBIT D

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,230,035 ("*Espinoza*")

'864 patent	**U.S. Patent No. 7,230,035 ("*Espinoza*")**
[3] The combustion promoter according to claim 1, comprising a plurality of microsphere sized porous particles comprising alumina.	*Espinoza* discloses a combustion promoter comprising a plurality of microsphere sized porous particles comprising alumina. *See, e.g.*, The support provided above for Claim 1[b]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Espinoza*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[6] The combustion promoter according to claim 1, wherein the microsphere sized porous particles collectively have an average (D50) size of between 60 and 90 microns as measured by laser diffraction.	*Espinoza* discloses a combustion promoter comprising microsphere sized porous particles, wherein the microsphere sized porous particles collectively have an average (D50) size of between 60 and 90 microns as measured by laser diffraction. *See, e.g.*, The support provided above for Claim 1[a]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Espinoza*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.

EXHIBIT D

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 7,230,035 ("*Espinoza*")

'864 patent	**U.S. Patent No. 7,230,035 ("*Espinoza*")**
[12] The combustion promoter according to claim 1, wherein the one or more Group VIII noble metals comprises one or more of platinum, palladium, iridium, ruthenium, and rhodium.	*Espinoza* discloses a combustion promoter comprising one or more Group VIII noble metals, wherein the one or more Group VIII noble metals comprises one or more of platinum, palladium, iridium, ruthenium, and rhodium. *See, e.g.*, The support provided above for Claim 1[c]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Espinoza*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[15] The combustion promoter according to claim 12, wherein the one or more Group VIII noble metals comprises palladium.	*Espinoza* discloses a combustion promoter comprising one or more Group VIII noble metals, wherein the one or more Group VIII noble metals comprises palladium. *See, e.g.*, The support provided above for Claim 1[c]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Espinoza*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.

EXHIBIT E

EXHIBIT E

Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

"*Optimal Distribution of Catalyst in Pellet*" ("*Gavriilidis Article*"), authored by Asterios Gavriilidis et al. and published in 1993, qualifies as prior art of U.S. Patent No. 11,224,864 ("the '864 patent") under at least 35 U.S.C. §§ 102 and/or 103 for the asserted claims of the '864 patent, as discussed in detail in the claim chart below.

The Court has not yet construed the claims and therefore the meaning of the terms in the claims has yet to be resolved by the Court. The support identified here for these limitations is responsive to Plaintiffs' infringement contentions, which Defendant disagrees with. As such, nothing in Defendant's claim charts should be construed as an admission regarding infringement, either literally or under the doctrine of equivalents, or as an admission regarding Defendant's understanding of the proper scope of the asserted claims. Defendant reserves the right to rely on additional citations or sources of evidence that also may be applicable, or that may become applicable in light of claim construction, changes in Plaintiffs' infringement contentions, and/or information obtained during discovery as the case progresses.

To the extent Plaintiffs allege that *Gavriilidis Article* does not disclose any particular limitation of the asserted claims of the '864 patent, either expressly or inherently, it would have been obvious to a person of ordinary skill in the art as of the priority date of the '864 patent to modify *Gavriilidis Article* and/or to combine the teachings of *Gavriilidis Article* with other prior art references, including but not limited to the present prior art references cited in the Cover Pleading and the relevant section(s) of claim charts for other prior art references for the '864 patent in a manner that would have rendered the asserted claims invalid as obvious.

Defendant reserves the right to amend or supplement this claim chart at a later date as more fully set forth in the Cover Pleading.

EXHIBIT E

Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

'864 patent	***"Optimal Distribution of Catalyst in Pellets" ("Gavriilidis Article")***
1[a] A CO to CO_2 combustion promoter comprising microsphere sized porous particles, each microsphere sized porous particle having a diameter of less than 1mm, and independently comprising	*Gavriilidis Article* discloses a CO to CO_2 combustion promoter comprising microsphere sized porous particles, each microsphere sized porous particle having a diameter of less than 1mm, and independently comprising the limitations discussed in the following sections. *See, e.g.*, [Page 400] "A large fraction of the chemical and refinery processes are catalytic in nature. While the worldwide sales of catalysts are only about $4 billion annually, the economic impact of catalysis comes from the fact that approximately $200 worth of products are manufactured for every $1 worth of catalyst consumed [1]. The active materials used as catalysts are often expensive metals, and in order to be utilized effectively, they are deposited on high surface area supports. This approach in many cases introduces intrapellet activity gradients during the preparation process, which were traditionally thought to be detrimental to catalyst performance. However, the effects of deliberate nonuniform distribution of the catalytic material within the support on the performance of a catalyst pellet started receiving attention in the late 1960's (see Refs. 2-6). These, as well as later studies, both experimental and theoretical, demonstrated that nonuniformly distributed catalysts can offer superior conversion, selectivity, durability, and thermal sensitivity characteristics over those wherein the activity is uniform." [Page 401-402] "In early studies, step-distributions of catalyst were analyzed for the simple case of a single reaction occurring under isothermal conditions. Researchers often treated bimolecular Langmuir- Hinshelwood kinetics, since it exhibits a maximum in the reaction rate as a function of reactant concentration. Thus, there is a range of reactant concentrations, where the reaction rate *increases* as the reactant concentration decreases. This feature occurs, for example, in the case of carbon monoxide or hydrocarbon oxidation, in excess oxygen, over noble metal catalysts such as platinum or palladium [22]."

EXHIBIT E

Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

'864 patent	***"Optimal Distribution of Catalyst in Pellets"* (*"Gavriilidis Article"*)**
	[Page 416] "The dimensionless steady-state mass balance of each component, and energy balance in a porous catalyst pellet, accounting for external film transport resistances, are given by…" [Page 422-423] "On Pt/γ-Al_2O_3 catalysts prepared by coimpregnation techniques, Chemburkar studied carbon monoxide oxidation [43], and Masi et al. studied ethylene hydrogenation [44]. Steady-state multiplicity behavior was observed in both studies. Additionally, it was shown theoretically that the catalyst performance was very sensitive to even small catalytic activity present in the portion of the support surrounding the active layer [43] (see Fig. 16). Similarly, Harold and Luss [45] demonstrated theoretically that multiplicity features for carbon monoxide oxidation are altered significantly even by small variations in the activity profile of an egg-shell-type catalyst." [Page 424] "One of the earliest works in this area was the modeling of CO oxidation in monolith reactors for automobile converters [24]. The CO conversion to CO_2 as a function of the Thiele modulus (or equivalently, the catalyst temperature) for a fixed inlet CO concentration is shown in Fig. 19, for four different catalyst distributions (cf. Fig. 1). The conversion of CO below 648°F was significantly higher in the inner catalyst, while above 648°F the middle catalyst attained 100% conversion. The uniform and outer catalysts also showed 100% conversion at progressively higher temperatures."

EXHIBIT E

Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

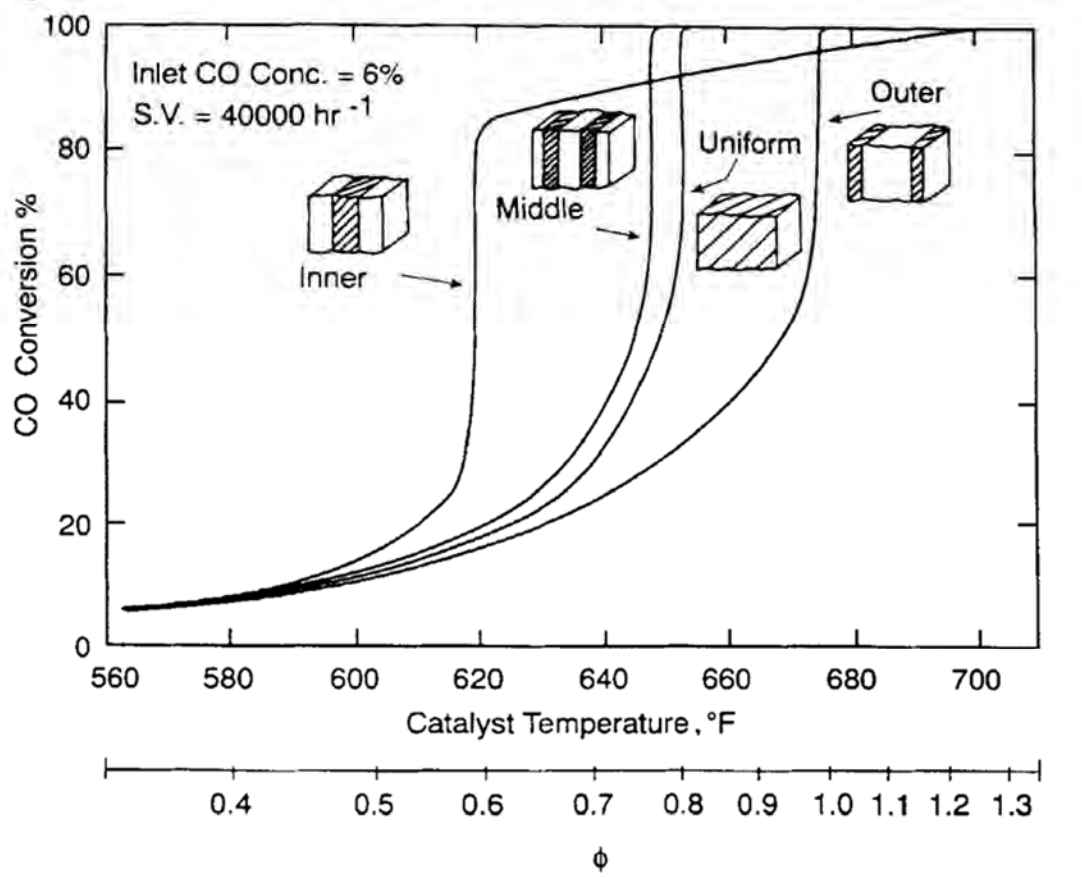
'864 patent	***"Optimal Distribution of Catalyst in Pellets" ("Gavriilidis Article")***
	[Fig. 19]  FIG. 19. Carbon monoxide conversion to CO_2 over a monolith reactor with platinum catalyst [24].

EXHIBIT E

Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

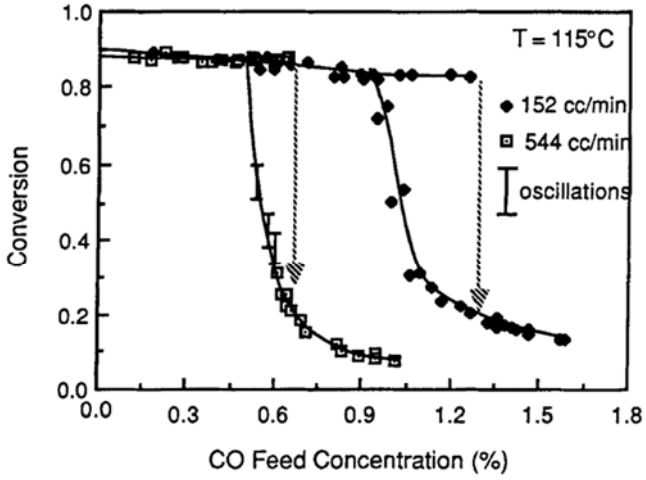
'864 patent	***"Optimal Distribution of Catalyst in Pellets"* (*"Gavriilidis Article"*)**
	[Page 427-428] "Propane oxidation was investigated on Pt/γ-Al_2O_3 catalyst by Kunimori et al. in a fixed-bed reactor under isothermal conditions [54]. They found that the activity was as follows: egg-shell > uniform > egg-white, which is expected for first-order kinetics. However in poisoning experiments, the egg-white catalyst was found to be more resistant which was reasonable for pore-mouth poisoning. The same investigators also studied CO oxidation on the same catalyst. As noted earlier, this reaction exhibits bimolecular Langmuir-Hinshelwood type of kinetics and egg-white catalysts are expected to be more active. Indeed, it was found that the activity was in the order: egg-white > uniform > egg-shell. Examining the steady-state multiplicity behavior of the above system, they found that the width of the conversion versus temperature hysteresis loop was much larger for the egg- white than for the egg-shell catalyst [55]. It is interesting to note that no difference in activity was observed between the powdered catalysts, prepared by pulverizing the egg-white and egg-shell catalyst pellets.  FIG. 22. Experimental results for a single zone fixed-bed reactor at various feed flowrates; $\bar{s} = 0.27$. Reaction: CO oxidation on Pt/γ-Al_2O_3 catalyst [56].

EXHIBIT E

Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

'864 patent	***"Optimal Distribution of Catalyst in Pellets"* (*"Gavriilidis Article"*)**
	Lee and Varma [56] studied experimentally the behavior of an isothermal fixed-bed reactor, where CO oxidation occurs on four different step-type $Pt/\gamma\text{-}Al_2O_3$ catalysts (with $\bar{s}$ = 0.27, 0.50, 0.65, 1), in which the thickness of the catalyst band did not exceed 8% of the pellet radius. Two steady states were observed in a reactor packed with one type of catalyst as shown in Fig. 22. Oscillations were observed frequently just before the transition from the low-conversion regime to the high-conversion regime. The reactor packed with the catalyst where the active layer was positioned at $\bar{s}$ = 0.65 had the widest range of high-conversion regime, among the four types of catalysts studied. This range was further increased by using a dual-zone reactor, with $\bar{s}$ = 0.27 and $\bar{s}$ = 1 in the first and second zones, respectively. This improvement was expected according to the discussion in the previous section. A direct experimental test of the impact of non-uniform catalyst distribution was demonstrated in a dual-zone reactor. Using the *same* catalyst pellets, the reactor performance (including the number of steady states) was dramatically altered when the packing order was first $\bar{s}$ = 0.27 and seconds = 1 in the two zones, and then it was reversed, as shown in Fig. 23. All of the above observations were in excellent qualitative agreement with theoretical predictions."

EXHIBIT E

Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

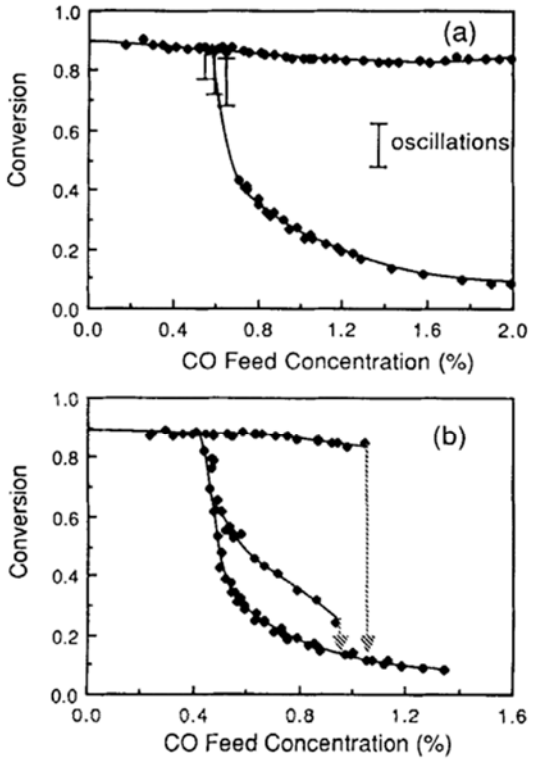
'864 patent	***"Optimal Distribution of Catalyst in Pellets" ("Gavriilidis Article")***
	 FIG. 23. Experimental results for a dual-zone fixed-bed reactor at 100°C and 152 cc/min flow rate. Reaction: CO oxidation on Pt/γ-Al_2O_3 catalyst. (a) $\bar{s}_1$ = 0.27, $\bar{s}_2$ = 1; (b) $\bar{s}_1$ = 1, $\bar{s}_2$ = 0.27 [56]. [Page 429-430] "Becker and Wei [58] studied the case of a first-order reaction with nonselective poisoning. They assumed that the poison adsorbs on the entire catalyst surface with no preference for the active element or the support (i.e., nonselective poisoning) and without ever reaching a saturation condition. The diffusion-reaction process for the poison is governed by the poison Thiele modulus defined as

EXHIBIT E

Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

'864 patent	***"Optimal Distribution of Catalyst in Pellets" ("Gavriilidis Article")***
	$\Phi_p = R\sqrt{k_p/D_{e,p}}$ (37) which represents the ratio between the rate of poison deposition (assumed to be first-order with respect to poison concentration) and the rate of the poison diffusion inside the pellet. It should be noted that in the above analysis all diffusion coefficients were assumed constant, which is true for situations where the poison loading is not sufficiently large to significantly plug the catalyst pore mouth. Depending upon the value of the poison Thiele modulus, two characteristic situations are obtained. For $\Phi_p << 1$, the poison deposits uniformly throughout the pellet (i.e., *uniform poisoning),* while for $\Phi_p >> 1$ the poison deposits preferentially in a thin layer close to the pellet's external surface (i.e., *pore-mouth poisoning).* In the analysis of Becker and Wei [58], four types of active element distributions in the pellet were considered: uniform, egg-shell, egg-white, and egg-yolk. In the case of uniform poisoning the best performance is provided by an egg-shell distribution, similarly to the nondeactivating case for positive-order reactions (Section II.A. I .a). However, under pore-mouth poisoning conditions a significant improvement in the catalyst performance could be achieved by distributing the active element in the inner region of the pellet. Similar improvement was also demonstrated by Polomski and Wolf [59] for egg-yolk catalysts, in which the outer protective layer was assumed to be comprised of a different support material than the support over which the catalyst was deposited, therefore having different diffusion properties." [Page 433] "In automotive emission control, a catalyst is required which simultaneously catalyzes three reactions, that is, oxidation of carbon monoxide and residual hydrocarbons, as well as reduction of nitrogen oxides, in a range of values of the air/fuel ratio centered around the stoichiometric point. This is a rather ambitious goal which needs to simultaneously utilize various active elements to perform the different required functions. Also, except under startup conditions when the catalyst is cold, under normal operating conditions the catalyst is at a sufficiently high temperature that the reactions are diffusion controlled, and for this reason the active elements are deposited close to the external surface of the pellet. However, a

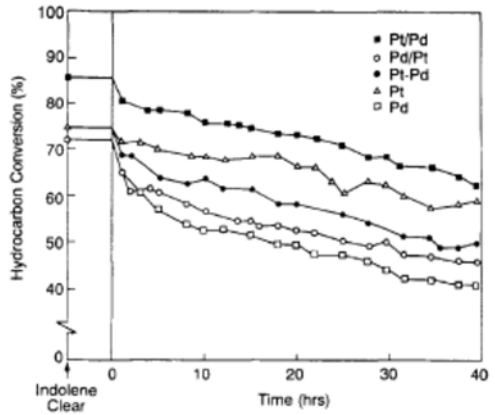
'864 patent	***"Optimal Distribution of Catalyst in Pellets" ("Gavriilidis Article")***
	complication arises from the occurrence of two deactivating processes: poisoning due to the presence of trace quantities of lead and phosphorous in the feed, and thermal sintering." [433-434] "The poisoning process is nonselective; that is, it involves both the support as well as the active catalyst. Since the poisoning process is diffusion controlled (i.e., pore-mouth poisoning), the poison penetrates the catalyst pellet in the form of a sharp front, whose velocity is substantially independent of the local concentration of the active elements. Summers and Hegedus [68] treated the case where only the two oxidizing reactions occur, and comparing several distributions of Pt and Pd, concluded that the best performance is obtained by a catalyst pellet which is impregnated with Pt at the outer shell, and Pd in a separate subsurface band, as illustrated in Fig. 26. This was attributed to the fact that Pt is more resistant to poisoning, with a nonzero residual activity, while Pd located in the interior provides higher hydrocarbon oxidation activity. The above configuration also guarantees a certain resistance to sintering, which is provided by the Pd band protected from poisoning by the external Pt band."  FIG. 26. Hydrocarbon conversion with time, during accelerated poisoning experiments. Pt/Pd: Pt (exterior)/Pd (interior); Pd/Pt: Pd (exterior)/Pt (interior); Pt–Pd: Pt and Pd (both exterior); Pt: Pt (exterior); Pd: Pd (exterior) [68].

EXHIBIT E

Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

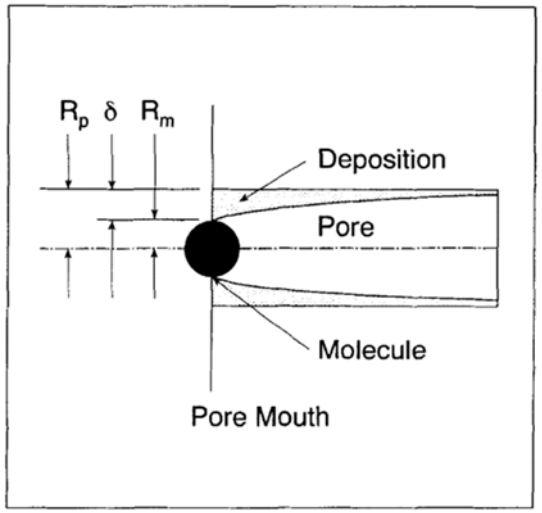
'864 patent	***"Optimal Distribution of Catalyst in Pellets"* (*"Gavriilidis Article"*)**
	[435-436] "Metal deposition usually occurs first at the pore mouth, which prevents the entrance of other reactant molecules, thus leaving a substantial fraction of the catalyst in the internal part of the pellet unutilized. This process is illustrated schematically in Fig. 27, where it is shown that when the thickness of the deposit equals the difference between the pore radius, RP and the reactant molecule radius, *Rm* then the transport through the pore mouth is inhibited and the pellet becomes inactive. For first-order deposition kinetics, the time *t*, at which such pore blocking occurs, can be estimated from the relationship $t = \rho_d(R_p - R_m)/kC$ (40) where ρ_d is the deposited layer density, *C* is the reactant concentration, and *k* is the rate constant of the deposition process.  FIG. 27. Schematic representation of pore mouth plugging [70].

EXHIBIT E

Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

'864 patent	***"Optimal Distribution of Catalyst in Pellets" ("Gavriilidis Article")***
	In order to use the catalyst more effectively, various strategies have been explored, involving optimization of the pore-size distribution and of the catalyst shape. In a detailed theoretical study, Limbach and Wei [70] investigated the use of catalyst pellets with nonuniform activity distributions. The case of deposition rate following first-order kinetics was considered. It was assumed that the intrinsic reaction rate per unit active surface area is not affected by deposition. The change of active surface area with deposition was accounted for by using an empirical relationship and similarly, the decrease of the effective diffusion coefficient with deposition was also taken into account because, as illustrated in Fig. 27, the deposited metal decreases the cross-section area of the pores available for transport. It was demonstrated that for hydrodemetallation, the overall deposited amount in a uniform pellet could be increased by up to 25% by simply decreasing the amount of catalyst located near the pore mouth, so as to delay its complete blockage. The optimal catalyst activity distributions were also computed numerically, and these exhibit a sharp maximum toward the center of the pellet followed by a decrease toward the external surface, so that the front of the fully deactivated catalyst originates at the center of the pellet and moves to the external surface. This distribution provides a good utilization of the catalyst, maximizing the overall amount of metals deposited." [Page 436-437] "Composite catalysts consist of small particles of the active catalyst (e.g., zeolite crystals) embedded in a support (e.g., silica-alumina) which may exhibit some modest catalytic activity itself. The support has much larger pores and therefore the diffusion coefficient is much larger in the support than in the active catalyst. As illustrated by Ruckenstein [72], this is the main advantage of composite catalysts, since the reactants have a much easier access to the active catalyst particles than in the case where the entire pellet is comprised of the active catalyst alone. The advantages of these catalytic systems in the presence of deactivation were studied by Varghese and Wolf [73] with reference to uniformly distributed catalyst pellets and pore-

EXHIBIT E

Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

'864 patent	***"Optimal Distribution of Catalyst in Pellets" ("Gavriilidis Article")***
	mouth poisoning. In this case the complete plugging of the larger pores of the support is significantly delayed relative to the case of a pellet which contains the active catalyst alone. Accordingly, the catalyst life is longer and allows for a better utilization of the active catalyst located in the inner portion of the pellet." [Page 439-440] "Nonuniform impregnation profiles usually arise due to complex interaction of intrapellet flow, diffusion, adsorption, electrochemical, and reaction phenomena, depending upon the particular system and the impregnation process employed. Several models reviewed by Lee and Aris [21] have been proposed to explain the distribution of the active ingredients in catalyst supports. Two rate processes are involved in the impregnation step: the transport of solute in the pores of the support and the uptake of solute by the pore walls. In most of the models developed so far, Fickian diffusion is assumed to take place in the liquid-filled pores, either during wet impregnation or in dry impregnation after the initial convective transport has ceased. Different variations of Langmuir-type adsorption (e.g., reversible or irreversible, kinetic or equilibrium, one or more types of adsorption *sites)* have been considered for the uptake of the solute on the support. The following studies are representative of those which have appeared in the literature. Vincent and Merrill used a simple model of plug-flow into a single pore to analyze the time-dependent flow of impregnating solution into a dry pellet and interior dispersal of the impregnate [96]. Two types of resistances at the liquid-solid interface were considered: mass transfer and adsorption kinetics; and these were shown to yield similar impregnation profiles in systems where saturation occurred. For the limiting cases where mass transfer or adsorption controls, Lee [97] developed analytical expressions for the intrapellet catalyst concentration at the end of the filling period." [Page 442-443] "The catalyst precursor deposited on the surface of the pores of a pellet is comprised of two parts: The first is the solute adsorbed on the support during the impregnation process, and the

EXHIBIT E

Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

’864 patent	***“Optimal Distribution of Catalyst in Pellets”* (*“Gavriilidis Article”*)**
	second is from the precipitation of unadsorbed solute from the pore liquid during the drying process. Depending upon the transport and adsorption characteristics of the system, catalyst deposition by adsorption can yield uniform or nonuniform distributions.” [Page 443-444] “The drying step which follows the impregnation phase, causes the transfer and evaporation of the liquid in the pore volume and in the process redistributes the impregnant remaining in the pore volume. The extent of this redistribution effect on the impregnation profile depends upon the physicochemical parameters of the system and in certain cases can be significant [116-119]. This process is also affected by the viscosity of the liquid in the pores [120]. The effect of drying is indeed complex, and differing conclusions about the consequences of slow and fast drying have been reported in the literature (cf. Ref. 21).”

EXHIBIT E

Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

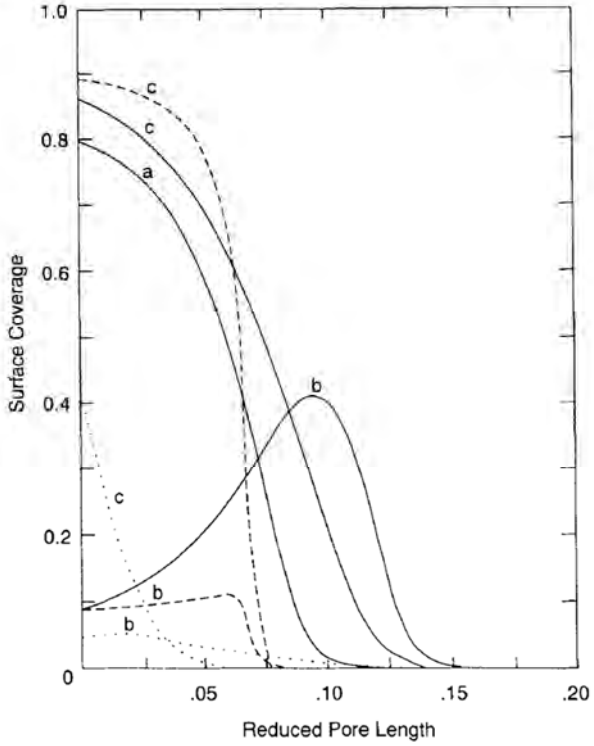
'864 patent	***"Optimal Distribution of Catalyst in Pellets"* (*"Gavriilidis Article"*)**
	[Fig. 29]  FIG. 29. Effect of coimpregnation on platinum distribution profile with citrate ion as coimpregnant: ——, electrokinetic and ionic effects considered; ····, electrokinetic effects neglected; – – –, electrokinetic and ionic dissociation effects neglected. (a) Adsorbed impregnant in the absence of coimpregnation; (b) adsorbed impregnant during coimpregnation; (c) adsorbed coimpregnant. Chloroplatinic acid 0.5×10^{-3} M, sodium citrate 0.5×10^{-3} M, $pH_0 = 3.3$, time = 10 h, $I_0 = 10^{-3}$ M [107]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Gavriilidis Article*, this limitation is obvious in combination with the knowledge of one of skill in the art

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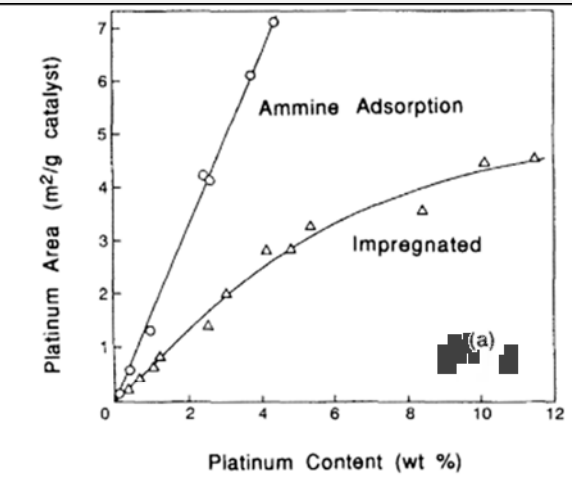
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'864 patent	***"Optimal Distribution of Catalyst in Pellets"* ("*Gavriilidis Article*")**
	and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
1[b] silica, alumina, or mixtures thereof; and	*Gavriilidis Article* discloses a combustion promoter comprising silica, alumina, or mixtures thereof. *See, e.g.*, [Page 418-419] "If the active element surface area is proportional to its loading, then the assumption of linear dependence of the reaction rate to the loading is valid. However, this is not always the case, and nonlinear dependence of surface area to loading is generally observed, especially at high loadings. This relationship is shown in Fig. 3, for the systems of Pt/SiO_2 [19] and $Ag/\alpha\text{-}Al_2O_3$ [40]."

EXHIBIT E

Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

'864 patent	***"Optimal Distribution of Catalyst in Pellets" ("Gavriilidis Article")***



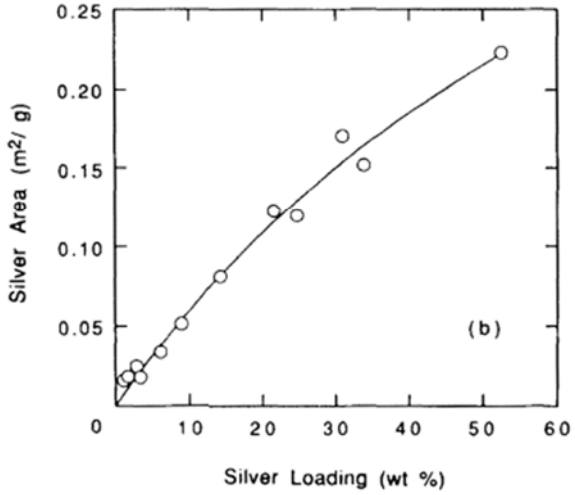


FIG. 13. (a) Variation of platinum area in impregnated- and ammine adsorption-type catalysts with platinum content for Pt/SiO_2 catalysts [19]. (b) Silver area as a function of silver loading for Ag/α-Al_2O_3 catalysts [40].

EXHIBIT E

Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

'864 patent	***"Optimal Distribution of Catalyst in Pellets"* (*"Gavriilidis Article"*)**
	[Page 421] "For a nonisothermal first-order reaction, the optimal catalyst distribution is obtained numerically; this is shown in Fig. 15. It broadens from a step-distribution to a more uniform one as *B* increases from 0.001 to 0.1, as expected according to the discussion above. It should be noted that such values of *B* for the Pt/SiO_2 system imply volume average loadings of platinum of 0.01 to l weight%, and thus cover the range of interest in most practical applications." [Page 422-423] "Kotter and Riekert investigated the triangular reaction network of partial oxidation of propene to acrolein on $CuO/\alpha\text{-}Al_2O_3$ [41, 42]. Three types of catalyst were prepared, having approximately the same amount of CuO, which was deposited in uniform, egg-shell, and egg-yolk distributions. They found that the selectivity to acrolein was higher for the egg-shell catalyst and progressively lower in the uniform and egg-yolk catalysts, in good agreement with theoretical predictions for the above reaction network. On $Pt/\gamma\text{-}Al_2O_3$ catalysts prepared by coimpregnation techniques, Chemburkar studied carbon monoxide oxidation [43], and Masi et al. studied ethylene hydrogenation [44]. Steady-state multiplicity behavior was observed in both studies. Additionally, it was shown theoretically that the catalyst performance was very sensitive to even small catalytic activity present in the portion of the support surrounding the active layer [43] (see Fig. 16). Similarly, Harold and Luss [45] demonstrated theoretically that multiplicity features for carbon monoxide oxidation are altered significantly even by small variations in the activity profile of an egg-shell-type catalyst. Wu et al. studied the hydrogenation of ethylene on $Pd/\gamma\text{-}Al_2O_3$ which follows first-order kinetics, and the methanation of CO on $Ni/\gamma\text{-}Al_2O_3$ which follows bimolecular Langmuir-Hinshelwood type of kinetics, in a single- pellet reactor [46]. The pellets were prepared by pressing the catalyst be- tween two alumina layers, so that step-type catalysts were

EXHIBIT E

Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

'864 patent	***"Optimal Distribution of Catalyst in Pellets" ("Gavriilidis Article")***
	produced. The investigators found for both reactions that there exists an optimal location for the active layer within the pellet (see Fig. 17), and that the optimal location moves toward the surface as the Thiele modulus increases, according to theoretical predictions. Gavriilidis and Varma studied the ethylene epoxidation reaction net- work on Ag/α-Al_2O_3 step catalyst in a novel single-pellet reactor with external recycle [47]. As shown in Fig. 18, the selectivity to ethylene oxide is higher when the active material is located at the external surface of the pellet, in qualitative agreement with previous theoretical results [37, 38]." [Page 427] "Propane oxidation was investigated on Pt/γ-Al_2O_3 catalyst by Kunimori et al. in a fixed-bed reactor under isothermal conditions [54]. They found that the activity was as follows: egg-shell > uniform > egg-white, which is expected for first-order kinetics." [Page 427-428] "Lee and Varma [56] studied experimentally the behavior of an isothermal fixed-bed reactor, where CO oxidation occurs on four different step-type Pt /γ-Al_2O_3 catalysts (with $\bar{s}$ = 0.27, 0.50, 0.65, 1), in which the thickness of the catalyst band did not exceed 8% of the pellet radius."

EXHIBIT E

Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

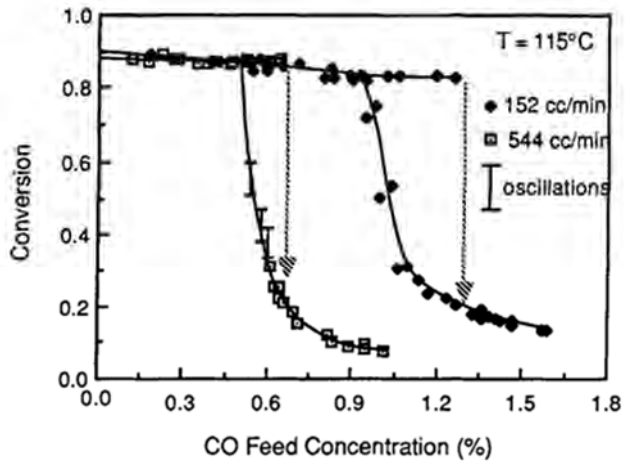
'864 patent	***"Optimal Distribution of Catalyst in Pellets" ("Gavriilidis Article")***
	[Fig. 22]  FIG. 22. Experimental results for a single zone fixed-bed reactor at various feed flowrates; $s = 0.27$. Reaction: CO oxidation on $Pt/\gamma\text{-}Al_2O_3$ catalyst [56].

EXHIBIT E

Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

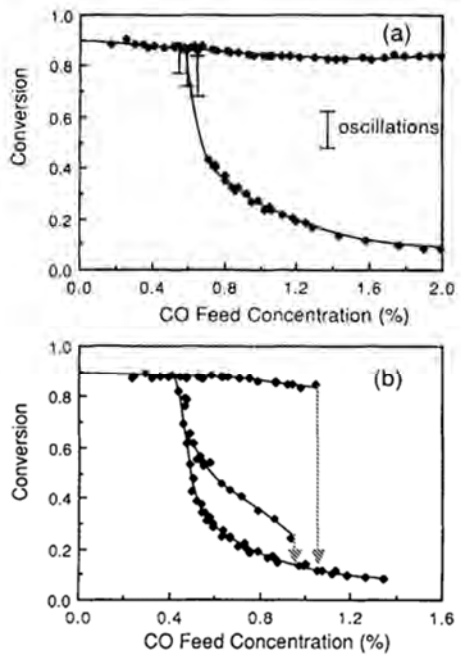
'864 patent	***"Optimal Distribution of Catalyst in Pellets" ("Gavriilidis Article")***
	[Fig. 23]  FIG. 23. Experimental results for a dual-zone fixed-bed reactor at 100°C and 152 cc/min flow rate. Reaction: CO oxidation on Pt/γ-Al_2O_3 catalyst. (a) $\bar{s}_1$ = 0.27, $\bar{s}_2$ = 1; (b) $\bar{s}_1$ = 1, $\bar{s}_2$ = 0.27 [56]. [Page 429] "Incineration of VOCs was recently investigated by Frost et al. [57] in a nonisothermal fixed-bed reactor with axial mass and heat dispersion, and external mass and heat transfer resistances, loaded with Pt/γ-Al_2O_3 pellets where the platinum was distributed as a thin layer slightly below the external surface."

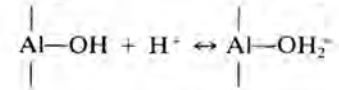
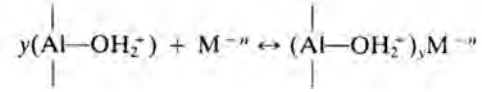
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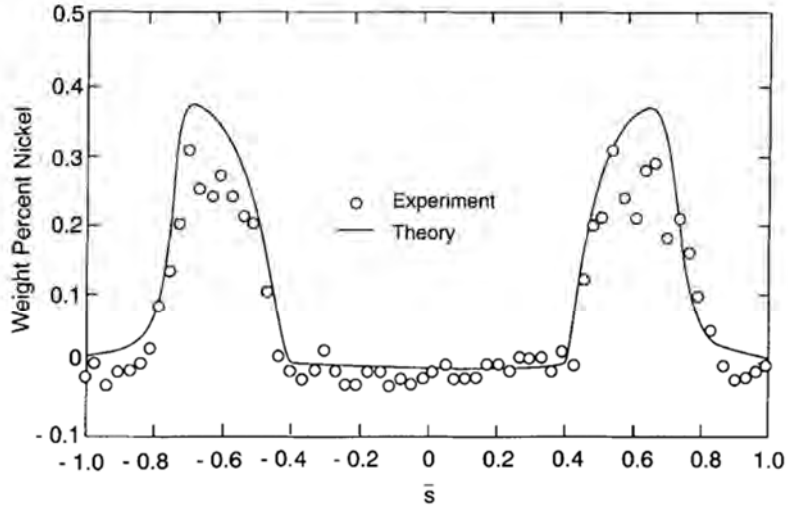
Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

'864 patent	***"Optimal Distribution of Catalyst in Pellets"* (*"Gavriilidis Article"*)**
	[Page 433] "An attempt to experimentally verify the improved performance of nonuniform catalyst pellets in the case of deactivating systems is reported by Rice and Hahn [66]. Three catalyst pellets (nickel supported on alumina) were tested: uniform, egg-shell, and egg-yolk. The methanation reaction $CO + 3H_2 \rightarrow CH_4 + H_2O$ (39) was studied, under deactivation resulting from the formation of carbonaceous compounds as well as from poisoning by hydrogen sulfide, added in small concentrations in the feed stream." [Page 436-437] "Composite catalysts consist of small particles of the active catalyst (e.g., zeolite crystals) embedded in a support (e.g., silica-alumina) which may exhibit some modest catalytic activity itself. The support has much larger pores and therefore the diffusion coefficient is much larger in the support than in the active catalyst. As illustrated by Ruckenstein [72], this is the main advantage of composite catalysts, since the reactants have a much easier access to the active catalyst particles than in the case where the entire pellet is comprised of the active catalyst alone." [Page 437] "The possibility of further improving the pellet performance through a nonuniform distribution of the active catalyst particles within the support was theoretically investigated by Dadyburjor [74]. In particular, a zeolite/silica-alumina cracking catalyst was examined." [Page 438-439] "For alumina, Jiratova [80] found isoelectric points in the range 4.5 to 9.7, depending on the type of foreign ions present on the surface. Among the common support materials zeolites, silica-aluminas and silicas adsorb cat- ions; aluminas, titanias, and chromia are amphoteric (i.e., they adsorb anions in acidic and cations in alkaline solutions); and magnesia adsorbs anions.

EXHIBIT E

Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

’864 patent	*“Optimal Distribution of Catalyst in Pellets” (“Gavriilidis Article”)*
	Although each individual adsorption process may be different, general rules of anionic adsorption have been demonstrated in a study by D'Aniello [81], on the adsorption of transition metal oxalate and cyanide complexes on a -y-alumina surface. The results have been interpreted with an adsorption mechanism of the type   involving simultaneous coadsorption of protons and anions. In the absence of chemical reactions the amounts of anion adsorbed could be easily controlled by the amount of acid present in the impregnating solution. However, not all ion-exchange processes follow the simple scheme outlined above; some involve a series of sequential reactions. For example, the adsorption of H_2PtCl_6 on γ-Al_2O_3 has been associated with an acid attack of the alumina with dissolution of aluminum and subsequent formation of aluminum ions. These ions are finally readsorbed on the support together with the hexachloroplatinic anion [82]. Adsorption characteristics of other systems that have been investigated in detail include calcium [83]; nickel [84]; chromium, cobalt, molybdenum, tungsten [85, 86]; and aromatic acids [87] on γ-alumina. Theoretical approaches of phenomena occurring at the oxide-water interface have been developed largely in the colloid and interface science literature. Based on the electric double-layer theory, James and Healy [88] modeled the adsorption of hydrolyzable metal ions on oxide surfaces. Their calculations were based on the total free energy of adsorption being comprised of a coulombic attraction, representing the energy change favorable for adsorption and a solvation term, representing an energy barrier to ad- sorption. However, the model required the use of an adjustable parameter which was associated with chemical interaction forces. This model has been employed for fitting experimental data for cobalt, calcium,

'864 patent	***"Optimal Distribution of Catalyst in Pellets" ("Gavriilidis Article")***
	chromium, and iron adsorption on silica [89], nickel adsorption on silica [90], and the adsorption of ammonium complexes of copper, nickel, and cobalt on alumina, titania, and iron oxide [91]." [Fig. 30]  FIG. 30. Nickel profile in a γ-alumina pellet after 26.75 h. Impregnation with 10^{-3} M $NiCl_2$ and 0.1 M NaCl after acid pretreatment for 120 h, pH_x = 5.2 [108]. [Page 440-441] "Hegedus et al. modeled competitive multicomponent adsorption on wet pellets by using adsorption constants determined from separate experiments [103]. The model was used to predict the uptake of Rh in γ-Al_2O_3 pellets, as well as the Rh distribution inside the pellet, when HF was used as the site-blocking agent, by using the diffusion constants as adjustable parameters. Scelza et al. [104], using a similar model, fitted the data of Castro et al. [105], for

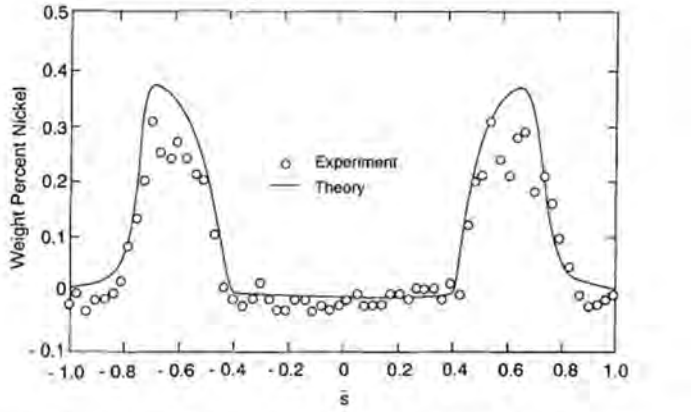
'864 patent	***"Optimal Distribution of Catalyst in Pellets" ("Gavriilidis Article")***
	coimpregnation of pellets with H_2PtCl_6 and HCl. However, not only the diffusion constants but also the adsorption constants were treated as adjustable parameters. Price [106], using a purely competitive Langmuir adsorption model for coimpregnation of chloroplatinic and citric acids, was able to successfully predict the location of the platinum layer inside a γ-Al_2O_3 pellet. For the case of sequential impregnation, the solution effect introduced by the competitive adsorbate was considered, and the resulting model agreed well with experimental results, as illustrated in Fig. 28." [Page 441] "Chu et al. [108] developed a transport-adsorption model for wet impregnation based on the continuity equations for dilute ionic species and on triple-layer electrostatic theory. This model predicted the Ni profile in γ- Al_2O_3 pellets very well, obtained with a system of $NiCl_2$, NaCl, and HCI, as shown in Fig. 30."  FIG. 30. Nickel profile in a γ-alumina pellet after 26.75 h. Impregnation with 10^{-3} M $NiCl_2$ and 0.1 M NaCl after acid pretreatment for 120 h, pH_{ss} = 5.2 [108].

EXHIBIT E

Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

'864 patent	***"Optimal Distribution of Catalyst in Pellets"* (*"Gavriilidis Article"*)**
	[Page 444] "Most multicomponent impregnation studies concern platinum supported on a -y-Al2O3 pellet. Shyr and Ernst [100] studied the influence of a large number of acids and salts used as coimpregnants with H_2PtCl_6, on the distribution of Pt within γ- Al_2O_3 pellets, and obtained profiles shown in Fig. 31. If along with chloroplatinic acid, HCl is used as coimpregnant, uniform platinum distribution is obtained [121, 122]. When dibasic organic acids (e.g., citric or oxalic acid) are used instead, the platinum is driven into the pellet interior forming a subsurface layer with controlled location and width [2, 123]. Employing HF as coimpregnant, Hegedus et al. [103] prepared egg-white Rh/γ- Al_2O_3 catalysts. Increase of the impregnation time has been found to increase the local loading, the width, and the distance of the active layer from the external surface, while increase of the cairn- pregnant concentration pushes the active layer deeper [103, 123, 124]. When the pellet is impregnated with the catalyst precursor and competitive ad- sorbate sequentially, in this or the reverse order, similar egg-white catalysts are obtained. The width of the catalytic layer is decreased if the metal precursor is impregnated first, while if the impregnation order is reversed, the resulting catalyst has a thicker active layer [124]. Some platinum catalysts, prepared by using sequential impregnation of chloroplatinic and citric acids, are shown in Fig. 32; theoretical predictions of catalyst locations for types (a)-(c) were previously shown in Fig. 28." To the extent that this limitation is not disclosed, either explicitly or inherently, by *Gavriilidis Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
1[c] one or more Group VIII noble metals distributed in the particle as an eggshell such that a higher concentration of the	*Gavriilidis Article* discloses a combustion promoter comprising one or more Group VIII noble metals, the metals distributed in the particle as an eggshell such that a higher concentration of the one or more Group VIII noble metals is present in the outer region of the microsphere

EXHIBIT E

Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

'864 patent	***"Optimal Distribution of Catalyst in Pellets"* ("*Gavriilidis Article*")**
one or more Group VIII noble metals is present in the outer region of the microsphere sized porous particle as compared to the concentration of the one or more Group VIII noble metals in the centre of the microsphere sized porous particle.	sized porous particle as compared to the concentration of the one or more Group VIII noble metals in the centre of the microsphere sized porous particle. *See, e.g.*, [Page 401-402] "In early studies, step-distributions of catalyst were analyzed for the simple case of a single reaction occurring under isothermal conditions. Researchers often treated bimolecular Langmuir- Hinshelwood kinetics, since it exhibits a maximum in the reaction rate as a function of reactant concentration. Thus, there is a range of reactant concentrations, where the reaction rate *increases* as the reactant concentration *decreases.* This feature occurs, for example, in the case of carbon monoxide or hydrocarbon oxidation, in excess oxygen, over noble metal catalysts such as platinum or palladium [22]. Becker and Wei [23, 24] numerically analyzed the effects of different catalyst distributions. In three of these (inner, middle, and outer; or alternatively, egg-yolk, egg-white, and egg-shell, respectively), the catalyst was deposited in only one third of the pellet, while in the fourth it was uniformly distributed [24]. The results are shown in Fig. 1, where the catalyst activity, represented by the effectiveness factor, η, is shown as a function of the Thiele modulus, ϕ. It may be seen that among these specific distributions, for low values of ϕ (i.e., kinetic control), the inner distribution is best, while for large values of ϕ (i.e., diffusion control), the inner distribution is best, while for large values of ϕ (i.e., diffusion control), the outer distribution is best. For intermediate values of the Thiele modulus, the middle distribution has the highest effectiveness factor." [Page 404] "From the above equations, it is seen that the catalyst location depends upon the physicochemical parameters of the system, that is, the Thiele modulus ϕ_0 and the adsorption constant σ. By increasing the Thiele modulus, or decreasing the adsorption constant, the optimal location of the active catalyst moves from the interior of the pellet to the external surface."

EXHIBIT E

Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

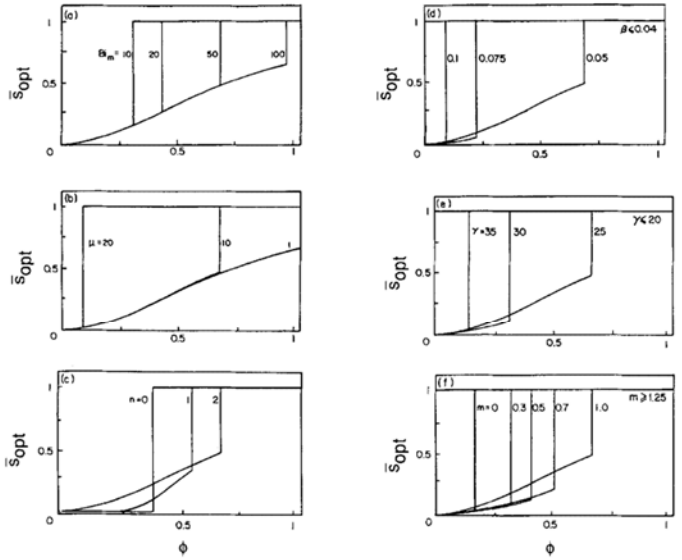
'864 patent	***"Optimal Distribution of Catalyst in Pellets"* (*"Gavriilidis Article"*)**
	[Page 407] "According to the effects of qi_0 and *a* described earlier, as β increases the optimal location eventually approaches the external surface of the pellet. For small values of *a*, the optimal location exhibits a minimum as a function of β, as illustrated in Fig. 5. Note that σ = 0 corresponds to a first-order reaction. In this case, under *isothermal* conditions, the reaction rate would be maximized at a location where the reactant concentration is the highest, that is, at the external surface of the pellet." [Page 408-409] "The effect of external transport resistances is illustrated in Figs. 6(a) and 6(b). As the resistance increase, that is, as the mass or heat Biot numbers decrease, the optimal location moves outward."  FIG. 6. Optimal location of active catalyst, $\bar{s}_{opt}$, as a function of Thiele modulus, ϕ, in the case of an *m*-th order reaction. $Bi_m = 50$, $\mu = 10$, $n = 2$, $\beta = 0.05$, $\gamma = 25$, $m = 1$, unless otherwise specified [32].

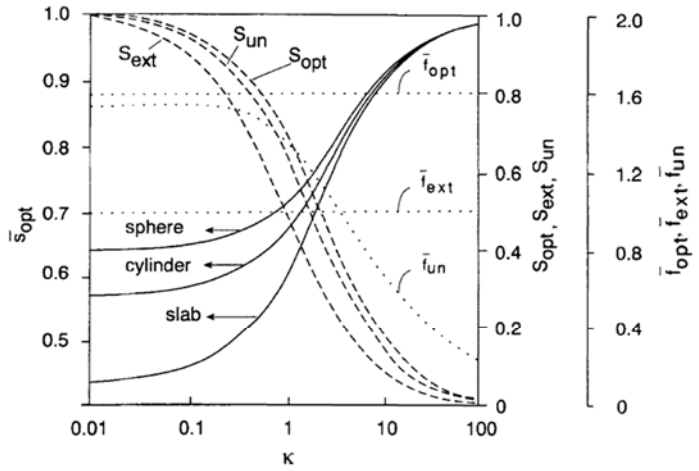
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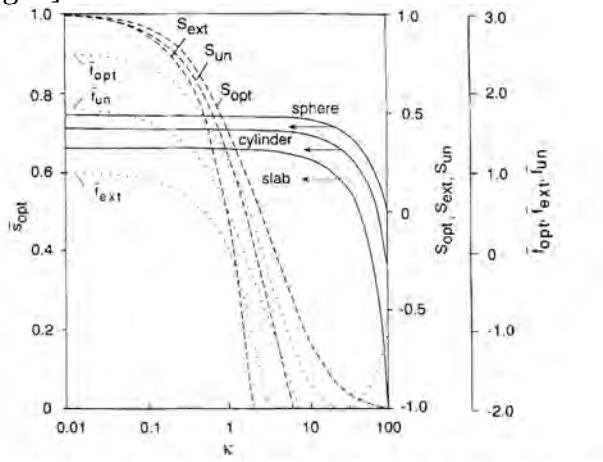
Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

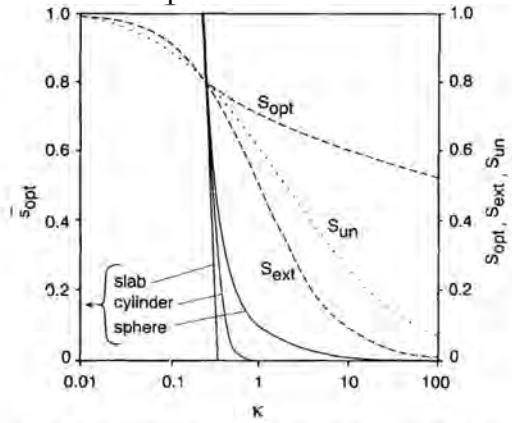
'864 patent	***"Optimal Distribution of Catalyst in Pellets"* (*"Gavriilidis Article"*)**
	[Page 409-411] "For *parallel* reactions that follow power-law kinetics, the optimal location of the Dirac-delta is at the surface of the pellet, if the order of the desired reaction is higher than the order of the undesired one; in the opposite case it is at the center of the pellet. Figure 8 shows the dependence of the optimum catalyst location, selectivity and dimensionless production rate of the desired product on the ratio of Thiele moduli, κ, for the optimum Dirac, uniform and external shell catalyst distributions, when the desired reaction follows bimolecular Langmuir-Hinshelwood kinetics, the undesired reaction follows unimolecular Langmuir-Hinshelwood kinetics and they both involve different adsorption sites. When the undesired reaction becomes faster (i.e., κ increases), the selectivities for all three types of distributions decrease. However, the Dirac-type catalyst always gives the highest selectivity. Simultaneously, the location of the Dirac distribution moves toward the external surface of the pellet. The net rate of production for the desired product is also consistently the highest for the Dirac distribution. The sensitivity of the selectivity to catalyst location behaves qualitatively in the same manner as already discussed for the effectiveness factor in the case of a single reaction."

EXHIBIT E

Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

’864 patent	***“Optimal Distribution of Catalyst in Pellets” (“Gavriilidis Article”)***
	 FIG. 8. Effect of the ratio of Thiele moduli, κ, on the optimal catalyst location, $\bar{s}_{opt}$ (solid lines), global selectivity, S (dashed lines), and dimensionless rate of production of the desired product A_2, $\bar{f}$ (dotted lines), for the optimal distribution (S_{opt}, $\bar{f}_{opt}$), uniform distribution of slab geometry (S_{un}, $\bar{f}_{un}$) and external shell distribution (S_{ext}, $\bar{f}_{ext}$). Parallel reactions: $A_1 \xrightarrow{1} A_2$, $A_1 \xrightarrow{2} A_3$; $f_1(u_1) = (1 + \sigma_1)^2 u_1/(1 + \sigma_1 u_1)^2$, $f_2(u_1) = (1 + \sigma_2)u_1/(1 + \sigma_2 u_1)$, $\sigma_1 = 5$, $\sigma_2 = 20$, $\phi_1\sqrt{(n + 1)/g_v} = 1$, $\xi = 1$ [34]. [Page 411] “The underlying reason for improved performance of Dirac catalysts, as demonstrated in previous sections, is the appropriate manipulation of concentration gradients which develop within the pellet. When detrimental, they are avoided by positioning the catalyst on the surface of the pellet. However, when they are beneficial, one can take advantage of them by placing the catalyst at an appropriate location inside the pellet. Temperature gradients are also manipulated in a similar way. However, their effect is generally more pronounced because of the exponential dependence of reaction rates on temperature. Similarly to the isothermal case, parallel, consecutive, and also triangular reaction net- works, occurring in nonisothermal pellets, show optimum performance when the catalyst is located at a specific position which

'864 patent	***"Optimal Distribution of Catalyst in Pellets"* (*"Gavriilidis Article"*)**
	depends upon the physicochemical parameters of the system as discussed by Vayenas and coworkers [35-37]. In this case, the activation energies and heats of reactions are also involved." [Fig. 9]  FIG. 9. Effect of the ratio of Thiele moduli, κ, on the optimal catalyst location, s_{opt} (solid lines), global selectivity, S (dashed lines), and dimensionless rate of production of the desired product A_2, $\bar{f}$ (dotted lines), for the optimal distribution (S_{opt}, $\bar{f}_{opt}$), uniform distribution for slab geometry (S_{un}, $\bar{f}_{un}$), and external shell distribution (S_{ext}, $\bar{f}_{ext}$). Consecutive reactions: $A_1 \xrightarrow{1} A_2 \xrightarrow{2} A_3$; $f_1(u_1) = (1 + \sigma)^2 u_1/(1 + \sigma u_1)^2$, $f_2(u_2) = u_2$, $\sigma = 8$, $\phi_1\sqrt{(n + 1)/g_s} = 1$, $\xi = 1$, $u_{1,2} = 1$ [34]. [Page 412-413] "As an example consider two parallel reactions, where the desired one is exothermic, has low activation energy, and exhibits second-order kinetics; while the undesired one is endothermic, has high activation energy, and exhibits linear kinetics. The effect of ratio of Thiele moduli, κ, on the optimum location and the selectivity, where the latter is to be optimized, is shown in Fig. 10. In order to obtain a physical understanding of this system, it should be realized that increasing reactant concentration and decreasing temperature favor the desired reaction. For small K values, the rate of the undesired reaction is relatively small and the desired exothermic

'864 patent	***"Optimal Distribution of Catalyst in Pellets"* (*"Gavriilidis Article"*)**
	reaction creates higher temperatures inside the pellet, if the catalyst is located there. This, however, has a detrimental effect on selectivity because of the relative activation energies, which adds to the detrimental concentration effect. Therefore, the optimum catalyst location is at the surface of the pellet."  FIG. 10. Effect of the ratio of Thiele moduli, κ, on the optimal catalyst location, $\bar{s}_{opt}$ (solid lines), optimal global selectivity, S_{opt}, and global selectivity obtained with uniform, S_{un}, and external shell, S_{ext} catalyst distributions. Parallel reactions: $A_1 \xrightarrow{1} A_2$, $A_1 \xrightarrow{2} A_3$; desired product A_2; $f_1(u_1) = u_1^2$, $f_2(u_1) = u_1$, $\beta_1 = 0.1$, $\beta_2 = -0.7$, $\gamma_1 = 5$, $\gamma_2 = 25$, $\phi_1\sqrt{(n+1)/g_v} = 1$ [35]. [Page 413-414] "A system of multiple reactions of great industrial importance, the ethylene epoxidation reaction network, was investigated in detail by Morbidelli et al. [38]. Ethylene reacts over silver catalyst to give ethylene oxide, which is the desired product, and carbon dioxide, which is the undesired by-product. Ethylene oxide is also combusted, but this reaction is not as important as the combustion of ethylene, and so it was not included in this study. It was shown that the selectivity to ethylene oxide is maximized when the catalyst is located at the external surface of the pellet. This is expected for parallel exothermic reactions with similar kinetics when the undesired reaction has a higher activation energy than the desired one. However, if the net production rate of ethylene oxide is to be maximized, then subsurface locations are

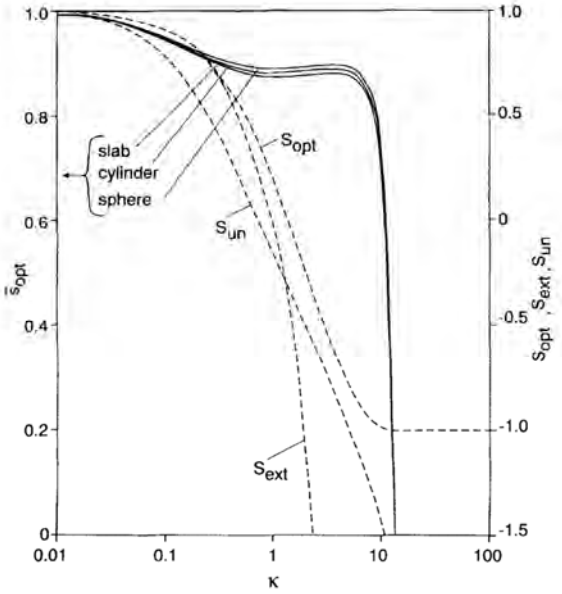
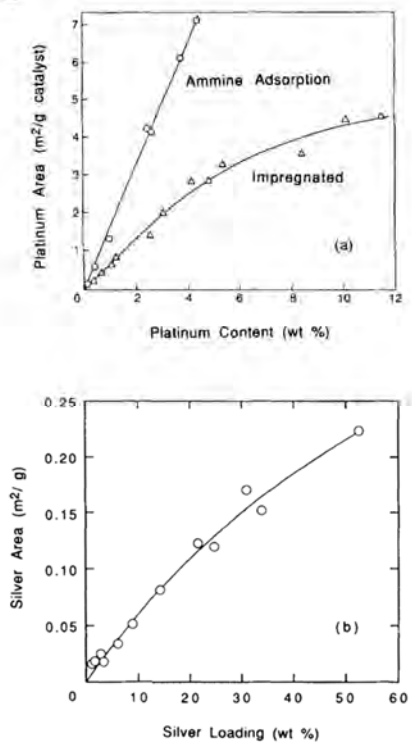
'864 patent	***"Optimal Distribution of Catalyst in Pellets"* (*"Gavriilidis Article"*)**
	optimal, a fact which arises from the nonisothermality of the pellet. The maximum selectivity is attained at the surface of the pellet, even if combustion of ethylene oxide is included [37]." [Fig. 11]  FIG. 11. Effect of the ratio of Thiele moduli, κ, on the optimal catalyst location, s_{opt} (solid lines), optimal global selectivity, S_{opt}, and global selectivity obtained with uniform, S_{un}, and external shell, S_{ext} catalyst distributions. Consecutive reactions: $A_1 \xrightarrow{1} A_2 \xrightarrow{2} A_3$; desired product A_2; $f_1(u_1) = u_1$, $f_2(u_2) = u_2$, $\beta_1 = 0.4$, $\beta_2 = 0.1$, $\gamma_1 = 10$, $\gamma_2 = 5$, $\phi_1\sqrt{(n+1)/g\nu} = 1$, $\xi = 1$, $u_{f,2} = 1$ [36]. [Page 417] "The solution of equations (25) and (26) can be obtained for various catalyst locations between the center and the external surface of the pellet (i.e., $\bar{s}$ = 0-1), and the performance index of interest (i.e., effectiveness factor, selectivity, or yield) can then easily be calculated using equations (22)—(24)."

EXHIBIT E

Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

'864 patent	***"Optimal Distribution of Catalyst in Pellets" ("Gavriilidis Article")***
	[Fig. 13]  FIG. 13. (a) Variation of platinum area in impregnated- and ammine adsorption-type catalysts with platinum content for Pt/SiO_2 catalysts [19]. (b) Silver area as a function of silver loading for Ag/α-Al_2O_3 catalysts [40]. [Page 420-421] "On the other hand, when $\psi \rightarrow \infty$, the optimal distribution becomes a progressively steeper monotonically increasing function of *s*, exhibiting its maxi- mum value at $s = 1$. This occurs when either $B \rightarrow 0$ or $\phi \rightarrow \infty$. For $B = 0$, since there is no problem of loss of catalyst surface area with loading, it is better to locate all the catalyst where the reactant concentration is maximum, that is, at the external surface of the pellet. For increasing values of ϕ, no matter

'864 patent	***"Optimal Distribution of Catalyst in Pellets"* (*"Gavriilidis Article"*)**
	how large B is, the reaction takes place in a narrower region near the pellet external surface, which is the only place where the active element can be profitably located." [Page 422-423] "Kotter and Riekert investigated the triangular reaction network of partial oxidation of propene to acrolein on $CuO/\alpha\text{-}Al_2O_3$ [41, 42]. Three types of catalyst were prepared, having approximately the same amount of CuO, which was deposited in uniform, egg-shell, and egg-yolk distributions. They found that the selectivity to acrolein was higher for the egg-shell catalyst and progressively lower in the uniform and egg-yolk catalysts, in good agreement with theoretical predictions for the above reaction network. On $Pt/\gamma\text{-}Al_2O_3$ catalysts prepared by coimpregnation techniques, Chemburkar studied carbon monoxide oxidation [43], and Masi et al. studied ethylene hydrogenation [44]. Steady-state multiplicity behavior was observed in both studies. Additionally, it was shown theoretically that the catalyst performance was very sensitive to even small catalytic activity present in the portion of the support surrounding the active layer [43] (see Fig. 16). Similarly, Harold and Luss [45] demonstrated theoretically that multiplicity features for carbon monoxide oxidation are altered significantly even by small variations in the activity profile of an egg-shell-type catalyst. Wu et al. studied the hydrogenation of ethylene on $Pd/\gamma\text{-}Al_2O_3$ which follows first-order kinetics, and the methanation of CO on $Ni/\gamma\text{-}Al_2O_3$ which follows bimolecular Langmuir-Hinshelwood type of kinetics, in a single-pellet reactor [46]. The pellets were prepared by pressing the catalyst between two alumina layers, so that step-type catalysts were produced. The investigators found for both reactions that there exists an optimal location for the active layer within the pellet (see Fig. 17), and that the optimal location moves toward the surface as the Thiele modulus increases, according to theoretical predictions. Gavriilidis and Varma studied the ethylene epoxidation reaction network on $Ag/\alpha\text{-}Al_2O_3$ step catalyst in a novel single-pellet reactor with external recycle [47]. As shown in Fig. 18, the

EXHIBIT E

Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

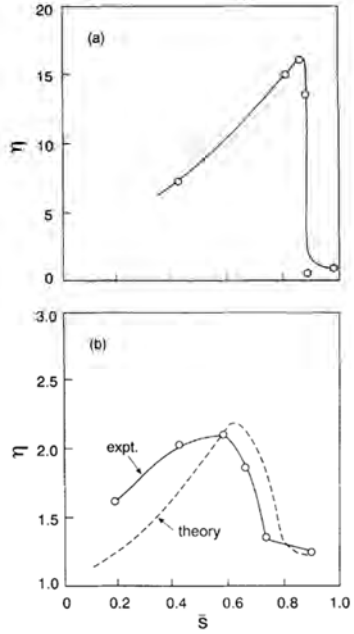
'864 patent	***"Optimal Distribution of Catalyst in Pellets" ("Gavriilidis Article")***
	selectivity to ethylene oxide is higher when the active material is located at the external surface of the pellet, in qualitative agreement with previous theoretical results [37, 38]."  FIG. 17. (a) Plot of experimental effectiveness factor, η, vs. active layer location $\bar{s}$, for ethylene hydrogenation: temperature 298 K, hydrogen concentration 5.89%, $\Delta = 0.05$. (b) Measured and simulant (dotted line) dependences of effectiveness factor, η, on the active layer location, $\bar{s}$, for CO methanation: temperature 533 K, CO concentration 5.0×10^{-6} mol/mL, $H_2/CO = 7.85$ [46].

EXHIBIT E

Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

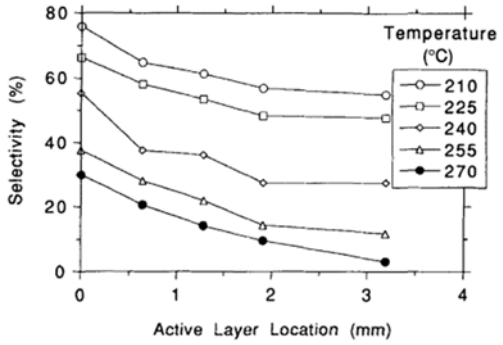
'864 patent	***"Optimal Distribution of Catalyst in Pellets"* (*"Gavriilidis Article"*)**
	 FIG. 18. Selectivity to ethylene oxide as a function of active layer location, for various bulk fluid temperatures; inlet ethylene concentration 7.2% [47]. [Page 424] "One of the earliest works in this area was the modeling of CO oxidation in monolith reactors for automobile converters [24]. The CO conversion to CO_2 as a function of the Thiele modulus (or equivalently, the catalyst temperature) for a fixed inlet CO concentration is shown in Fig. 19, for four different catalyst distributions (cf. Fig. 1). The conversion of CO below 648°F was significantly higher in the inner catalyst, while above 648°F the middle catalyst attained 100% conversion. The uniform and outer catalysts also showed 100% conversion at progressively higher temperatures."

EXHIBIT E

Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

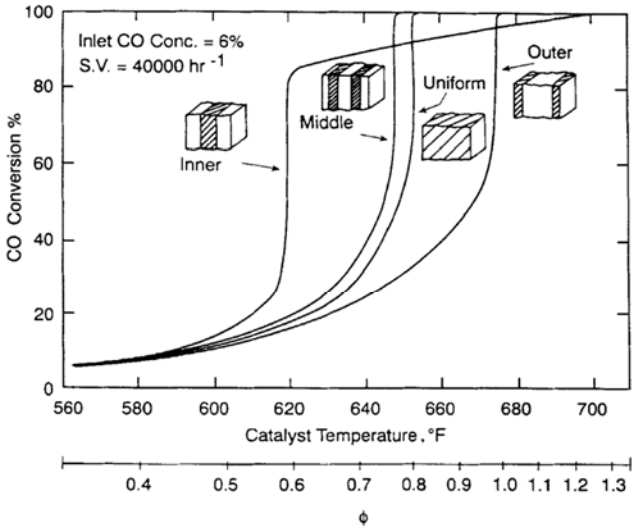
'864 patent	***"Optimal Distribution of Catalyst in Pellets" ("Gavriilidis Article")***
	 FIG. 19. Carbon monoxide conversion to CO_2 over a monolith reactor with platinum catalyst [24]. [Page 427-428] "Propane oxidation was investigated on Pt/γ-Al_2O_3 catalyst by Kunimori et al. in a fixed-bed reactor under isothermal conditions [54]. They found that the activity was as follows: egg-shell > uniform > egg-white, which is expected for first-order kinetics. However in poisoning experiments, the egg-white catalyst was found to be more resistant which was reasonable for pore-mouth poisoning. The same investigators also studied CO oxidation on the same catalyst. As noted earlier, this reaction exhibits bimolecular Langmuir-Hinshelwood type of kinetics and egg-white catalysts are expected to be more active. Indeed, it was found that the activity was in the order: egg-white > uniform > egg-shell. Examining the steady-state multiplicity behavior of the above system, they found that the width of the conversion versus temperature hysteresis loop was much larger for the egg- white than for the egg-shell catalyst [55]. It is interesting to note that no difference in activity was observed between the powdered catalysts, prepared by pulverizing the egg-white and egg-shell catalyst pellets.

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Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

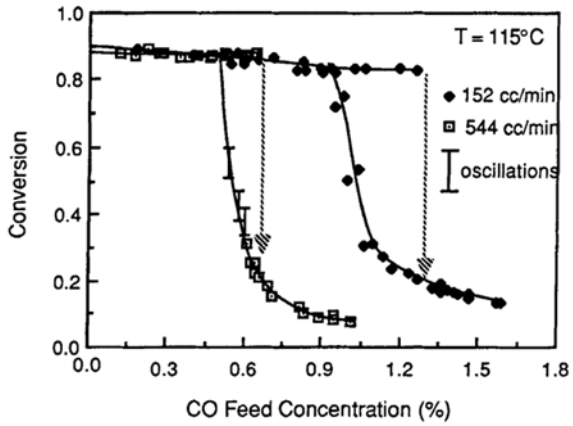
'864 patent	***"Optimal Distribution of Catalyst in Pellets" ("Gavriilidis Article")***
	 FIG. 22. Experimental results for a single zone fixed-bed reactor at various feed flowrates; $\bar{s} = 0.27$. Reaction: CO oxidation on Pt/γ-Al_2O_3 catalyst [56]. Lee and Varma [56] studied experimentally the behavior of an isothermal fixed-bed reactor, where CO oxidation occurs on four different step-type Pt/γ-Al_2O_3 catalysts (with $\bar{s}$ = 0.27, 0.50, 0.65, 1), in which the thickness of the catalyst band did not exceed 8% of the pellet radius." [Page 429] "Incineration of VOCs was recently investigated by Frost et al. [57] in a nonisothermal fixed-bed reactor with axial mass and heat dispersion, and external mass and heat transfer resistances, loaded with Pt/γ-Al_2O_3 pellets where the platinum was distributed as a thin layer slightly below the external surface. The reaction was considered to follow Langmuir-Hinshelwood type of kinetics. The authors developed a model which was fitted to experimental temperature-conversion data by optimizing the values of the kinetic parameters and the effective diffusivity of reactants in the pellet. Once these constants were determined, design parameters for commercial reactors could be obtained."

EXHIBIT E

Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

'864 patent	***"Optimal Distribution of Catalyst in Pellets"* (*"Gavriilidis Article"*)**
	[Page 430] "In the analysis of Becker and Wei [58], four types of active element distributions in the pellet were considered: uniform, egg-shell, egg-white, and egg-yolk. In the case of uniform poisoning the best performance is provided by an egg-shell distribution, similarly to the nondeactivating case for positive-order reactions (Section II.A. I .a). However, under pore-mouth poisoning conditions a significant improvement in the catalyst performance could be achieved by distributing the active element in the inner region of the pellet. Similar improvement was also demonstrated by Polomski and Wolf [59] for egg-yolk catalysts, in which the outer protective layer was assumed to be comprised of a different support material than the support over which the catalyst was deposited, therefore having different diffusion properties. Pavlou and Vayenas [60] addressed the problem of optimal initial activity distribution which maximizes the total reactant conversion accounting for saturation of active sites with poison. In this study, which was limited to the specific case of first-order kinetics and large Thiele modulus values, the deactivation process was simulated by a shell progressive model where the deactivation front, s_p, advances towards the pellet interior independently of the concentrations of the catalyst, Reactants, or poison, according to the Voorhies equation: $S_p = 1 - t^w$ (38) where t is dimensionless time and *w* is an empirical coefficient between 0.5 and 1. The obtained results indicate that in general, the optimal initial activity distribution tends to exhibit a bimodal behavior, concentrating one portion of the active element near the pellet external surface and another portion close to its center, as illustrated in Fig. 24. The specific shape depends on the pellet geometry, on the values of the physicochemical parameters involved, and on the kinetics of the deactivating process through the value of the parameter *w*. It should be noted that in this particular case, contrary to nondeactivating systems, it was found that the performance of the optimal catalyst pellet was not significantly higher than that of a uniformly active pellet."

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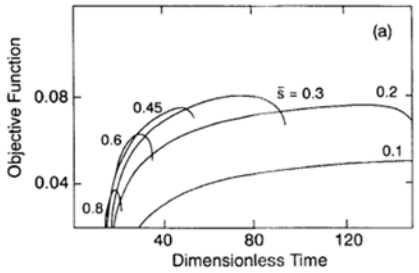
Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

'864 patent	***"Optimal Distribution of Catalyst in Pellets"* (*"Gavriilidis Article"*)**
	[Page 431] "It was found that for low values of the ratio of the two Thiele moduli characteristic of the poisoning and the main reactions, the optimal location is close to the pellet external surface. For increasing values of this parameter, the optimal location moves towards the pellet interior." [Page 432] "It was found that the desired catalyst lifetime plays an important role. In general, for shorter catalyst lifetimes, locations closer to the external surface show superior performance, while for longer lifetimes, deeper lo- cations are better. Also, when the catalyst cost is high [see Fig. 25(a)], it is better to locate the active element in the pellet interior, while if the catalyst cost is low [see Fig. 25(b)] the optimal location moves toward the pellet external surface, in order to favor the main reactant transport process. Comparisons were made only among Dirac-type catalysts, and in all cases significant improvements of reactor performance were reported when using the appropriate Dirac-delta distribution."

EXHIBIT E

Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

'864 patent	***"Optimal Distribution of Catalyst in Pellets" ("Gavriilidis Article")***



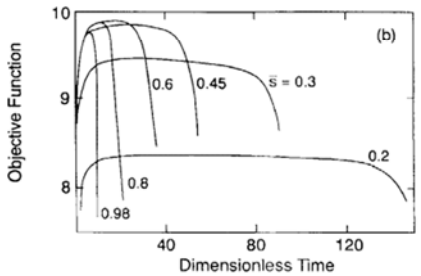


FIG. 25. Objective function time dependence for various active layer locations; $\phi_p = 10$, $Da_p = 10$, (a) $(a_1 - a_2)/a_3 = 0.1$, (b) $(a_1 - a_2)/a_3 = 10$. Isothermal first-order reaction, poisoning kinetics first-order. Objective function: (Cost of products per operating time − cost of raw materials and catalyst per operating time)/operating time [64].

[Page 433]

"An attempt to experimentally verify the improved performance of nonuniform catalyst pellets in the case of deactivating systems is reported by Rice and Hahn [66]. Three catalyst pellets (nickel supported on alumina) were tested: uniform, egg-shell, and egg-yolk. The methanation reaction

$$CO + 3H_2 \rightarrow CH_4 + H_2O \qquad (39)$$

was studied, under deactivation resulting from the formation of carbonaceous compounds as well as from poisoning by hydrogen sulfide, added in small concentrations in the feed stream. Substantial differences in the behavior of the three pellets were observed in terms of both product selectivity and catalyst lifetime. However, as noted by the authors, since methanation

'864 patent	***"Optimal Distribution of Catalyst in Pellets"* (*"Gavriilidis Article"*)**
	is a structure-sensitive reaction, its kinetics depend upon the active element dispersion which indeed was quite different for the three pellets examined. This aspect was strong enough to mask the effect of the active element distribution throughout the support, thus preventing any definitive conclusions to be drawn about the effectiveness of the catalyst design itself. The same reaction system was also studied in step-type catalyst pellets by Wu et al. [67]. It was found that when the depth of the active layer was greater than a certain value, the influence of carbon deposition was negligible." [Page 433-435] "The poisoning process is nonselective; that is, it involves both the support as well as the active catalyst. Since the poisoning process is diffusion controlled (i.e., pore-mouth poisoning), the poison penetrates the catalyst pellet in the form of a sharp front, whose velocity is substantially independent of the local concentration of the active elements. Summers and Hegedus [68] treated the case where only the two oxidizing reactions occur, and comparing several distributions of Pt and Pd, concluded that the best performance is obtained by a catalyst pellet which is impregnated with Pt at the outer shell, and Pd in a separate subsurface band, as illustrated in Fig. 26. This was attributed to the fact that Pt is more resistant to poisoning, with a nonzero residual activity, while Pd located in the interior provides higher hydrocarbon oxidation activity. The above configuration also guarantees a certain resistance to sintering, which is provided by the Pd band protected from poisoning by the external Pt band. When the reduction of nitric oxide is also desired, it is necessary to include in the catalyst pellet a certain amount of Rh. which catalyzes this reaction even in the presence of traces of oxygen. However, the high cost of Rh as compared to that of Pt and Pd, and its strong sensitivity to phosphorous and lead poisoning, suggest using a small amount of Rh in the pellet and protecting it from poisoning. Based upon the considerations reported above, a three-way catalyst was designed by Hegedus et al. [69]. On the whole, the catalyst pellets contained 1.12 g of Pt, 0.44 g of Pd, and 0.06 g of Rh per car. These were located in three separate bands, one underneath the other. The first, at

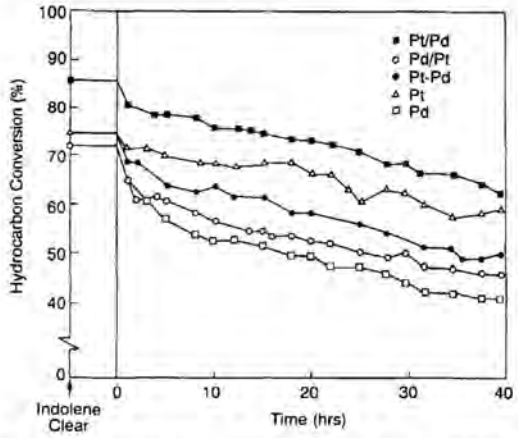
'864 patent	***"Optimal Distribution of Catalyst in Pellets" ("Gavriilidis Article")***
	the pellet's external surface, was the Pt band, whose depth was determined so as to entirely contain the expected penetration depth of the poison during the desired catalyst lifetime. Next was the Rh band, so that diffusional resistances in the nitric oxide reduction process would be minimized and it would also be protected from poisoning. Finally, the Pd band was placed at the deepest location, providing reasonable activity even at relatively low temperatures, typically encountered during engine startup conditions."  FIG. 26. Hydrocarbon conversion with time, during accelerated poisoning experiments. Pt/Pd: Pt (exterior)/Pd (interior); Pd/Pt: Pd (exterior)/Pt (interior); Pt–Pd: Pt and Pd (both exterior); Pt: Pt (exterior); Pd: Pd (exterior) [68]. [Page 436] "It was demonstrated that for hydrodemetallation, the overall deposited amount in a uniform pellet could be increased by up to 25% by simply decreasing the amount of catalyst located near the pore mouth, so as to delay its complete blockage. The optimal catalyst activity distributions were also computed numerically, and these exhibit a sharp maximum toward the center of the pellet followed by a decrease toward the external surface, so that the front of the fully deactivated catalyst originates at the center of the pellet and moves to the external surface. This distribution provides a good utilization of the catalyst, maximizing the overall amount of metals deposited.

EXHIBIT E

Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

'864 patent	***"Optimal Distribution of Catalyst in Pellets"* (*"Gavriilidis Article"*)**
	For hydrodesulfurization, Asua and Delman [71] have made a theoretical study for catalysts containing molybdenum and cobalt sulfides. In this case, catalyst performance optimization can be pursued by varying both the overall catalyst concentration (i.e., Mo + Co) as well as its composition (i.e., Mo/Co). They used a detailed kinetic model which accounted explicitly for the catalyst composition, as well as for the concentration of active sites, and showed that the catalyst performance could indeed be improved by using nonuniformly distributed catalyst pellets." [Page 440-441] "Do [98] modeled the impregnation of one adsorbing agent in a wet pellet. The steady-state solution of the diffusion-adsorption equation was obtained analytically, while the transient solution was obtained numerically. It was shown that when the impregnation is diffusion controlled, a sharp egg-shell catalyst distribution is obtained, particularly when there is no desorption of the active species, so that there is no chance for redistribution. A higher adsorption capacity of the support results in an increase of the overall loading, but it does not have a significant effect on the activity profile. Coimpregnation of pellets with metal precursor solution and a site-blocking agent can be used to manipulate intrapellet distribution of the active ingredient. The model calculations of Vincent and Merrill for dry impregnation were extended for such multicomponent adsorption by Kulkarni et al. [99]. The authors obtained qualitative agreement with the data of Shyr and Ernst [100]. Analytical solutions were obtained by Lee and Aris [101] and Lee [102]. Both models were able to produce a variety of distributions of the active material as, for example, uniform, sharply defined egg-shell, egg-white, and egg-yolk. The former study [101] also addressed the effect of diffusion after impregnation was complete, as well as the effect of drying. It was shown that the above phenomena had a smoothing effect on the nonuniformities of the catalyst distribution developed at the end of the impregnation step.

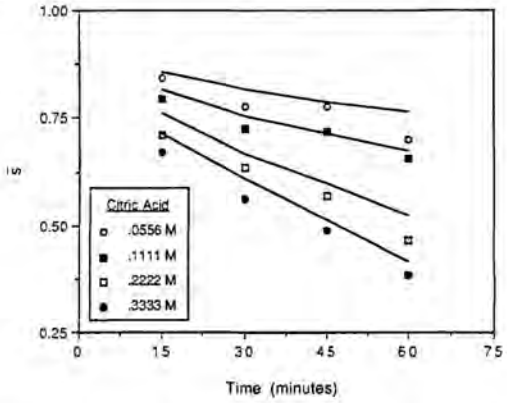
'864 patent	***"Optimal Distribution of Catalyst in Pellets" ("Gavriilidis Article")***
	Hegedus et al. modeled competitive multicomponent adsorption on wet pellets by using adsorption constants determined from separate experiments [103]. The model was used to predict the uptake of Rh in -y-Al $_2O_3$ pellets, as well as the Rh distribution inside the pellet, when HF was used as the site-blocking agent, by using the diffusion constants as adjustable parameters. Scelza et al. [104], using a similar model, fitted the data of Castro et al. [105], for coimpregnation of pellets with H 2PtC16 and HCl. However, not only the diffusion constants but also the adsorption constants were treated as adjustable parameters. Price [106], using a purely competitive Langmuir adsorption model for coimpregnation of chloroplatinic and citric acids, was able to successfully predict the location of the platinum layer inside a γ-Al_2O_3 pellet. For the case of sequential impregnation, the solution effect introduced by the competitive adsorbate was considered, and the resulting model agreed well with experimental results, as illustrated in Fig. 28."  FIG. 28. Comparison of model predictions and experimental platinum band locations for sequential impregnation with 0.00589 M hexachloroplatinic acid and various citric acid concentrations [106]. [Page 443] "When only one component is present during impregnation (which then must be the catalyst precursor), and in the case where drying does not affect distribution of the catalytic material,

EXHIBIT E

Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

'864 patent	***"Optimal Distribution of Catalyst in Pellets"* (*"Gavriilidis Article"*)**
	the relative rates of the transport and adsorption processes determine the type of distribution actually obtained. Fast transport or weak adsorption of the solute yield relatively uniform distributions, while strong adsorption or slow transport yield egg-shell distribution [109, 110]. The impregnating solution concentration and the impregnation time determine the thickness of the shell [111-113]. When impregnation with short contact time produces catalysts with decreasing concentration profile toward the pellet center, increasing time of impregnation smooths out catalyst nonuniformities [114]. The effect of the state of the support, dry or wet before impregnation, depends upon the adsorptivity of the catalyst precursor and is more pronounced for short impregnation time, in which case the effect of initial convective transport is significant. In the case of weak adsorption, dry impregnation yields a catalyst with more total loading and relatively more uniform than wet impregnation [115]. For strong adsorption, Summers and Ausen [110] observed that the penetration depth obtained from wet impregnation was slightly less than that from dry impregnation." [Page 444-446] "Most multicomponent impregnation studies concern platinum supported on a γ-Al_2O_3 pellet. Shyr and Ernst [100] studied the influence of a large number of acids and salts used as coimpregnants with H_2PtCl_6, on the distribution of Pt within γ-Al_2O_3 pellets, and obtained profiles shown in Fig. 31. If along with chloroplatinic acid, HCl is used as coimpregnant, uniform platinum distribution is obtained [121, 122]. When dibasic organic acids (e.g., citric or oxalic acid) are used instead, the platinum is driven into the pellet interior forming a subsurface layer with controlled location and width [2, 123]. Employing HF as coimpregnant, Hegedus et al. [103] prepared egg-white Rh/γ-Al $_2O_3$ catalysts. Increase of the impregnation time has been found to increase the local loading, the width, and the distance of the active layer from the external surface, while increase of the coimpregnant concentration pushes the active layer deeper [103, 123, 124]. When the pellet is impregnated with the catalyst precursor and competitive ad- sorbate sequentially, in this or the reverse order, similar egg-white catalysts are obtained. The width of the catalytic layer is decreased if the metal precursor is impregnated first, while if the impregnation order is reversed, the resulting catalyst has a thicker active layer [124]. Some platinum catalysts, prepared by using sequential impregnation

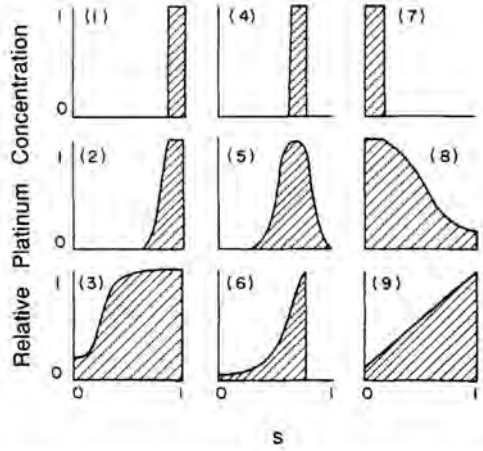
'864 patent	***"Optimal Distribution of Catalyst in Pellets"* (*"Gavriilidis Article"*)**
	of chloroplatinic and citric acids, are shown in Fig. 32; theoretical predictions of catalyst locations for types (a)-(c) were previously shown in Fig. 28. Maitra et al. studied the effect of pH by carefully monitoring and adjusting it during the impregnation process [125]. In this way, egg-shell or uniform tungsten catalysts were prepared. Schwarz and Heise, in an attempt to predict catalyst nonuniformities in their impregnation studies, classified the coimpregnants according to their effect on three interfacial phenomena [126]. The first class consists of inorganic salts such as NaCl, $NaNO_3$, and $CaCl_2$, which affect electrostatics at the solution-surface interface. The second class includes simple inorganic acids and bases such as HCI, HNO_3, and NaOH, which change the potential difference between the adsorbing ions and the surface. The third class of ingredients, for example, acetates, citrates, and phosphates, compete with the metal ion for possible adsorption sites. This competition is similar to a chromatographic process. These ingredients can also alter the electrostatics and potential of the heterogeneous system."  FIG. 31. Types of Pt profiles obtained in coimpregnation experiments [100].

EXHIBIT E

Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

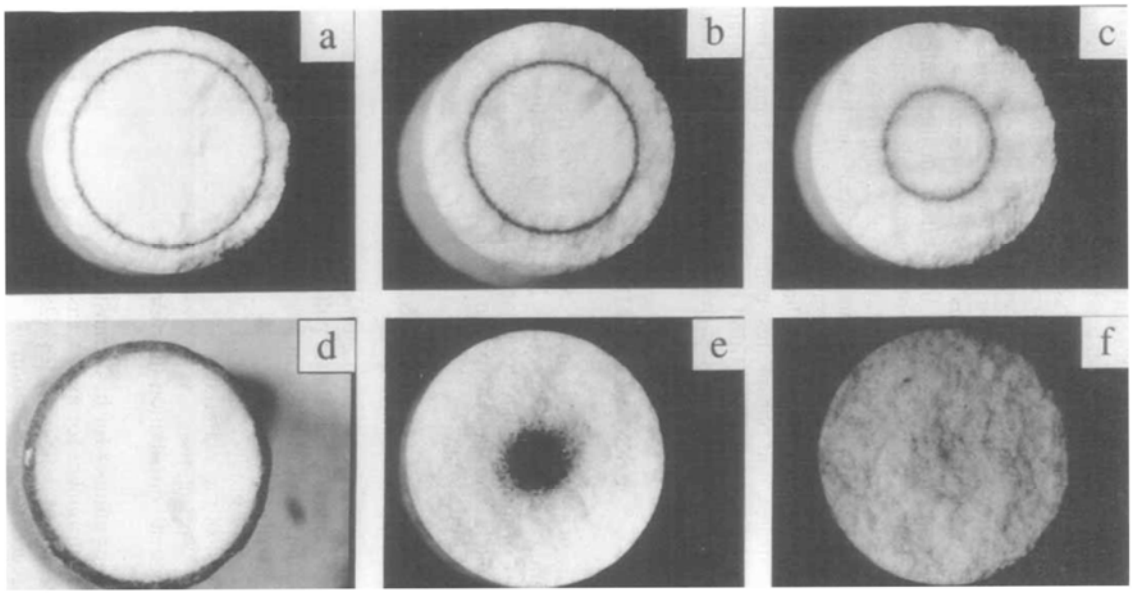
'864 patent	***"Optimal Distribution of Catalyst in Pellets" ("Gavriilidis Article")***
	 FIG. 32. Step-type platinum catalysts prepared by sequential and coimpregnation. (a) 0.106 M chloroplatinic acid for 15 min, followed by 1.0 M citric acid for 15 min; (b) 0.106 M chloroplatinic acid for 15 min, followed by 1.0 M citric acid for 60 min; (c) 0.106 M chloroplatinic acid for 15 min, followed by 3.0 M citric acid for 60 min; (d) 0.106 M chloroplatinic acid for 15 min; (e) 0.106 M chloroplatinic acid and 1.5 M citric acid coimpregnation for 3 h; (f) 0.106 M chloroplatinic acid and 1.5 M citric acid coimpregnation for 10 h [124]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Gavriilidis Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[3] The combustion promoter according to claim 1,	*Gavriilidis Article* discloses a combustion promoter comprising a plurality of microsphere sized porous particles comprising alumina. *See, e.g.*,

EXHIBIT E

Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

'864 patent	***"Optimal Distribution of Catalyst in Pellets"* ("*Gavriilidis Article*")**
comprising a plurality of microsphere sized porous particles comprising alumina.	The support provided above for Claim 1[b]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Gavriilidis Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[6] The combustion promoter according to claim 1, wherein the microsphere sized porous particles collectively have an average (D50) size of between 60 and 90 microns as measured by laser diffraction.	*Gavriilidis Article* discloses a combustion promoter comprising microsphere sized porous particles, wherein the microsphere sized porous particles collectively have an average (D50) size of between 60 and 90 microns as measured by laser diffraction. *See, e.g.*, The support provided above for Claim 1[a]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Gavriilidis Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[12] The combustion promoter according to claim 1, wherein the one or more Group VIII noble metals comprises one or more of platinum, palladium, iridium, ruthenium, and rhodium.	*Gavriilidis Article* discloses a combustion promoter comprising one or more Group VIII noble metals, wherein the one or more Group VIII noble metals comprises one or more of platinum, palladium, iridium, ruthenium, and rhodium. *See, e.g.*, The support provided above for Claim 1[c].

EXHIBIT E

Invalidity of U.S. Patent No. 11,224,864 in View of *Gavriilidis Article*

'864 patent	***"Optimal Distribution of Catalyst in Pellets" ("Gavriilidis Article")***
	To the extent that this limitation is not disclosed, either explicitly or inherently, by *Gavriilidis Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[15] The combustion promoter according to claim 12, wherein the one or more Group VIII noble metals comprises palladium.	*Gavriilidis Article* discloses a combustion promoter comprising one or more Group VIII noble metals, wherein the one or more Group VIII noble metals comprises palladium. *See, e.g.*, The support provided above for Claim 1[c]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Gavriilidis Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.

EXHIBIT F

EXHIBIT F

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2010/0190638 ("*Hagemeyer*")

U.S. Patent Application No. 2010/0190638 ("*Hagemeyer*"), filed on May 31, 2007 and published on July 29, 2010, qualifies as prior art under at least 35 U.S.C. §§ 102 and/or 103 for the asserted claims of U.S. Patent No. 11,224,864 ("the '864 patent"), as discussed in detail in the claim chart below.

The Court has not yet construed the claims and therefore the meaning of the terms in the claims has yet to be resolved by the Court. The support identified here for these limitations is responsive to Plaintiffs' infringement contentions, which Defendant disagrees with. As such, nothing in Defendant's claim charts should be construed as an admission regarding infringement, either literally or under the doctrine of equivalents, or as an admission regarding Defendant's understanding of the proper scope of the asserted claims. Defendant reserves the right to rely on additional citations or sources of evidence that also may be applicable, or that may become applicable in light of claim construction, changes in Plaintiffs' infringement contentions, and/or information obtained during discovery as the case progresses.

To the extent Plaintiffs allege that *Hagemeyer* does not disclose any particular limitation of the asserted claims of the '864 patent, either expressly or inherently, it would have been obvious to a person of ordinary skill in the art as of the priority date of the '864 patent to modify *Hagemeyer* and/or to combine the teachings of *Hagemeyer* with other prior art references, including but not limited to the present prior art references cited in the Cover Pleading and the relevant section(s) of claim charts for other prior art references for the '864 patent in a manner that would have rendered the asserted claims invalid as obvious.

Defendant reserves the right to amend or supplement this claim chart at a later date as more fully set forth in the Cover Pleading.

EXHIBIT F

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2010/0190638 ("*Hagemeyer*")

'864 patent	**U.S. Patent Application No. 2010/0190638 ("*Hagemeyer*")**
1[a] A CO to CO_2 combustion promoter comprising microsphere sized porous particles, each microsphere sized porous particle having a diameter of less than 1mm, and independently comprising	*Hagemeyer* discloses a CO to CO_2 combustion promoter comprising microsphere sized porous particles, each microsphere sized porous particle having a diameter of less than 1mm, and independently comprising the limitations discussed in the following sections. *See, e.g.*, [Abstract] "A method for producing a shell catalyst comprising a porous catalyst support shaped body with an outer shell containing at least one transition metal in metal form. To provide a shell catalyst with a relatively small shell thickness, a device is set up to circulate the catalyst support shaped bodies by means of process gases with a reductive effect. The device is charged with catalyst support shaped bodies that are circulated by means of a process gas with a reductive effect, an outer shell of the catalyst support shaped bodies is impregnated with a transition-metal precursor compound by spraying the circulating catalyst support shaped bodies with a solution containing the transition-metal precursor compound, the metal component of the transition-metal precursor compound is converted into the metal form by reduction by means of the process gas, and the catalyst support shaped bodies sprayed with the solution are dried." [0002] "The present invention relates to a method for producing a shell catalyst which comprises a porous catalyst support shaped body with an outer shell in which at least one transition metal in metal form is contained." [0003] "Supported transition-metal catalysts in the form of shell catalysts and also methods for their production are known in the state of the art. The catalytically active species-often also the promoters-are contained in shell catalysts only in an outer area (shell) of greater or lesser width of a catalyst support shaped body, i.e. they do not fully penetrate the catalyst support shaped body (cf. for example EP 565 952 A1, EP

EXHIBIT F

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2010/0190638 ("*Hagemeyer*")

'864 patent	**U.S. Patent Application No. 2010/0190638 ("*Hagemeyer*")**
	634 214Al, EP634 209Al and EP 634 208Al). With shell catalysts, a more selective reaction control is possible in many cases than with catalysts in which the support is loaded into the core of the support with the catalytically active species ('impregnated through')." [0004] "Vinyl acetate monomer (VAM) for example is currently produced predominantly by means of shell catalysts in high selectivity. The great majority of the shell catalysts used at present for producing VAM are shell catalysts with a Pd/Au shell on a porous amorphous aluminosilicate support, formed as a sphere, based on natural sheet silicates, wherein the supports are impregnated through with potassium acetate as promoter. In the Pd/Au system of these catalysts, the active metals Pd and Au are probably not present in the form of metal particles of the respective pure metal, but rather in the form of Pd/Au-alloy particles of possibly different composition, although the presence of unalloyed particles cannot be ruled out." [0059] "In the method according to aspects of the invention, porous shaped bodies of any shape can be used as catalyst supports, wherein the supports can be formed from any materials or material mixtures. However, catalyst supports which comprise at least one metal oxide or are formed from a metal oxide or a metal oxide mixture are preferred according to aspects of the invention. However, the catalyst support preferably comprises a silicon oxide, an aluminium oxide, an aluminosilicate, a zirconium oxide, a titanium oxide, a niobium oxide or a natural sheet silicate, preferably a calcined acid-treated bentonite." [0075] "To produce a shell catalyst for oxidation reactions, it is provided according to a further preferred embodiment of the method according to aspects of the invention that the solution of the transition-metal precursor compound contains a Pd compound as transition-metal precursor compound."

EXHIBIT F

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2010/0190638 ("*Hagemeyer*")

'864 patent	**U.S. Patent Application No. 2010/0190638 ("*Hagemeyer*")**
	[0093] "The present invention furthermore relates to a shell catalyst comprising a porous catalyst support shaped body with an outer shell in which at least one transition metal is contained in particulate metal form, characterized in that the proportion by mass of the transition metal in the catalyst is more than 0.3 mass-%, preferably more than 0.5 mass-% and by preference more than 0.8 mass-%, and the average dispersion of the transition-metal particles is greater than 20%, preferably greater than 23%, by preference greater than 25% and more preferably greater than 27%." [0099] "Catalysts preferred according to aspects of the invention contain two different metals in metal form in the shell, wherein the two metals are combinations of one of the following pairs: Pd and Ag; Pd and Au; Pd and Pt. Catalysts with a Pd/Au shell are suitable in particular for producing VAM, those with a Pd/Pt shell are suitable in particular as oxidation and hydrogenation catalysts and those with a Pd/Ag shell are suitable in particular for the selective hydrogenation of alkynes and dienes in olefin streams, thus for example for producing purified ethylene by selective hydrogenation of acetylene contained in the untreated product." [0115] "In order to ensure an adequate activity, a preferred oxidation or hydrogenation catalyst according to aspects of the invention contains Pd and Pt as noble metals, wherein the proportion of Pd in the catalyst is 0.5 to 5 mass-%, preferably 0.1 to 2.5 mass-% and by preference 0.15 to 0.8 mass-%, relative to the mass of the catalyst support loaded with noble metal." [0129] "The catalyst support of the catalyst according to aspects of the invention is formed as a shaped body. The catalyst support can in principle assume the form of any geometric body to which a corresponding shell can be applied. However, it is

EXHIBIT F

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2010/0190638 ("*Hagemeyer*")

'864 patent	**U.S. Patent Application No. 2010/0190638 ("*Hagemeyer*")**
	preferred if the catalyst support is formed as a sphere, cylinder (also with rounded end surfaces), perforated cylinder (also with rounded end surfaces), trilobe, 'capped tablet,' tetralobe, ring, doughnut, star, cartwheel, 'reverse' cartwheel, or as a strand, preferably as a ribbed strand or star strand." [0130] "The diameter or the length and thickness of the catalyst support of the catalyst according to aspects of the invention is preferably 2 to 9 mm, depending on the geometry of the reactor tube in which the catalyst is to be used." To the extent that this limitation is not disclosed, either explicitly or inherently, by *Hagemeyer*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
1[b] silica, alumina, or mixtures thereof; and	*Hagemeyer* discloses a combustion promoter comprising silica, alumina, or mixtures thereof. *See, e.g.*, [0004] "Vinyl acetate monomer (VAM) for example is currently produced predominantly by means of shell catalysts in high selectivity. The great majority of the shell catalysts used at present for producing VAM are shell catalysts with a Pd/Au shell on a porous amorphous aluminosilicate support, formed as a sphere, based on natural sheet silicates, wherein the supports are impregnated through with potassium acetate as promoter. In the Pd/Au system of these catalysts, the active metals Pd and Au are probably not present in the form of metal particles of the respective pure metal, but rather in the form of Pd/Au-alloy particles of possibly different composition, although the presence of unalloyed particles cannot be ruled out."

EXHIBIT F

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2010/0190638 ("*Hagemeyer*")

'864 patent	**U.S. Patent Application No. 2010/0190638 ("*Hagemeyer*")**
	[0059] "In the method according to aspects of the invention, porous shaped bodies of any shape can be used as catalyst supports, wherein the supports can be formed from any materials or material mixtures. However, catalyst supports which comprise at least one metal oxide or are formed from a metal oxide or a metal oxide mixture are preferred according to aspects of the invention. However, the catalyst support preferably comprises a silicon oxide, an aluminium oxide, an aluminosilicate, a zirconium oxide, a titanium oxide, a niobium oxide or a natural sheet silicate, preferably a calcined acid-treated bentonite." [0120] "According to a further preferred embodiment of the catalyst according to aspects of the invention, it is provided that the catalyst support is a support based on a silicon oxide, an aluminium oxide, an aluminosilicate, a zirconium oxide, a titanium oxide, a niobium oxide or a natural sheet silicate, preferably a calcined acid-treated bentonite. The expression 'based on' means that the catalyst support comprises one or more of the named materials." To the extent that this limitation is not disclosed, either explicitly or inherently, by *Hagemeyer*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
1[c] one or more Group VIII noble metals distributed in the particle as an eggshell such that a higher concentration of the one or more Group VIII noble metals is present in	*Hagemeyer* discloses a combustion promoter comprising one or more Group VIII noble metals, the metals distributed in the particle as an eggshell such that a higher concentration of the one or more Group VIII noble metals is present in the outer region of the microsphere sized porous particle as compared to the concentration of

EXHIBIT F

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2010/0190638 ("*Hagemeyer*")

'864 patent	**U.S. Patent Application No. 2010/0190638 ("*Hagemeyer*")**
the outer region of the microsphere sized porous particle as compared to the concentration of the one or more Group VIII noble metals in the centre of the microsphere sized porous particle.	the one or more Group VIII noble metals in the centre of the microsphere sized porous particle. *See, e.g.*, [Abstract] "A method for producing a shell catalyst comprising a porous catalyst support shaped body with an outer shell containing at least one transition metal in metal form. To provide a shell catalyst with a relatively small shell thickness, a device is set up to circulate the catalyst support shaped bodies by means of process gases with a reductive effect. The device is charged with catalyst support shaped bodies that are circulated by means of a process gas with a reductive effect, an outer shell of the catalyst support shaped bodies is impregnated with a transition-metal precursor compound by spraying the circulating catalyst support shaped bodies with a solution containing the transition-metal precursor compound, the metal component of the transition-metal precursor compound is converted into the metal form by reduction by means of the process gas, and the catalyst support shaped bodies sprayed with the solution are dried." [0002] "The present invention relates to a method for producing a shell catalyst which comprises a porous catalyst support shaped body with an outer shell in which at least one transition metal in metal form is contained." [0003] "Supported transition-metal catalysts in the form of shell catalysts and also methods for their production are known in the state of the art. The catalytically active species-often also the promoters-are contained in shell catalysts only in an outer area (shell) of greater or lesser width of a catalyst support shaped body, i.e. they do not fully penetrate the catalyst support shaped body (cf. for example EP 565 952 Al, EP 634 214Al, EP634 209Al and EP 634 208Al). With shell catalysts, a more selective reaction control is possible in many cases than with catalysts in which the support is

EXHIBIT F

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2010/0190638 ("*Hagemeyer*")

'864 patent	**U.S. Patent Application No. 2010/0190638 ("*Hagemeyer*")**
	loaded into the core of the support with the catalytically active species ('impregnated through')." [0004] "Vinyl acetate monomer (VAM) for example is currently produced predominantly by means of shell catalysts in high selectivity. The great majority of the shell catalysts used at present for producing VAM are shell catalysts with a Pd/Au shell on a porous amorphous aluminosilicate support, formed as a sphere, based on natural sheet silicates, wherein the supports are impregnated through with potassium acetate as promoter. In the Pd/Au system of these catalysts, the active metals Pd and Au are probably not present in the form of metal particles of the respective pure metal, but rather in the form of Pd/Au-alloy particles of possibly different composition, although the presence of unalloyed particles cannot be ruled out." [0006]-[0009] "The Pd/Au shell of a VAM shell catalyst is produced for example by first steeping the catalyst support shaped body in a first step in an Na_2PdC1_4 solution and then in a second step fixing the Pd component with NaOH solution onto the catalyst support in the form of a Pd-hydroxide compound. In a subsequent, separate third step, the catalyst support is then steeped in an $NaAuC1_4$ solution and then the Au component is likewise fixed by means of NaOH. It is also possible for example to firstly steep the support in lye and then apply the precursor compounds to the thus-pretreated support. After the fixing of the noble-metal components to the catalyst support, the loaded catalyst support is then very largely washed free of chloride and Na ions, then dried and finally reduced with ethylene at 150° C. The produced Pd/Au shell is usually approximately 100 to 500 μm thick, wherein normally the smaller the thickness of its shell, the higher the product selectivity of a shell catalyst. Usually, the catalyst support loaded with the noble metals is then loaded with potassium acetate after the fixing or reducing step wherein, rather than the loading

EXHIBIT F

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2010/0190638 ("*Hagemeyer*")

'864 patent	U.S. Patent Application No. 2010/0190638 ("*Hagemeyer*")
	with potassium acetate taking place only in the outer shell loaded with noble metals, the catalyst support is completely impregnated through with the promoter. According to the state of the art, the active metals Pd and Au, starting from chloride compounds in the area of a shell of the support, are applied to same by means of steeping. However, this technique has reached its limits as regards minimum shell thicknesses. The smallest achievable shell thickness of correspondingly produced VAM catalysts is at best approx. 100 µm and it is not foreseen that even thinner shells can be obtained by means of steeping. In addition, the catalysts produced by means of steeping have a relatively large average dispersion of the noble-metal particles, which can have a disadvantageous effect in particular on the activity of the catalyst. An object of the present invention is to provide a shell catalyst production method by means of which supported transition-metal catalysts formed as shell catalysts can be produced which have a relatively small shell thickness." [0024] "In particular in respect of noble metals as metal components to be reduced, gas mixtures of hydrogen with nitrogen or argon can be preferred, preferably with a hydrogen content between 1 vol.-% and 15 vol.-%. The method according to aspects of the invention is carried out for example with hydrogen (5 vol.-%) in nitrogen as process gas at a temperature of approximately 150° C. over a period of for example 5 hours. If the desired quantity of transition-metal precursor compound solution has been deposited onto the shaped bodies, the spraying can be stopped and the circulation continued until the deposited metal component has been completely reduced." [0074] "According to a further preferred embodiment of the method according to aspects of the invention, it is provided that the noble-metal compound is selected from the

EXHIBIT F

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2010/0190638 ("*Hagemeyer*")

'864 patent	**U.S. Patent Application No. 2010/0190638 ("*Hagemeyer*")**
	halides, in particular chlorides, oxides, nitrates, nitrites, formates, pro-pionates, oxalates, acetates, citrates, tartrates, lactates, hydroxides, hydrogen carbonates, amine complexes or organic complexes, for example triphenylphosphine complexes or acetylacetonate complexes, of the noble metals." [0077] "To produce a platinum-containing shell catalyst, it is provided according to a further preferred embodiment of the method according to aspects of the invention that the solution of the transition-metal precursor compound contains a Pt compound as transition-metal precursor compound." [0080]-[0081] "With the methods described in the state of the art for producing VAM shell catalysts based on Pd and Au, commercially available solutions of the precursor compounds such as Na_2PdCl_4, $NaAuCl_4$ or $HAuCl_4$ solutions are customarily used. In the more recent literature, chloride-free Pd or Au precursor compounds such as for example $Pd(NH_3)_4(OH)_2$, $Pd(NH_3)_2(NO_2)_2$ and $KAuO_2$ are also used. These precursor compounds react base in solution, while the standard chloride, nitrate and acetate precursor compounds all react acid in solution. In principle, any Pd or Au compound by means of which a degree of dispersion of the metal particles high enough for VAM synthesis can be achieved can be used as Pd and Au precursor compound. By 'degree of dispersion' is meant the ratio of the number of all the surface metal atoms (of the metal concerned) of all the metal/alloy particles of a supported metal catalyst to the total number of all the metal atoms of the metal/alloy particles. In general it is preferred if the degree of dispersion corresponds to a relatively high numerical value, since in this case as many metal atoms as possible are freely accessible for a catalytic reaction. This means that, given a relatively high degree of dispersion of a supported metal catalyst, a specific catalytic activity of same can be achieved with a relatively small quantity of metal used."

EXHIBIT F

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2010/0190638 ("*Hagemeyer*")

'864 patent	**U.S. Patent Application No. 2010/0190638 ("*Hagemeyer*")**
	[0093]-[0100] "The present invention furthermore relates to a shell catalyst comprising a porous catalyst support shaped body with an outer shell in which at least one transition metal is contained in particulate metal form, characterized in that the proportion by mass of the transition metal in the catalyst is more than 0.3 mass-%, preferably more than 0.5 mass-% and by preference more than 0.8 mass-%, and the average dispersion of the transition-metal particles is greater than 20%, preferably greater than 23%, by preference greater than 25% and more preferably greater than 27%. Transition-metal shell catalysts with such high metal loadings with a simultaneously high metal dispersion can be obtained by means of the method according to aspects of the invention. The transition-metal dispersion is determined by means of the DIN standard for the respective metal in question. On the other hand, the dispersion of the noble metals Pt, Pd and Rh is determined by means of CO chemisorption according to "Journal of Catalysis 120, 370 -376 (1989)". The dispersion of Cu is determined by means of N_2O. According to a preferred embodiment of the shell catalyst according to aspects of the invention, the concentration of the transition metal varies, over an area of 90% of the shell thickness, the area being at a distance of 5% of the shell thickness from each of the outer and inner shell limit, from the average concentration of transition metal of this area by a maximum of +/-20%, preferably by a maximum of +/-15% and by preference by a maximum of +/-10%. Due to the largely uniform distribution of the transition metal within the shell, a largely uniform activity of the catalyst according to aspects of the invention over the thickness of the shell is ensured, as the concentration of transition metal varies only relatively little over the shell thickness. In other words, the profile of the concentration of transition metal describes an approximately rectangular function over the shell thickness.

EXHIBIT F

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2010/0190638 ("*Hagemeyer*")

'864 patent	**U.S. Patent Application No. 2010/0190638 ("*Hagemeyer*")**
	To further increase the selectivity of the catalyst according to aspects of the invention, it can be provided that, seen over the thickness of the shell of the catalyst, the maximum concentration of transition metal is in the area of the outer shell limit and the concentration decreases towards the inner shell limit. It can be preferred if the concentration of transition metal decreases constantly towards the inner shell limit over an area of at least 25% of the shell thickness, preferably over an area of at least 40% of the shell thickness and by preference over an area of 30 to 80% of the shell thickness. According to a further preferred embodiment of the catalyst according to aspects of the invention, the concentration of transition metal decreases roughly constantly towards the inner shell limit to a concentration of 50 to 90% of the maximum concentration, preferably to a concentration of 70 to 90% of the maximum concentration. It is preferred if the transition metal is selected from the group of the noble metals. Catalysts preferred according to aspects of the invention contain two different metals in metal form in the shell, wherein the two metals are combinations of one of the following pairs: Pd and Ag; Pd and Au; Pd and Pt. Catalysts with a Pd/Au shell are suitable in particular for producing VAM, those with a Pd/Pt shell are suitable in particular as oxidation and hydrogenation catalysts and those with a Pd/Ag shell are suitable in particular for the selective hydrogenation of alkynes and dienes in olefin streams, thus for example for producing purified ethylene by selective hydrogenation of acetylene contained in the untreated product. With regard to the provision of a VAM shell catalyst with adequate VAM activity, it is preferred that the catalyst contains Pd and Au as noble metals and the proportion of Pd in the catalyst is 0.6 to 2.5 mass-%, preferably 0.7 to 2.3 mass-% and by preference 0.8 to 2 mass-%, relative to the mass of the catalyst support loaded with noble metal."

EXHIBIT F

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2010/0190638 ("*Hagemeyer*")

'864 patent	**U.S. Patent Application No. 2010/0190638 ("*Hagemeyer*")**
	[0111] "According to an alternative embodiment of the catalyst according to aspects of the invention, it contains Pd and Ag as noble metals and, to ensure an adequate activity of the catalyst, preferably in the hydrogenation of acetylene, the proportion of Pd in the catalyst is 0.01 to 1.0 mass-%, preferably 0.02 to 0.8 mass-% and by preference 0.03 to 0.7 mass-%, relative to the mass of the catalyst support loaded with noble metal." [0115] "In order to ensure an adequate activity, a preferred oxidation or hydrogenation catalyst according to aspects of the invention contains Pd and Pt as noble metals, wherein the proportion of Pd in the catalyst is 0.5 to 5 mass-%, preferably 0.1 to 2.5 mass-% and by preference 0.15 to 0.8 mass-%, relative to the mass of the catalyst support loaded with noble metal." [0121] "As already stated above, the catalyst support of the catalyst according to aspects of the invention is subjected to a degree of mechanical stress during production of the catalyst. In addition, the catalyst according to aspects of the invention can be subjected to a strong mechanical load stress during the filling of a reactor, which can result in an undesired formation of dust and damage to the catalyst support, in particular to its catalytically active shell lying in an outer area." [0131]-[0132] "In general, the smaller the thickness of the shell of the catalyst, the higher the product selectivity of the catalyst according to aspects of the invention. According to a further preferred embodiment of the catalyst according to aspects of the invention, the shell of the catalyst therefore has a thickness of less than 300 μm, preferably less than 200 μm, by preference less than 150 μm, further preferably less than 100 μm and more preferably less than 80 μm. As a rule in the case of supported

EXHIBIT F

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2010/0190638 ("*Hagemeyer*")

'864 patent	**U.S. Patent Application No. 2010/0190638 ("*Hagemeyer*")**
	metal catalysts, the thickness of the shell can be measured visually by means of a microscope. The area in which the metals are deposited appears black, while the areas free of metals appear white. As a rule, the boundary between areas containing metals and areas free of them is very sharp and can clearly be recognized visually. If the above-named boundary is not sharply defined and accordingly not clearly recognizable visually or the shell thickness cannot be determined visually for other reasons, the thickness of the shell corresponds to the thickness of a shell, measured starting from the outer surface of the catalyst support, which contains 95% of the transition metal deposited on the support. However, it was likewise found that in the case of the catalyst according to aspects of the invention the shell can be formed with a relatively large thickness effecting a high activity of the catalyst, without effecting an appreciable reduction of the product selectivity of the catalyst according to aspects of the invention. Catalyst supports with a relatively small surface area are to be used for this. According to another preferred embodiment of the catalyst according to aspects of the invention, the shell of the catalyst therefore has a thickness of between 200 and 2000 µm, preferably between 250 and 1800 µm, by preference between 300 and 1500 µm and further preferably between 400 and 1200 µm." To the extent that this limitation is not disclosed, either explicitly or inherently, by *Hagemeyer*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[3] The combustion promoter according to claim 1, comprising a plurality of	*Hagemeyer* discloses a combustion promoter comprising a plurality of microsphere sized porous particles comprising alumina. *See, e.g.*,

EXHIBIT F

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2010/0190638 ("*Hagemeyer*")

'864 patent	**U.S. Patent Application No. 2010/0190638 ("*Hagemeyer*")**
microsphere sized porous particles comprising alumina.	The support provided above for Claim 1[b]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Hagemeyer*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[6] The combustion promoter according to claim 1, wherein the microsphere sized porous particles collectively have an average (D50) size of between 60 and 90 microns as measured by laser diffraction.	*Hagemeyer* discloses a combustion promoter comprising microsphere sized porous particles, wherein the microsphere sized porous particles collectively have an average (D50) size of between 60 and 90 microns as measured by laser diffraction. *See, e.g.*, The support provided above for Claim 1[a]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Hagemeyer*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[12] The combustion promoter according to claim 1, wherein the one or more Group VIII noble metals comprises one or more of platinum, palladium, iridium, ruthenium, and rhodium.	*Hagemeyer* discloses a combustion promoter comprising one or more Group VIII noble metals, wherein the one or more Group VIII noble metals comprises one or more of platinum, palladium, iridium, ruthenium, and rhodium. *See, e.g.*, The support provided above for Claim 1[c].

EXHIBIT F

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2010/0190638 ("*Hagemeyer*")

'864 patent	**U.S. Patent Application No. 2010/0190638 ("*Hagemeyer*")**
	To the extent that this limitation is not disclosed, either explicitly or inherently, by *Hagemeyer*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[15] The combustion promoter according to claim 12, wherein the one or more Group VIII noble metals comprises palladium.	*Hagemeyer* discloses a combustion promoter comprising one or more Group VIII noble metals, wherein the one or more Group VIII noble metals comprises palladium. *See, e.g.*, The support provided above for Claim 1[c]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Hagemeyer*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.

EXHIBIT G

EXHIBIT G

Invalidity of U.S. Patent No. 11,224,864 in View of *Harold Article*

"*Impact of the Catalytic Activity Profile on Observed Multiplicity Features: CO Oxidation on Pt/Al_2O_3*" ("*Harold Article*"), authored by Michael P. Harold et. al. and published in 1987, qualifies as prior art of U.S. Patent No. 11,224,864 ("the '864 patent") under at least 35 U.S.C. §§ 102 and/or 103 for the asserted claims of the '864 patent, as discussed in detail in the claim chart below.

The Court has not yet construed the claims and therefore the meaning of the terms in the claims has yet to be resolved by the Court. The support identified here for these limitations is responsive to Plaintiffs' infringement contentions, which Defendant disagrees with. As such, nothing in Defendant's claim charts should be construed as an admission regarding infringement, either literally or under the doctrine of equivalents, or as an admission regarding Defendant's understanding of the proper scope of the asserted claims. Defendant reserves the right to rely on additional citations or sources of evidence that also may be applicable, or that may become applicable in light of claim construction, changes in Plaintiffs' infringement contentions, and/or information obtained during discovery as the case progresses.

To the extent Plaintiffs allege that *Harold Article* does not disclose any particular limitation of the asserted claims of the '864 patent, either expressly or inherently, it would have been obvious to a person of ordinary skill in the art as of the priority date of the '864 patent to modify *Harold Article* and/or to combine the teachings of *Harold Article* with other prior art references, including but not limited to the present prior art references cited in the Cover Pleading and the relevant section(s) of claim charts for other prior art references for the '864 patent in a manner that would have rendered the asserted claims invalid as obvious.

Defendant reserves the right to amend or supplement this claim chart at a later date as more fully set forth in the Cover Pleading.

EXHIBIT G

Invalidity of U.S. Patent No. 11,224,864 in View of *Harold Article*

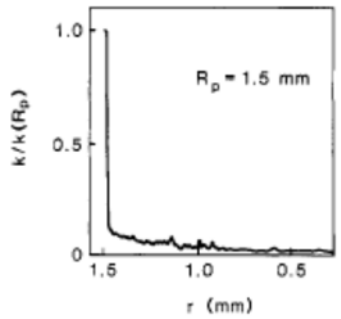
'864 patent	***"Impact of the Catalytic Activity Profile on Observed Multiplicity Features: CO Oxidation on Pt/Al_2O_3" ("Harold Article")***
1[a] A CO to CO_2 combustion promoter comprising microsphere sized porous particles, each microsphere sized porous particle having a diameter of less than 1mm, and independently comprising	*Harold Article* discloses a CO to CO_2 combustion promoter comprising microsphere sized porous particles, each microsphere sized porous particle having a diameter of less than 1mm, and independently comprising the limitations discussed in the following sections. *See, e.g.*, [Page 1617] "Previous studies predict that the optimal activity profile is rather sharp, i.e., in the form of a delta or step function. Metal dispersion during impregnation and drying (Foger, 1984) makes it difficult to attain such a sharp metal profile. However, if needed, it is possible to prepare catalysts with rather sharp metal profiles (Shyr and Ernst, 1980). For example, Figure 1 shows a scanning electron microprobe (SEM) analysis of a typical fresh, commercial 0.3% Pt on alumina pellet (3.0-mm diameter) made by OxyCatalyst (division of Met-Pro Corporation). The profile indicates that most of the Pt is contained in a thin surface shell (about 40 μm thick). The remaining Pt is dispersed throughout the inner core. A comparison of the profiles of several fresh pellets from the same batch revealed similar qualitative features.  **Figure 1.** Measured Pt profile (arbitrary units) of a fresh Pt/Al_2O_3 pellet.

EXHIBIT G

Invalidity of U.S. Patent No. 11,224,864 in View of *Harold Article*

'864 patent	***"Impact of the Catalytic Activity Profile on Observed Multiplicity Features: CO Oxidation on Pt/Al_2O_3" ("Harold Article")***
	Sintering and metal surface diffusion may alter the metal profile during operation. Figure 2 shows a Pt profile of an aged larger pellet (4.5-mm diameter) from the same batch as the fresh pellet (Figure 1). The aged pellet was used extensively for about a 6-month period to study the multiplicity feature of ethane and carbon monoxide oxidation (Harold and Luss, 1985)." [Page 1617] "Matsuura and Kato (1967) showed that when a homogeneous reaction with bimolecular Langmuir-Hinshelwood kinetics is carried out in an isothermal CSTR, the conversion can be a multivalued function of the feed concentration. Similarly, the interaction between intraparticle diffusion and a bimolecular Langmuir-Hinshelwood reaction may cause the rate of a single pellet to be a multivalued function of the limiting reactant surface concentration (Elnashaie and Yates, 1973; Wei and Becker, 1974). Hegedus et al. (1977) and Oh et al. (1979) confirmed experimentally the existence of rate multiplicity caused by this interaction during CO oxidation on Pt/Al_2O_3. The bifurcation diagrams of such systems, say of the rate or pellet temperature vs. ambient temperature or limiting reactant concentration, may give useful information about the functional form of the rate expression (Harold et al., 1987a,b). Kunimori et al. (1986) observed recently that the range of temperatures for which multiplicity occurred during the oxidation of CO on Pt/ Al_2O_3 pellets depended on the metal profile." [Page 1617] "The kinetics of many reactions are described by a bi-molecular Langmuir-Hinshelwood rate expression. The concentration profile of the reactant within a porous, spherical catalytic pellet in which such a reaction is carried out isothermally satisfies the equation

EXHIBIT G

Invalidity of U.S. Patent No. 11,224,864 in View of *Harold Article*

'864 patent	***"Impact of the Catalytic Activity Profile on Observed Multiplicity Features: CO Oxidation on Pt/Al_2O_3" ("Harold Article")***

$$\frac{1}{s^2}\frac{\mathrm{d}}{\mathrm{d}s}\left(s^2\frac{\mathrm{d}x}{\mathrm{d}s}\right) - \frac{\phi^2 a(s)x}{(1+Qx)^2} = 0 \qquad (1)$$

$$x = 1 \qquad s = 3 \qquad (2)$$

$$\frac{\mathrm{d}x}{\mathrm{d}s} = 0 \qquad s = 0 \qquad (3)$$

where

$$x = C/C_s \qquad Q = KC_s \qquad s = \frac{3r}{R_p} \qquad \phi^2 = \frac{R_p^2\bar{k}}{9D_e} \qquad (4)$$

$$a(s) = \frac{k(s)}{\bar{k}} \qquad \bar{k} = \frac{1}{9}\int_0^3 s^2\,k(s)\,\mathrm{d}s$$

We assume that the activity is proportional to the active metal concentration and that its profile depends only on the radial position. The analysis can be readily modified to cases in which this assumption is not valid."

[Page 1618]
"Luss (1971) proved that multiplicity cannot occur within a uniformly active, porous, symmetric pellet for $Q_c < 8$. This limiting value is rather close to $Q_c \approx 11$, found numerically for this specific activity distribution. The cusp-shaped bifurcation set is qualitatively similar to that found for a uniformly active catalytic slab. This is not surprising since for sufficiently large $\phi/(1 + Q)$, the reaction occurs almost entirely near the surface."

[Page 1619]
"Vega and Linan (1982) and Witmer et al. (1986) showed that for bimolecular Langmuir-Hinshelwood kinetics, an infinite number of solutions exist for a bounded range of ϕ as $Q \rightarrow \infty$. A similar behavior was found by Aris (1979) for a first-order exothermic reaction in a spherical pellet. Jackson (1973) has shown that only the

'864 patent	***"Impact of the Catalytic Activity Profile on Observed Multiplicity Features: CO Oxidation on Pt/Al_2O_3" ("Harold Article")***
	two bounding solutions are stable. As we are concerned with observed features, the unstable solutions are only of academic interest." [Page 1620] "The position of the maximal activity relative to the multiplicity region may provide a useful indication about the activity profile. For example, Figure 10 describes the observed isothermal rate vs. C_s for three different nonuniform pellets. The kinetics were assumed to follow the rate expression $$R = \frac{k_0 \exp(-E/RT)C_s}{[1 + K_0 \exp(-\Delta H_a/R_g T)C_s]^2} \quad (14)$$ using the parameter values of $k_0 = 2.357 \times 10^{11}\ s^{-1}$ $E = 13243$ cal/gmol $K_0 = 1.449 \times 10^7\ cm^3/gmol$ (15) $(-\Delta H_a) = 1561$ cal/gmol determined by Harold and Luss (1987) for the nonisothermal oxidation of CO. The pellet temperature is assumed to be 150 °C which, for R_P = 2 mm and D_e = 0.039 cm^2/s, corresponds to a surface Thiele modulus of $\phi_s = (R_p/3)\,[k(3)/D_e]^{1/2} = 62.1$. Case 1 is for a pellet with a uniformly active surface shell of 13% of the pellet radius and an inert core (ϕ = 36.2). Case 2 is for a pellet with a three-zone activity profile with s_1 = 2.25, s_2 = 2.97, and σ = 1 (ϕ = 35.6). Case 3 is for a pellet with a profile fit to the polynomial expression in eq 8 (ϕ = 36.2). Cases 1 and 3 have the same volume-averaged activity which differs only by a factor of 0.983 from that of case 2. Each profile predicts rate multiplicity; however, the features are quite different. The range of C_s, for which multiple rates exist, decreases and moves to a higher C_s range as the activity is distributed more uniformly. In addition, the C_s corresponding to the maximum rate is outside the multiplicity region for profiles 2 and 3. This feature is consistent with some of the data found by Harold (1985) for CO oxidation on Pt/Al_2O_3. Beusch et al. (1972) also found the maximum rate to be

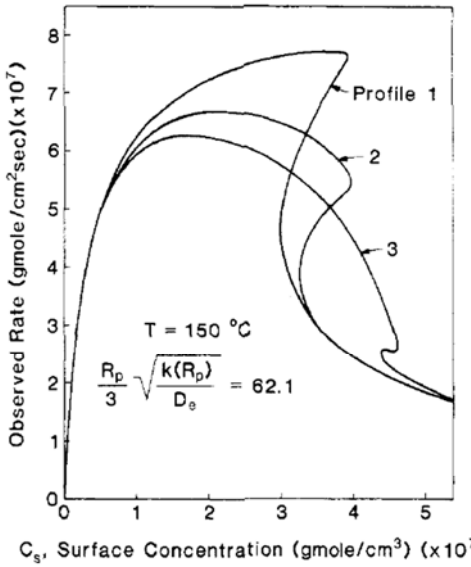
'864 patent	***"Impact of the Catalytic Activity Profile on Observed Multiplicity Features: CO Oxidation on Pt/Al_2O_3" ("Harold Article")***
	outside the multiplicity region during CO oxidation on a single porous Pt/Al_2O_3 pellet. These data suggest an interaction between negative-order kinetics and pore diffusion in a pellet with a nonuniform activity. Beusch et al.'s (1972) contention that the multiplicity is due solely to the intrinsic kinetics may be erroneous."  **Figure 10.** Dependence of the observed isothermal rate per unit pellet area on the CO surface concentration for three different activity profiles. Profile 1 corresponds to a uniformly active surface shell with a thickness of $0.13R_p$. Profile 2 is of a three-zone profile with $s_1 = 2.25$, $s_2 = 2.97$, and $\sigma = 1$. Profile 3 is defined by eq 8. [Page 1620] "The simulations show that surface diffusion of the metal may affect significantly the multiplicity features and performance of catalytic pellets. Thus, changes in bifurcation diagrams may serve as an indication of possible changes in the activity

EXHIBIT G

Invalidity of U.S. Patent No. 11,224,864 in View of *Harold Article*

'864 patent	***"Impact of the Catalytic Activity Profile on Observed Multiplicity Features: CO Oxidation on Pt/Al_2O_3" ("Harold Article")***
	profile of the metal. This possibility can then be checked by slicing the pellet and measuring the metal by a SEM. In a recent study of the oxidation of CO on a nonuniformly impregnated Pt/Al_2O_3 catalyst, Kunimori et al. (1986) noted an increase in the range of temperatures for which multiplicity occurred and a reduction in the temperature of the high conversion state. Following Hegedus et al. (1977), they attributed this to pore deposits which increase the diffusional resistance. Our simulations suggest that this behavior may have been caused by surface migration of the dispersed platinum to give a more uniform profile." To the extent that this limitation is not disclosed, either explicitly or inherently, by *Harold Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
1[b] silica, alumina, or mixtures thereof; and	*Harold Article* discloses a combustion promoter comprising silica, alumina, or mixtures thereof. *See, e.g.*,

EXHIBIT G

Invalidity of U.S. Patent No. 11,224,864 in View of *Harold Article*

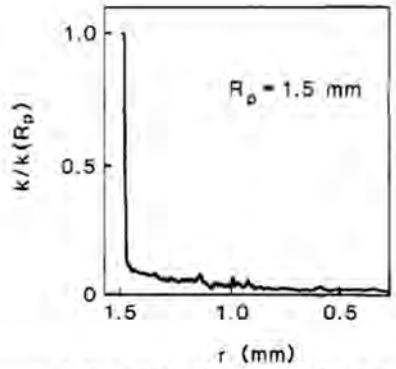
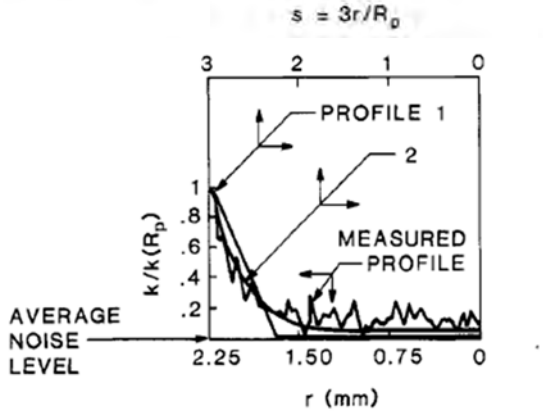
'864 patent	***"Impact of the Catalytic Activity Profile on Observed Multiplicity Features: CO Oxidation on Pt/Al_2O_3" ("Harold Article")***
	[Fig. 1 & 2]  **Figure 1.** Measured Pt profile (arbitrary units) of a fresh Pt/Al_2O_3 pellet.  **Figure 2.** Measured Pt profile (in counts) of aged 4.5-mm-diameter Pt/Al_2O_3 pellet. The three-zone model with $s_1 = 2.25$, $s_2 = 2.97$, $\sigma = 1$ (profile 1; eq 6 and 7) and the polynomial profile (2; eq 8) are also shown.

EXHIBIT G

Invalidity of U.S. Patent No. 11,224,864 in View of *Harold Article*

'864 patent	***"Impact of the Catalytic Activity Profile on Observed Multiplicity Features: CO Oxidation on Pt/Al_2O_3" ("Harold Article")***
	[Page 1617] "Previous studies predict that the optimal activity profile is rather sharp, i.e., in the form of a delta or step function. Metal dispersion during impregnation and drying (Foger, 1984) makes it difficult to attain such a sharp metal profile. However, if needed, it is possible to prepare catalysts with rather sharp metal profiles (Shyr and Ernst, 1980). For example, Figure 1 shows a scanning electron microprobe (SEM) analysis of a typical fresh, commercial 0.3% Pt on alumina pellet (3.0-mm diameter) made by OxyCatalyst (division of Met-Pro Corporation). The profile indicates that most of the Pt is contained in a thin surface shell (about 40 μm thick). The remaining Pt is dispersed throughout the inner core. A comparison of the profiles of several fresh pellets from the same batch revealed similar qualitative features." [Page 1617] "Matsuura and Kato (1967) showed that when a homogeneous reaction with bimolecular Langmuir-Hinshelwood kinetics is carried out in an isothermal CSTR, the conversion can be a multivalued function of the feed concentration. Similarly, the interaction between intraparticle diffusion and a bimolecular Langmuir-Hinshelwood reaction may cause the rate of a single pellet to be a multivalued function of the limiting reactant surface concentration (Elnashaie and Yates, 1973; Wei and Becker, 1974). Hegedus et al. (1977) and Oh et al. (1979) confirmed experimentally the existence of rate multiplicity caused by this interaction during CO oxidation on Pt/Al_2O_3. The bifurcation diagrams of such systems, say of the rate or pellet temperature vs. ambient temperature or limiting reactant concentration, may give useful information about the functional form of the rate expression (Harold et al., 1987a,b). Kunimori et al. (1986) observed recently that the range of temperatures for which multiplicity occurred during the oxidation of CO on Pt/Al_2O_3 pellets depended on the metal profile."

EXHIBIT G

Invalidity of U.S. Patent No. 11,224,864 in View of *Harold Article*

’864 patent	***“Impact of the Catalytic Activity Profile on Observed Multiplicity Features: CO Oxidation on Pt/Al_2O_3” (“Harold Article”)***
	[Page 1620] “The range of C_s, for which multiple rates exist, decreases and moves to a higher C_s range as the activity is distributed more uniformly. In addition, the C_s corresponding to the maximum rate is outside the multiplicity region for profiles 2 and 3. This feature is consistent with some of the data found by Harold (1985) for CO oxidation on Pt/ Al_2O_3. Beusch et al. (1972) also found the maximum rate to be outside the multiplicity region during CO oxidation on a single porous Pt/Al_2O_3 pellet. These data suggest an interaction between negative-order kinetics and pore diffusion in a pellet with a nonuniform activity. Beusch et al.'s (1972) contention that the multiplicity is due solely to the intrinsic kinetics may be erroneous.” [Page 1620] “The simulations show that surface diffusion of the metal may affect significantly the multiplicity features and performance of catalytic pellets. Thus, changes in bifurcation diagrams may serve as an indication of possible changes in the activity profile of the metal. This possibility can then be checked by slicing the pellet and measuring the metal by a SEM. In a recent study of the oxidation of CO on a nonuniformly impregnated Pt/Al_2O_3 catalyst, Kunimori et al. (1986) noted an increase in the range of temperatures for which multiplicity occurred and a reduction in the temperature of the high conversion state. Following Hegedus et al. (1977), they attributed this to pore deposits which increase the diffusional resistance. Our simulations suggest that this behavior may have been caused by surface migration of the dispersed platinum to give a more uniform profile.” To the extent that this limitation is not disclosed, either explicitly or inherently, by *Harold Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the ’864 patent to combine this reference with any of the other references as

EXHIBIT G

Invalidity of U.S. Patent No. 11,224,864 in View of *Harold Article*

'864 patent	***"Impact of the Catalytic Activity Profile on Observed Multiplicity Features: CO Oxidation on Pt/Al_2O_3" ("Harold Article")***
	charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
1[c] one or more Group VIII noble metals distributed in the particle as an eggshell such that a higher concentration of the one or more Group VIII noble metals is present in the outer region of the microsphere sized porous particle as compared to the concentration of the one or more Group VIII noble metals in the centre of the microsphere sized porous particle.	*Harold Article* discloses a combustion promoter comprising one or more Group VIII noble metals, the metals distributed in the particle as an eggshell such that a higher concentration of the one or more Group VIII noble metals is present in the outer region of the microsphere sized porous particle as compared to the concentration of the one or more Group VIII noble metals in the centre of the microsphere sized porous particle. *See, e.g.*, [Page 1616] "The observed multiplicity features of a catalytic pellet in which an isothermal Langmuir-Hinshelwood reaction occurs are sensitive to the distribution of the catalytic components. Specifically, surface migration of the catalytic components may change the range of reactant concentrations or temperatures over which rate multiplicity occurs or shift the maximal rate outside the concentration region in which multiple solutions occur. These changes in the multiplicity features may be used to detect qualitative changes in the metal concentration profile without having to carry out a destructive test." [Page 1616-1617] "The supported metal profile of many industrial precious metal catalysts is nonuniform. This catalytic activity profile may affect considerably the observed catalytic activity. Previous studies have shown that the performance may be optimized by the use of a nonuniform catalytic activity profile. For positive-order reactions, the catalytic activity should be concentrated near the surface to minimize the detrimental influence of intraparticle diffusional limitations (Carberry and Minhas, 1969)."

EXHIBIT G

Invalidity of U.S. Patent No. 11,224,864 in View of *Harold Article*

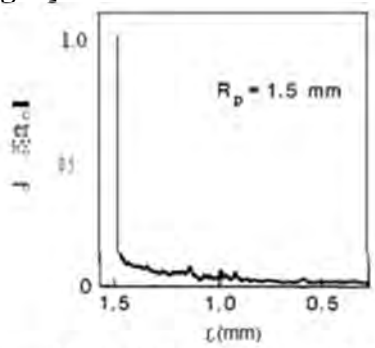
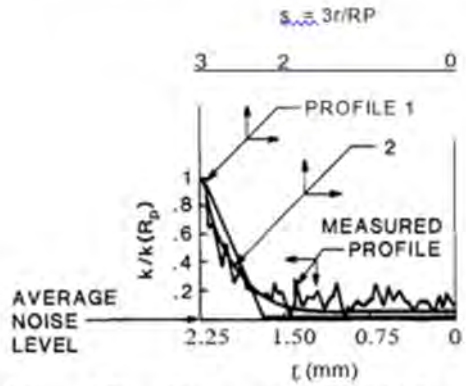
'864 patent	***"Impact of the Catalytic Activity Profile on Observed Multiplicity Features: CO Oxidation on Pt/Al_2O_3" ("Harold Article")***
	[Fig. 1 & Fig. 2]  **Figure 1.** Measured Pt profile (arbitrary units) of a fresh Pt/ Al_2O_3 pellet.  **Figure 2.** Measured Pt profile (in counts) of aged 4.5-mm-diameter Pt/ Al_2O_3 pellet. The three-zone model with $s_1 = 2.25$, $s_2 = 2.97$, $u = 1$ (profile 1; eq 6 and 7) and the polynomial profile (2; eq 8) are also shown.

EXHIBIT G

Invalidity of U.S. Patent No. 11,224,864 in View of *Harold Article*

'864 patent	***"Impact of the Catalytic Activity Profile on Observed Multiplicity Features: CO Oxidation on Pt/Al_2O_3" ("Harold Article")***
	[Page 1617] "These studies show that the observed reaction rate may be increased significantly by placing the catalytic material at a proper distance from the surface. Shyr and Ernst (1980) devised a method of coimpregnation for getting different types of Pt profiles. Previous studies predict that the optimal activity profile is rather sharp, i.e., in the form of a delta or step function. Metal dispersion during impregnation and drying (Foger, 1984) makes it difficult to attain such a sharp metal profile. However, if needed, it is possible to prepare catalysts with rather sharp metal profiles (Shyr and Ernst, 1980). For example, Figure 1 shows a scanning electron microprobe (SEM) analysis of a typical fresh, commercial 0.3% Pt on alumina pellet (3.0-mm diameter) made by OxyCatalyst (division of Met-Pro Corporation). The profile indicates that most of the Pt is contained in a thin surface shell (about 40 μm thick). The remaining Pt is dispersed throughout the inner core. A comparison of the profiles of several fresh pellets from the same batch revealed similar qualitative features. Sintering and metal surface diffusion may alter the metal profile during operation. Figure 2 shows a Pt profile of an aged larger pellet (4.5-mm diameter) from the same batch as the fresh pellet (Figure 1). The aged pellet was used extensively for about a 6-month period to study the multiplicity feature of ethane and carbon monoxide oxidation (Harold and Luss, 1985). The maximal temperature of the pellet in these experiments was 550 °C. The profile reveals that the Pt concentration is a linear decreasing function of the radial distance in a 0.5-mm-thick exterior shell and is distributed uniformly throughout the interior core at a concentration of about 20% of the surface value. This redistribution is most probably due to surface diffusion and migration of Pt atoms. It is of interest to know the sensitivity of the behavioral features of the pellet to changes in the profile.

EXHIBIT G

Invalidity of U.S. Patent No. 11,224,864 in View of *Harold Article*

'864 patent	***"Impact of the Catalytic Activity Profile on Observed Multiplicity Features: CO Oxidation on Pt/Al_2O_3" ("Harold Article")***
	Matsuura and Kato (1967) showed that when a homogeneous reaction with bimolecular Langmuir-Hinshelwood kinetics is carried out in an isothermal CSTR, the conversion can be a multivalued function of the feed concentration. Similarly, the interaction between intraparticle diffusion and a bimolecular Langmuir-Hinshelwood reaction may cause the rate of a single pellet to be a multivalued function of the limiting reactant surface concentration (Elnashaie and Yates, 1973; Wei and Becker, 1974). Hegedus et al. (1977) and Oh et al. (1979) confirmed experimentally the existence of rate multiplicity caused by this interaction during CO oxidation on Pt/Al_2O_3. The bifurcation diagrams of such systems, say of the rate or pellet temperature vs. ambient temperature or limiting reactant concentration, may give useful information about the functional form of the rate expression (Harold et al., 1987a,b). Kunimori et al. (1986) observed recently that the range of temperatures for which multiplicity occurred during the oxidation of CO on Pt/ Al_2O_3 pellets depended on the metal profile." [Page 1617] "We limit the analysis to activity profiles which decrease monotonically from the pellet surface to the center, as in Figures 1 and 2." [Page 1617] "The kinetics of many reactions are described by a bi-molecular Langmuir-Hinshelwood rate expression. The concentration profile of the reactant within a porous, spherical catalytic pellet in which such a reaction is carried out isothermally satisfies the equation

EXHIBIT G

Invalidity of U.S. Patent No. 11,224,864 in View of *Harold Article*

'864 patent	***"Impact of the Catalytic Activity Profile on Observed Multiplicity Features: CO Oxidation on Pt/Al_2O_3" ("Harold Article")***

$$\frac{1}{s^2}\frac{d}{ds}\left(s^2\frac{dx}{ds}\right) - \frac{\phi^2 a(s)x}{(1+Qx)^2} = 0 \qquad (1)$$

$$x = 1 \qquad s = 3 \qquad (2)$$

$$\frac{dx}{ds} = 0 \qquad s = 0 \qquad (3)$$

where

$$x = C/C_s \qquad Q = KC_s \qquad s = \frac{3r}{R_p} \qquad \phi^2 = \frac{R_p^2\bar{k}}{9D_e} \qquad (4)$$

$$a(s) = \frac{k(s)}{\bar{k}} \qquad \bar{k} = \frac{1}{9}\int_0^3 s^2\, k(s)\, ds$$

We assume that the activity is proportional to the active metal concentration and that its profile depends only on the radial position. The analysis can be readily modified to cases in which this assumption is not valid."

[Fig. 3]

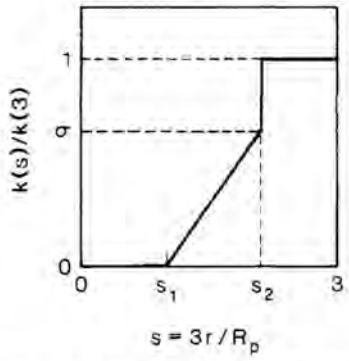


Figure 3. Schematic of three-zone profile (eq 6 and 7).

EXHIBIT G

Invalidity of U.S. Patent No. 11,224,864 in View of *Harold Article*

'864 patent	***"Impact of the Catalytic Activity Profile on Observed Multiplicity Features: CO Oxidation on Pt/Al_2O_3" ("Harold Article")***
	[Page 1618] "Consider a catalytic pellet, the activity of which is described by a three-zone profile (Figure 3) $$a(s) = \left[H(s - s_2) - H(s - 3) + \sigma \frac{s - s_1}{s_2 - s_1}(H(s - s_1) - H(s - s_2)) \right] \Big/ A \quad (6)$$ where $$0 \le s_1 \le s_2 \le 3 \qquad 0 \le \sigma \le 1$$ $$A = \frac{1}{9}\left[\frac{27 - s_2^3}{3} + \frac{\sigma}{s_2 - s_1}\left(\frac{s_2^4 - s_1^4}{4} - \frac{s_1(s_2^3 - s_1^3)}{3} \right) \right] \quad (7)$$ and H is the Heaviside function, i.e., the activity is uniform in an exterior shell of thickness 3 - s_2. The activity decreases linearly from σ at s_2^+ to zero, in an adjacent layer of thickness s_2 - s_1. No activity exists in a central core of radius s_1. The metal profile of the aged catalyst may be fitted by this profile by choosing s_1, s_2, and σ to be 2.25, 2.97, and 1, respectively, as shown in Figure 2. Note that the three-term polynomial fit does not account for the small fluctuations found experimentally and describes an 'averaged' metal profile. Figure 4 describes the dependence of η on Q for several ϕ. The numerical calculations show that η is a unique function of Q for all ϕ smaller than about 15. For larger values of ϕ, η is a multivalued function for a bounded range of Q values. Thus, one expects to find an inverse-S-type hysteresis with changing surface concentrations. Note that the maximum effectiveness factor is within the multiplicity region in all these cases."

EXHIBIT G

Invalidity of U.S. Patent No. 11,224,864 in View of *Harold Article*

'864 patent	***"Impact of the Catalytic Activity Profile on Observed Multiplicity Features: CO Oxidation on Pt/Al_2O_3" ("Harold Article")***
	[Page 1618-1619] "Luss (1971) proved that multiplicity cannot occur within a uniformly active, porous, symmetric pellet for $Q_c < 8$. This limiting value is rather close to $Q_c \approx 11$, found numerically for this specific activity distribution. The cusp-shaped bifurcation set is qualitatively similar to that found for a uniformly active catalytic slab. This is not surprising since for sufficiently large $\phi/(1 + Q)$, the reaction occurs almost entirely near the surface. Figure 6 compares the effectiveness factor vs. Q diagrams for five different catalysts with the same volume-averaged activity ($\phi = 150$). Case 1 is the uniformly active sphere ($s_1 = s_2 = 0$), while case 5 corresponds to a catalyst with all the activity concentrated uniformly in a thin surface shell, the thickness of which is 1% of the pellet radius ($s_1 = s_2 = 2.97$). The activity profiles for cases 2-4 are intermediate within the two extreme cases, 1 and 5. They were obtained by changing s_1 and s_2 in the three-zone model and adjusting the surface activity ($k(3)_i$, i = 2-4) to get $\phi = 150$. The η in each diagram approaches asymptotically $[a(3)]^{1/2}(1 + Q)/\phi$ for large $\phi/(1 + Q)$. A comparison of cases 1-5 indicates that the maximal effectiveness factor attainable for a fixed volume-averaged activity and for activity profiles, which decreases monotonically with distance from the surface, is a step distribution with thickness less than the pellet radius. For activity profiles with vanishing surface activity, the optimal one corresponds to a delta function positioned at some point away from the surface (Morbidelli et al., 1982)."

EXHIBIT G

Invalidity of U.S. Patent No. 11,224,864 in View of *Harold Article*

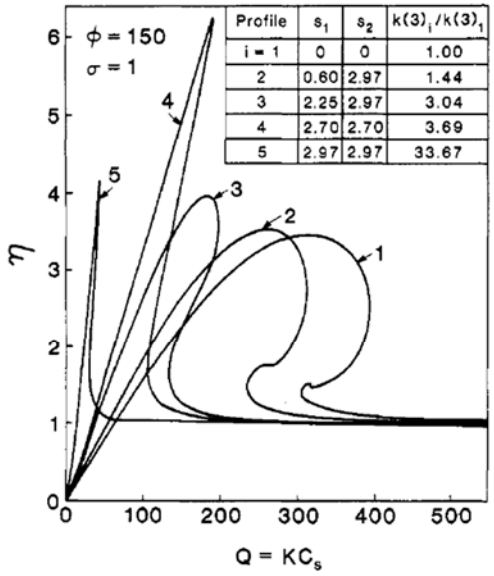
'864 patent	***"Impact of the Catalytic Activity Profile on Observed Multiplicity Features: CO Oxidation on Pt/Al_2O_3" ("Harold Article")***
	 **Figure 6.** Dependence of the effectiveness factor on Q for a fixed volume-averaged activity ($\phi = 150$) for five different activity profiles with values of the surface activity ratio $k(3)_i/k(3)_1$ and s_1 and s_2 shown in the table. [1619-1620] "The simulations show that catalyst effectiveness for a bimolecular Langmuir-Hinshelwood reaction is a sensitive function of the activity distribution. The behavioral features of the effectiveness factor vs. Q (= KC_s) diagrams may be altered by changes in the activity profile. Most notably the position of the maximal activity relative to the multiplicity region is strongly influenced by the activity profile. A maximal η located very close to the extinction is typically found when the activity is concentrated in a thin shell. On the other hand, a maximal η which is well removed from the extinction, or which is located outside of the multiplicity region, is usually found when the activity is distributed throughout the pellet."

EXHIBIT G

Invalidity of U.S. Patent No. 11,224,864 in View of *Harold Article*

'864 patent	***"Impact of the Catalytic Activity Profile on Observed Multiplicity Features: CO Oxidation on Pt/Al_2O_3" ("Harold Article")***
	[Page 1620] "The position of the maximal activity relative to the multiplicity region may provide a useful indication about the activity profile. For example, Figure 10 describes the observed isothermal rate vs. C_s for three different nonuniform pellets. The kinetics were assumed to follow the rate expression $$R = \frac{k_0 \exp(-E/RT)C_s}{[1 + K_0 \exp(-\Delta H_a/R_g T)C_s]^2} \quad (14)$$ using the parameter values of $k_0 = 2.357 \times 10^{11}\ s^{-1}$ $E = 13243$ cal/gmol $K_0 = 1.449 \times 10^7\ cm^3/gmol$ (15) $(-\Delta H_a) = 1561$ cal/gmol determined by Harold and Luss (1987) for the nonisothermal oxidation of CO. The pellet temperature is assumed to be 150 °C which, for R_P = 2 mm and D_e = 0.039 cm^2/s, corresponds to a surface Thiele modulus of $\phi_s = (R_p/3)\,[k(3)/D_e]^{1/2} = 62.1$. Case 1 is for a pellet with a uniformly active surface shell of 13% of the pellet radius and an inert core (ϕ = 36.2). Case 2 is for a pellet with a three-zone activity profile with s_1 = 2.25, s_2 = 2.97, and σ = 1 (ϕ = 35.6). Case 3 is for a pellet with a profile fit to the polynomial expression in eq 8 (ϕ = 36.2). Cases 1 and 3 have the same volume-averaged activity which differs only by a factor of 0.983 from that of case 2. Each profile predicts rate multiplicity; however, the features are quite different. The range of C_s, for which multiple rates exist, decreases and moves to a higher C_s range as the activity is distributed more uniformly. In addition, the C_s corresponding to the maximum rate is outside the multiplicity region for profiles 2 and 3. This feature is consistent with some of the data found by Harold (1985) for CO oxidation on Pt/Al_2O_3. Beusch et al. (1972) also found the maximum rate to be outside the multiplicity region during CO oxidation on a single porous Pt/Al_2O_3 pellet. These data suggest an interaction between negative-order kinetics and pore

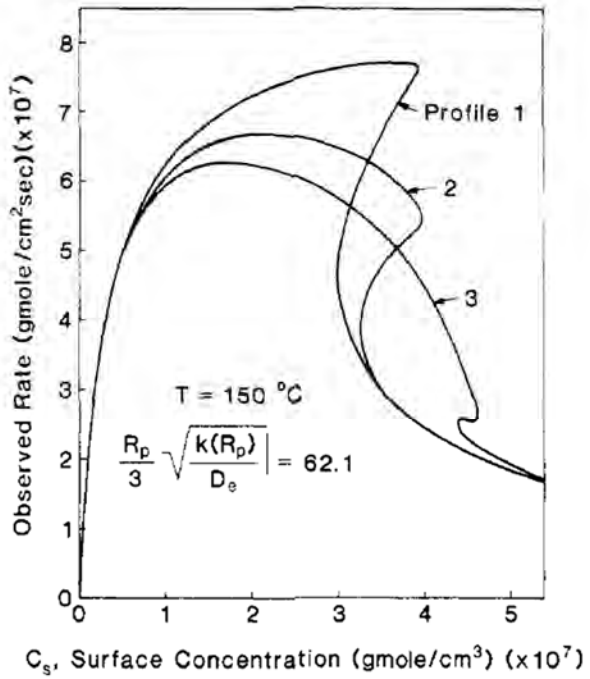
'864 patent	***"Impact of the Catalytic Activity Profile on Observed Multiplicity Features: CO Oxidation on Pt/Al_2O_3" ("Harold Article")***
	diffusion in a pellet with a nonuniform activity. Beusch et al.'s (1972) contention that the multiplicity is due solely to the intrinsic kinetics may be erroneous."  **Figure 10.** Dependence of the observed isothermal rate per unit pellet area on the CO surface concentration for three different activity profiles. Profile 1 corresponds to a uniformly active surface shell with a thickness of $0.13R_p$. Profile 2 is of a three-zone profile with $s_1 = 2.25$, $s_2 = 2.97$, and $\sigma = 1$. Profile 3 is defined by eq 8. [Page 1620] "The simulations show that surface diffusion of the metal may affect significantly the multiplicity features and performance of catalytic pellets. Thus, changes in

EXHIBIT G

Invalidity of U.S. Patent No. 11,224,864 in View of *Harold Article*

'864 patent	***"Impact of the Catalytic Activity Profile on Observed Multiplicity Features: CO Oxidation on Pt/Al_2O_3" ("Harold Article")***
	bifurcation diagrams may serve as an indication of possible changes in the activity profile of the metal. This possibility can then be checked by slicing the pellet and measuring the metal by a SEM. In a recent study of the oxidation of CO on a nonuniformly impregnated Pt/Al_2O_3 catalyst, Kunimori et al. (1986) noted an increase in the range of temperatures for which multiplicity occurred and a reduction in the temperature of the high conversion state. Following Hegedus et al. (1977), they attributed this to pore deposits which increase the diffusional resistance. Our simulations suggest that this behavior may have been caused by surface migration of the dispersed platinum to give a more uniform profile." To the extent that this limitation is not disclosed, either explicitly or inherently, by *Harold Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[3] The combustion promoter according to claim 1, comprising a plurality of microsphere sized porous particles comprising alumina.	*Harold Article* discloses a combustion promoter comprising a plurality of microsphere sized porous particles comprising alumina. *See, e.g.*, The support provided above for Claim 1[b]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Harold Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and

EXHIBIT G

Invalidity of U.S. Patent No. 11,224,864 in View of *Harold Article*

'864 patent	***"Impact of the Catalytic Activity Profile on Observed Multiplicity Features: CO Oxidation on Pt/Al_2O_3" ("Harold Article")***
	predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[6] The combustion promoter according to claim 1, wherein the microsphere sized porous particles collectively have an average (D50) size of between 60 and 90 microns as measured by laser diffraction.	*Harold Article* discloses a combustion promoter comprising microsphere sized porous particles, wherein the microsphere sized porous particles collectively have an average (D50) size of between 60 and 90 microns as measured by laser diffraction. *See, e.g.*, The support provided above for Claim 1[a]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Harold Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[12] The combustion promoter according to claim 1, wherein the one or more Group VIII noble metals comprises one or more of platinum, palladium, iridium, ruthenium, and rhodium.	*Harold Article* discloses a combustion promoter comprising one or more Group VIII noble metals, wherein the one or more Group VIII noble metals comprises one or more of platinum, palladium, iridium, ruthenium, and rhodium. *See, e.g.*, The support provided above for Claim 1[c]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Harold Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and

EXHIBIT G

Invalidity of U.S. Patent No. 11,224,864 in View of *Harold Article*

'864 patent	***"Impact of the Catalytic Activity Profile on Observed Multiplicity Features: CO Oxidation on Pt/Al_2O_3" ("Harold Article")***
	predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[15] The combustion promoter according to claim 12, wherein the one or more Group VIII noble metals comprises palladium.	*Harold Article* discloses a combustion promoter comprising one or more Group VIII noble metals, wherein the one or more Group VIII noble metals comprises palladium. *See, e.g.*, The support provided above for Claim 1[c]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Harold Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.

EXHIBIT H

EXHIBIT H

Invalidity of U.S. Patent No. 11,224,864 in View of UK Patent Application No. GB 2,310,384 ("*Lin*")

UK Patent Application No. GB 2,310,384 ("*Lin*"), filed on February 22, 1996 and published on August 27, 1997, qualifies as prior art under at least 35 U.S.C. §§ 102 and/or 103 for the asserted claims of U.S. Patent No. 11,224,864 ("the '864 patent"), as discussed in detail in the claim chart below.

The Court has not yet construed the claims and therefore the meaning of the terms in the claims has yet to be resolved by the Court. The support identified here for these limitations is responsive to Plaintiffs' infringement contentions, which Defendant disagrees with. As such, nothing in Defendant's claim charts should be construed as an admission regarding infringement, either literally or under the doctrine of equivalents, or as an admission regarding Defendant's understanding of the proper scope of the asserted claims. Defendant reserves the right to rely on additional citations or sources of evidence that also may be applicable, or that may become applicable in light of claim construction, changes in Plaintiffs' infringement contentions, and/or information obtained during discovery as the case progresses.

To the extent Plaintiffs allege that *Lin* does not disclose any particular limitation of the asserted claims of the '864 patent, either expressly or inherently, it would have been obvious to a person of ordinary skill in the art as of the priority date of the '864 patent to modify *Lin* and/or to combine the teachings of *Lin* with other prior art references, including but not limited to the present prior art references cited in the Cover Pleading and the relevant section(s) of claim charts for other prior art references for the '864 patent in a manner that would have rendered the asserted claims invalid as obvious.

Defendant reserves the right to amend or supplement this claim chart at a later date as more fully set forth in the Cover Pleading.

EXHIBIT H

Invalidity of U.S. Patent No. 11,224,864 in View of UK Patent Application No. GB 2,310,384 (“*Lin*”)

’864 patent	UK Patent Application No. GB 2,310,384 (“*Lin*”)
1[a] A CO to CO_2 combustion promoter comprising microsphere sized porous particles, each microsphere sized porous particle having a diameter of less than 1mm, and independently comprising	*Lin* discloses a CO to CO_2 combustion promoter comprising microsphere sized porous particles, each microsphere sized porous particle having a diameter of less than 1mm, and independently comprising the limitations discussed in the following sections. *See, e.g.*, [Abstract] “Disclosed is a new method for preparing supported metal catalysts with eggshell active metal profile. The organometallic compounds with metal as Pd, Ni, Co, Mo, Cu, Pt, Fe, Ag, Ir, Pb, Ti, Sn, V, and Zn, etc., are dissolved in a pure organic solvent, such as benzene, toluene, xylene, methanol, ethanol, and tetrahydrofuran (THF), etc., and/or an organic solvent mixture thereof. The catalyst precursors (i.e., the reactive metal of the prepared organic or inorganic metal concentrated solution) are carried to the surface of MgO, Al_2O_3, SiO_2, TiO_2, ZrO_2, zeolite, active carbon, and polymer support by wet impregnation and/or spray method. The metal concentration profile and metal loading can be precisely controlled by choosing suitable solvent and/or operation conditions.” [Page 1] “The present invention is related to an eggshell catalyst and process for preparation of the same, to precisely control the thickness of single- or multiple-metal eggshell of the finished catalyst and some important properties, such as, metal content and metal dispersion. The porous supports with active metals non-uniformly-distributed have been widely-studied and applied in petroleum refining and petroleum chemical industries currently. They have been theoretically and experimentally proved to own advantages of improving yield and selectivity, reducing thermal sensitivity, resisting poisoning and activity decay, and increasing wear-resistance, etc.” [Page 1-2] “Currently, eggshell catalysts have been widely employed in the industrially selective hydrogenation. such as, acetylene's hydrogenation to ethylene, dienes' hydrogenation to monoenes in gasoline cracking, etc., selective isomerization, such as, α-olefins' conversion to β-olefins, etc., selective oxidation, for example, ethylene's reaction to ethyloxides, oxidation

EXHIBIT H

Invalidity of U.S. Patent No. 11,224,864 in View of UK Patent Application No. GB 2,310,384 (“*Lin*”)

’864 patent	UK Patent Application No. GB 2,310,384 (“*Lin*”)
	of SO_2, oxidation of CO, etc. The yield and selectivity of the desired products of the above reactions can be increased by using eggshell catalysts.” [Page 5] “Example 1: Effect of conversion efficiency. selectivity, and life of eggshell and uniformly distributed Pd/Al_2O_3 catalyst on isoprene selective hydrogenation Step 1: Preparation of 0.2 wt% of Pd - uniformly distributed catalyst Pd/Al_2O_3 0.1523g of $Pd(NH_3)_4$ $(NO_3)_2$ having a Pd purity of 36.04 wt% were dissolved in 50ml of deionized water. To the obtained solution, 10.045g of δ-Al_2O_3 (spherical particles with diameter of 2mm, surface area of 82.4 m^2/g, and pore volume of 0.570 cc/g) were added and then stirred with a shaker at 100 rpm for 3 hours. After filtration, the obtained solid was dried at 100°C for 1 hour and then treated by raising temperature from ambient temperature to 350°C at a rate of 10°C/min and maintained at 350°C for 6 hours.” [Page 8-9] “Example 2: Thickness of Pd-eggshell and Pd content controlled by impregnation time. Step 1: Several 10g portions of δ-Al_2O_3 spherical support (with diameter of 4mm) were separately impregnated in 200ml of over-saturated $Pd(CH_3COO)_2$/toluene solution under stirring of a shaker at 100 rpm. The impregnation time for each portion of the support was controlled respectively to be 0.5, 1, 6, 12, and 24 hours. After filtration, the obtained solid was dried at 100°C for 1 hour and then treated by raising temperature from ambient temperature to 350°C at a rate of 10°C/min and maintained at 350°C for 6 hours.”

EXHIBIT H

Invalidity of U.S. Patent No. 11,224,864 in View of UK Patent Application No. GB 2,310,384 ("*Lin*")

'864 patent	**UK Patent Application No. GB 2,310,384 ("*Lin*")**
	[Page 9] "Example 3: Pd dispersion on Pd-eggshell catalyst controlled by calcining temperature Step 1: 0.534g of $Pd(CH_3COO)_2$ having a Pd purity of 47 wt% was dissolved in 750 ml of toluene. To the resulted solution, 125.24g of δ-Al_2O_3 (spherical particles with diameter of· 2 mm, surface area of 82.4 m^2/g, and pore volume of 0.570 cc/g) were added and then stirred with a shaker at 100 rpm for 3 hours. After filtration, the obtained solid was dried at 100°C for 1 hour." [Page 10] "Example 4: Preparation of catalyst with linear decrease of Pd metal distribution from outer layer toward spherical core Step 1: $Ni(NO_2)_2$-$6H_2O$ was dissolved in deionized water to form a solution of 1M concentration. Spherical γ-Al_2O_3 (with diameter of 4mm, surface area of 173 m^2/g, and porous volume of 0.760 cc/g) was placed in the solution and treated by wet impregnation for 1 hour. Filtered and dried at 110°C for 1 hour." [Page 10-11] "Example 5: Preparation of catalyst by controlling Ni eggshell thickness with impregnation time Step 1: $Ni(N0_3)_2$·6H2O was dissolved in THF solvent to make a solution of 3.0M concentration. Each of an impregnation solution was made by impregnating 1g of spherical support γ-Al_2O_3 (with diameter of 2mm, surface area of 173m^2/g, and porous volume of 0.760 cc/g) in a 5cc of the above Ni-containing solution. The impregnation time was controlled to be 5min., 10min., 20min., and 40min, respectively. Filtered and dried at 120°C for 12 hours."

EXHIBIT H

Invalidity of U.S. Patent No. 11,224,864 in View of UK Patent Application No. GB 2,310,384 ("*Lin*")

'864 patent	**UK Patent Application No. GB 2,310,384 ("*Lin*")**
	[Page 13] "1. A process for the preparation of an eggshell-shaped catalyst for use in selective reduction or oxidation, the process utilising an organo- or an inorgano-metallic compound and comprising the steps of…" To the extent that this limitation is not disclosed, either explicitly or inherently, by *Lin*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
1[b] silica, alumina, or mixtures thereof; and	*Lin* discloses a combustion promoter comprising silica, alumina, or mixtures thereof. *See, e.g.*, [Abstract] "Disclosed is a new method for preparing supported metal catalysts with eggshell active metal profile. The organometallic compounds with metal as Pd, Ni, Co, Mo, Cu, Pt, Fe, Ag, Ir, Pb, Ti, Sn, V, and Zn, etc., are dissolved in a pure organic solvent, such as benzene, toluene, xylene, methanol, ethanol, and tetrahydrofuran (THF), etc., and/or an organic solvent mixture thereof. The catalyst precursors (i.e., the reactive metal of the prepared organic or inorganic metal concentrated solution) are carried to the surface of MgO, Al_2O_3, SiO_2, TiO_2, ZrO_2, zeolite, active carbon, and polymer support by wet impregnation and/or spray method. The metal concentration profile and metal loading can be precisely controlled by choosing suitable solvent and/or operation conditions." [Page 2] "H_2PtCl_6 and citric acid are co-impregnated to produce various nonuniformly distributed Pt on spherical alumina support and the experimental error of reproducibility is about 10% [see USP Nos.3,259,454 and 3,259,589 (July 5, 1966); E.R. Becker et al., in 'Preparation of catalysts II :Proceedings of the 2nd international Sympogereous Catalysts', Elsevier, Amsterdam, 1979,

’864 patent	UK Patent Application No. GB 2,310,384 (“*Lin*”)
	P159]. The precursors of CrO_3, $(NH_4)_2Cr_2O_7$, and $Cu(NO_3)_2$, etc. are used by means of various impregnation technologies to produce non-uniformly distributed Cr and Cu metal in different state on spherical Al_2O_3 support [see J. Catal., 43, (1976) 200]. Platinum can be modified into nine different states non-uniformly distributed on spherical Al_2O_3 support by adding salts of HCl, NaBr, and the like to an impregnating solution of H_2PtCl_6 [see J. Catal., 63, (1980) 425].” [Page 3] “Accordingly, the present invention is a process for preparation of eggshell metal catalysts. Said process utilizes organo- or inorgano-metallic compounds, in which the metal preferably comprises Pd, Ni, Co, Mo, Cu, Pt, Fe, Ag, Ir, Pb, Re, Ti, Sn, V, and Zn, etc., by dissolving in organic solvents, such as aromatic solvents, e.g., toluene, benzene, and xylenes, alcohols, eg. methanol, and tetrahydrofuran (THF), etc. or in a variety of mixtures thereof under heating or at ambient temperature. The active metal in the prepared organo- or inorgano-metallic concentrate solution is attached to a support by wet impregnation or spray method, in which the support comprises Al_2O_3, SiO_2, zeolite, TiO_2, ZrO_2, MgO, active carbon, polymer or the mixtures thereof. The process of preparation according to the present invention can employ different operating variables, for example, Al_2O_3 be calcined at different temperatures for changing its surface area and acidity, or, the impregnated catalyst may be calcined at different temperatures, or the type and the concentration of the impregnating solution may be varied, etc. so as to precisely control the thickness of single- or multiple-metal eggshell of the finished catalyst and some important properties, such as, metal content and metal dispersion.” [Page 5] “Fig. 14 shows an EPMA spectrum of 0.2 wt% Pd/Al_2O_3 catalyst calcined at 500°C. Fig. 15 shows a temperature programmed reduction (TPR) spectrum of 0.2 wt% Pd/Al_2O_3 catalyst calcined at various temperatures (a)300°C, (b)400°C, (c)500°C, (d)600°C, and (e)700°C.

EXHIBIT H

Invalidity of U.S. Patent No. 11,224,864 in View of UK Patent Application No. GB 2,310,384 (“*Lin*”)

’864 patent	UK Patent Application No. GB 2,310,384 (“*Lin*”)
	Fig. 16 shows a plot of Pd metal distribution vs. calcining temperature of 0.2 wt% Pd/Al_2O_3 catalyst.” [Page 5-6] “Example 1: Effect of conversion efficiency. selectivity, and life of eggshell and uniformly distributed Pd/Al_2O_3 catalyst on isoprene selective hydrogenation Step 1: Preparation of 0.2 wt% of Pd - uniformly distributed catalyst Pd/Al_2O_3 0.1523g of $Pd(NH_3)_4 (NO_3)_2$ having a Pd purity of 36.04 wt% were dissolved in 50ml of deionized water. To the obtained solution, 10.045g of δ-Al_2O_3 (spherical particles with diameter of 2mm, surface area of 82.4 m^2/g, and pore volume of 0.570 cc/g) were added and then stirred with a shaker at 100 rpm for 3 hours. After filtration, the obtained solid was dried at 100°C for 1 hour and then treated by raising temperature from ambient temperature to 350°C at a rate of 10°C/min and maintained at 350°C for 6 hours. The obtained catalyst sample has 0.2 wt% of Pd content from elementary analysis and Pd distribution is uniform resulted from an EPMA analysis as shown in Fig. 1. Step 2: Preparation of 0.2 wt% of Pd eggshell catalyst Pd/Al_2O_3 0.1068g of $Pd(CH_3COO)_2$ having a Pd purity of 47wt% were dissolved in 150ml of toluene. To the obtained solution, 25.048g of δ - Al_2O_3 were added and followed by the same treatment as described in step 1. The obtained catalyst sample has an eggshell distribution of Pd metal from an EPMA analysis as shown in Fig. 2.”

EXHIBIT H

Invalidity of U.S. Patent No. 11,224,864 in View of UK Patent Application No. GB 2,310,384 ("*Lin*")

'864 patent	**UK Patent Application No. GB 2,310,384 ("*Lin*")**
	[Page 8-9] "Example 2: Thickness of Pd-eggshell and Pd content controlled by impregnation time. Step 1: Several 10g portions of δ-Al_2O_3 spherical support (with diameter of 4mm) were separately impregnated in 200ml of over-saturated $Pd(CH_3COO)_2$/toluene solution under stirring of a shaker at 100 rpm. The impregnation time for each portion of the support was controlled respectively to be 0.5, 1, 6, 12, and 24 hours. After filtration, the obtained solid was dried at 100°C for 1 hour and then treated by raising temperature from ambient temperature to 350°C at a rate of 10°C/min and maintained at 350°C for 6 hours." [Page 9] "Example 3: Pd dispersion on Pd-eggshell catalyst controlled by calcining temperature Step 1: 0.534g of $Pd(CH_3COO)_2$ having a Pd purity of 47 wt% was dissolved in 750 ml of toluene. To the resulted solution, 125.24g of δ-Al_2O_3 (spherical particles with diameter of 2 mm, surface area of 82.4 m^2/g, and pore volume of 0.570 cc/g) were added and then stirred with a shaker at 100 rpm for 3 hours. After filtration, the obtained solid was dried at 100°C for 1 hour." [Page 10] "Example 4: Preparation of catalyst with linear decrease of Pd metal distribution from outer layer toward spherical core Step 1: $Ni(NO_2)_2 \cdot 6H_2O$ was dissolved in deionized water to form a solution of IM concentration. Spherical γ-Al_2O_3 (with diameter of 4mm, surface area of 173 m^2/g, and porous volume of 0.760 cc/g) was placed in the solution and treated by wet impregnation for 1 hour. Filtered and dried at 110 °C for 1 hour."

EXHIBIT H

Invalidity of U.S. Patent No. 11,224,864 in View of UK Patent Application No. GB 2,310,384 (“*Lin*”)

’864 patent	UK Patent Application No. GB 2,310,384 (“*Lin*”)
	[Page 10-11] “Example 5: Preparation of catalyst by controlling Ni eggshell thickness with impregnation time Step 1: $Ni(NO_3)_2 \cdot 6H2O$ was dissolved in THF solvent to make a solution of 3.0M concentration. Each of an impregnation solution was made by impregnating 1g of spherical support γ-Al_2O_3 (with diameter of 2mm, surface area of 173m^2/g, and porous volume of 0.760 cc/g) in a 5cc of the above Ni-containing solution. The impregnation time was controlled to be 5min., 10min., 20min., and 40min, respectively. Filtered and dried at 120°C for 12 hours.” [Page 11] “It can be seen from the foregoing that a new method is disclosed for preparing supported metal catalysts with eggshell active metal profile. The inorgano- or organometallic compounds, preferably comprising Pd, Ni, Co, Mo, Cu, Pt, Fe, Ag, Ir, Pb, Ti, Sn, V, and Zn, etc. are dissolved in an organic solvent, such as benzene, toluene, xylene, methanol, ethanol, and tetrahydrofuran (THF), etc. and/or an organic solvent mixture thereof. The catalyst precursors (i.e. the reactive metal of the prepared organic or inorganic metal concentrated solution) are carried to the surface of MgO, Al_2O_3, SiO_2, TiO_2, ZrO_2, zeolite, active carbon, and polymer support by wet impregnation and/or spray method. The metal concentration profile and metal loading can be precisely controlled by choosing suitable solvent and/or operation conditions.” To the extent that this limitation is not disclosed, either explicitly or inherently, by *Lin*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the ’864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.

EXHIBIT H

Invalidity of U.S. Patent No. 11,224,864 in View of UK Patent Application No. GB 2,310,384 (“*Lin*”)

’864 patent	UK Patent Application No. GB 2,310,384 (“*Lin*”)
1[c] one or more Group VIII noble metals distributed in the particle as an eggshell such that a higher concentration of the one or more Group VIII noble metals is present in the outer region of the microsphere sized porous particle as compared to the concentration of the one or more Group VIII noble metals in the centre of the microsphere sized porous particle.	*Lin* discloses a combustion promoter comprising one or more Group VIII noble metals, the metals distributed in the particle as an eggshell such that a higher concentration of the one or more Group VIII noble metals is present in the outer region of the microsphere sized porous particle as compared to the concentration of the one or more Group VIII noble metals in the centre of the microsphere sized porous particle. *See, e.g.*, [Abstract] “Disclosed is a new method for preparing supported metal catalysts with eggshell active metal profile. The organometallic compounds with metal as Pd, Ni, Co, Mo, Cu, Pt, Fe, Ag, Ir, Pb, Ti, Sn, V, and Zn, etc., are dissolved in a pure organic solvent, such as benzene, toluene, xylene, methanol, ethanol, and tetrahydrofuran (THF), etc., and/or an organic solvent mixture thereof. The catalyst precursors (i.e., the reactive metal of the prepared organic or inorganic metal concentrated solution) are carried to the surface of MgO, Al_2O_3, SiO_2, TiO_2, ZrO_2, zeolite, active carbon, and polymer support by wet impregnation and/or spray method. The metal concentration profile and metal loading can be precisely controlled by choosing suitable solvent and/or operation conditions.”

EXHIBIT H

Invalidity of U.S. Patent No. 11,224,864 in View of UK Patent Application No. GB 2,310,384 (“*Lin*”)

’864 patent	UK Patent Application No. GB 2,310,384 (“*Lin*”)
	[Fig. 1 & Fig. 2]

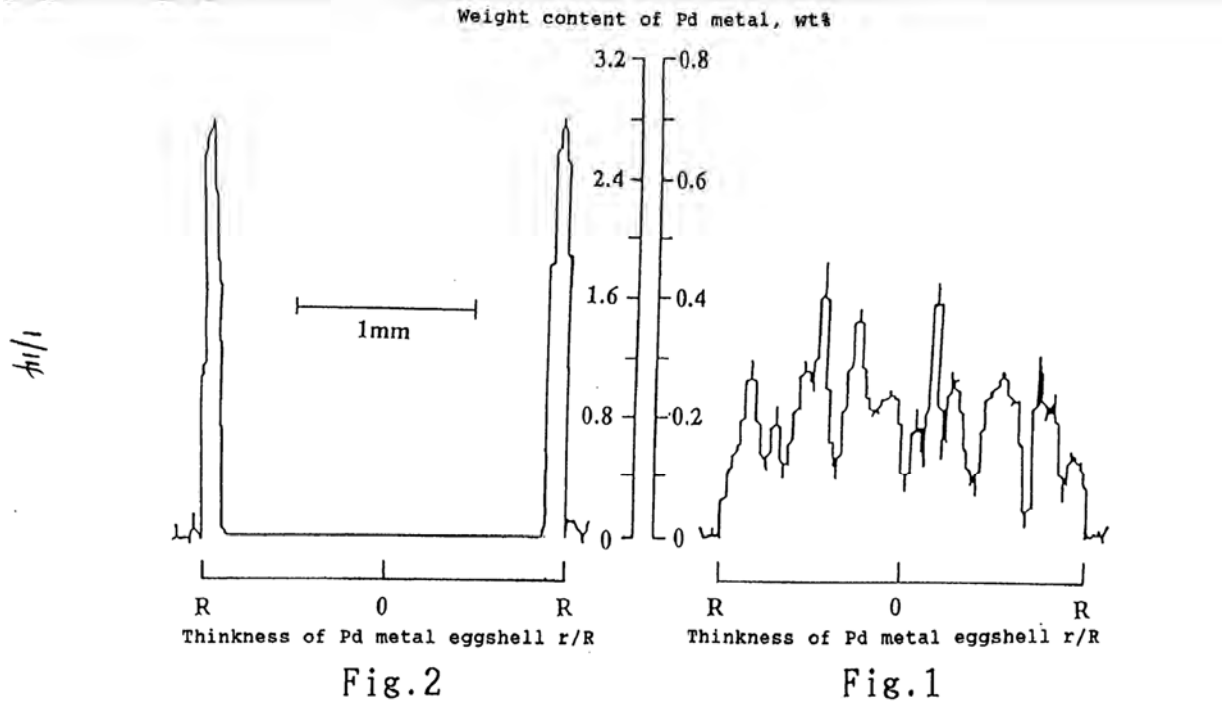


EXHIBIT H

Invalidity of U.S. Patent No. 11,224,864 in View of UK Patent Application No. GB 2,310,384 (“*Lin*”)

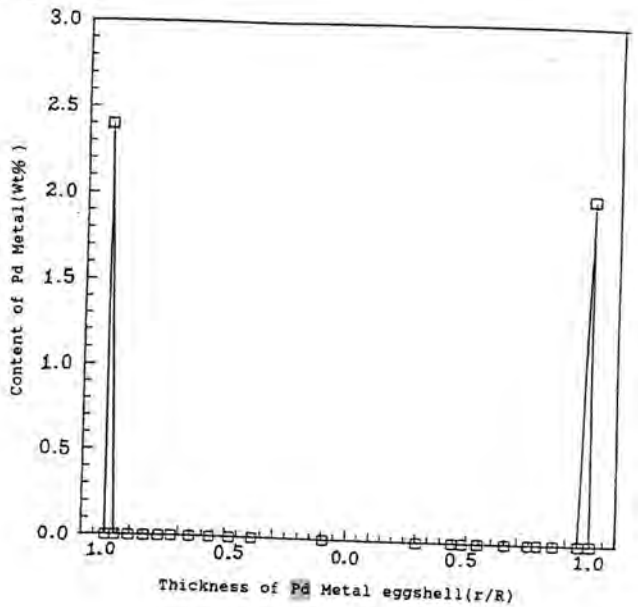
’864 patent	UK Patent Application No. GB 2,310,384 (“*Lin*”)
	[Fig. 14]  Fig.14

EXHIBIT H

Invalidity of U.S. Patent No. 11,224,864 in View of UK Patent Application No. GB 2,310,384 (“*Lin*”)

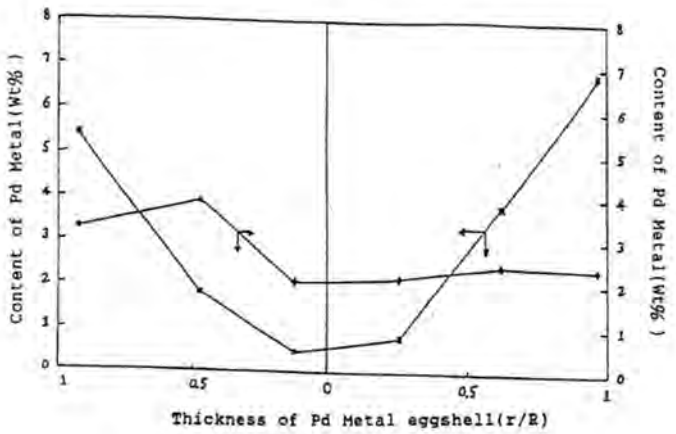
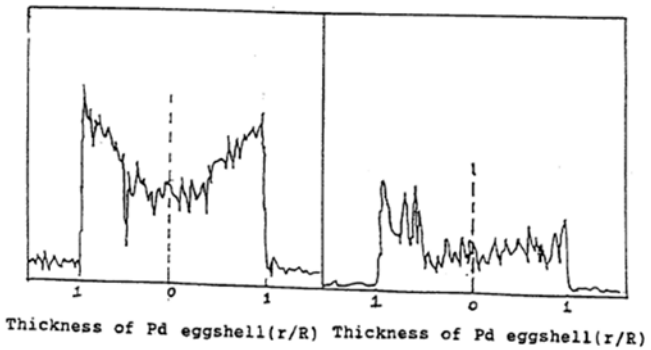
’864 patent	UK Patent Application No. GB 2,310,384 (“*Lin*”)
	[Fig. 17]  Fig.17(a)  Fig.17(b)

EXHIBIT H

Invalidity of U.S. Patent No. 11,224,864 in View of UK Patent Application No. GB 2,310,384 (“*Lin*”)

’864 patent	UK Patent Application No. GB 2,310,384 (“*Lin*”)
	[Page 1-2] “The present invention is related to an eggshell catalyst and process for preparation of the same, to precisely control the thickness of single- or multiple-metal eggshell of the finished catalyst and some important properties, such as, metal content and metal dispersion. The porous supports with active metals non-uniformly-distributed have been widely-studied and applied in petroleum refining and petroleum chemical industries currently. They have been theoretically and experimentally proved to own advantages of improving yield and selectivity, reducing thermal sensitivity, resisting poisoning and activity decay, and increasing wear-resistance, etc. According to the description in Ind. Eng. Chem. Prod. Res. Dev. 1981, Vol.20, p439, non-uniformly active metal distribution can be classified into four types. 1. Metal is uniformly distributed, which is usually used in reactions with catalyst having fair activity and no mass transfer resistance. 2. Metal is distributed on the outer layer of the support (with eggshell shape), which is usually used in fast reactions and capable of improving the selectivity and yield of the desired products. 3. Metal is distributed inside the support (with eggyolk shape), which is usually used in a fluidized bed or a moving bed and the outer layer of the catalyst may easily show wear or poisoning. 4. Metal is distributed between eggshell shape and yolk shape, as egg white shape, which is suitable for a reaction range between those conducted by the eggshell- and the eggyolk shaped catalyst, with mass transfer resistance inside the particle and wear or poisoning easily shown on the outer layer. Currently, eggshell catalysts have been widely employed in the industrially selective hydrogenation, such as, acetylene's hydrogenation to ethylene, dienes' hydrogenation to

EXHIBIT H

Invalidity of U.S. Patent No. 11,224,864 in View of UK Patent Application No. GB 2,310,384 (“*Lin*”)

’864 patent	UK Patent Application No. GB 2,310,384 (“*Lin*”)
	monoenes in gasoline cracking, etc., selective isomerization, such as, α-olefins' conversion to β-olefins, etc., selective oxidation, for example, ethylene's reaction to ethyloxides, oxidation of SO_2, oxidation of CO, etc. The yield and selectivity of the desired products of the above reactions can be increased by using eggshell catalysts. In particular for consecutive reactions A →B →C, it is verified (see Chem. Eng. Sci., 27, (1972) 227; Supra, 29, (1974) 1473) that the yield and selectivity of product B can be increased by eggshell catalyst. According to the disclosure from J. of Chem. Eng. of Japan, 22(3), (1989) 287, it is verified that the order of ethylene selectivity in the selective hydrogenation of acetylene with major product of ethylene depends upon the metal distribution on catalyst Ni/Al_20_3 as the following: eggshell of thin layer> eggshell of thick layer> uniform distribution> eggyolk distribution” [Page 3] “Accordingly, the present invention is a process for preparation of eggshell metal catalysts. Said process utilizes organo- or inorgano-metallic compounds, in which the metal preferably comprises Pd, Ni, Co, Mo, Cu, Pt, Fe, Ag, Ir, Pb, Re, Ti, Sn, V, and Zn, etc., by dissolving in organic solvents, such as aromatic solvents, e.g., toluene, benzene, and xylenes, alcohols, e.g. methanol, and tetrahydrofuran (THF), etc. or in a variety of mixtures thereof under heating or at ambient temperature. The active metal in the prepared organo or inorgano-metallic concentrate solution is attached to a support by wet impregnation or spray method, in which the support comprises Al_2O_3, SiO_2, zeolite, TiO_2, ZrO_2, MgO, active carbon, polymer or the mixtures thereof. The process of preparation according to the present invention can employ different operating variables, for example, Al_2O_3 maybe calcined at different temperatures for changing its surface area and acidity, or, the impregnated catalyst may be calcined at different temperatures, or the type and the concentration of the impregnating solution may be varied, etc. so as to precisely control the thickness of single- or multiple-metal eggshell of the finished catalyst and some important properties, such as, metal content and metal dispersion.”

EXHIBIT H

Invalidity of U.S. Patent No. 11,224,864 in View of UK Patent Application No. GB 2,310,384 (“*Lin*”)

’864 patent	UK Patent Application No. GB 2,310,384 (“*Lin*”)
	[Page 3-4] “Fig. 1 shows a result of Pd metal distribution from EPMA (electron probe microanalyser) of a uniform catalyst (X-axis : thickness of Pd metal eggshell, r/R; Y-axis : weight content of Pd metal, wt%). Fig. 2 shows a result of Pd metal distribution from EPMA of an eggshell catalyst (X-axis: thickness of Pd metal eggshell, r/R; Y-axis : weight content of Pd metal, wt%).” [Page 5] “Fig. 11 shows a result of Pd metal distribution from EPMA of the catalysts obtained after different impregnation time (a) time =0.5h, (b) time = 1h, (c) time =3h, (d) time =6h, (e) time =12h, and (f) time =24h. Fig. 12 shows a variation of the thickness of Pd metal eggshell vs. impregnation time. Fig. 13 shows a variation of the content of Pd metal vs. impregnation time. Fig. 14 shows an EPMA spectrum of 0.2 wt% Pd/Al_2O_3 catalyst calcined at 500°C. Fig. 15 shows a temperature programmed reduction (TPR) spectrum of 0.2 wt% Pd/Al_2O_3 catalyst calcined at various temperatures (a) 300°C, (b) 400°C, (c) 500°C, (d) 600°C, and (e) 700°C. Fig. 16 shows a plot of Pd metal distribution vs. calcining temperature of 0.2 wt% Pd/Al_2O_3 catalyst. Fig. 17a & Fig. 17b shows an EPMA spectrum of metal distribution of Pd and Ni eggshell. Fig. 18 shows an EPMA spectrum of Ni metal distribution; (a) 5min, (b) 10min, (c)20min, and (d) 40min.”

EXHIBIT H

Invalidity of U.S. Patent No. 11,224,864 in View of UK Patent Application No. GB 2,310,384 (“*Lin*”)

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	[Page 5-6] “Example 1: Effect of conversion efficiency. selectivity, and life of eggshell and uniformly distributed Pd/Al_2O_3 catalyst on isoprene selective hydrogenation Step 1: Preparation of 0.2 wt% of Pd - uniformly distributed catalyst Pd/Al_2O_3 0.1523g of $Pd(NH_3)_4 (NO_3)_2$ having a Pd purity of 36.04 wt% were dissolved in 50ml of deionized water. To the obtained solution, 10.045g of δ-Al_2O_3 (spherical particles with diameter of 2mm, surface area of 82.4 m^2/g, and pore volume of 0.570 cc/g) were added and then stirred with a shaker at 100 rpm for 3 hours. After filtration, the obtained solid was dried at 100°C for 1 hour and then treated by raising temperature from ambient temperature to 350°C at a rate of 10°C/min and maintained at 350°C for 6 hours. The obtained catalyst sample has 0.2 wt% of Pd content from elementary analysis and Pd distribution is uniform resulted from an EPMA analysis as shown in Fig. 1. Step 2: Preparation of 0.2 wt% of Pd eggshell catalyst Pd/Al_2O_3 0.1068g of $Pd(CH_3COO)_2$ having a Pd purity of 47wt% were dissolved in 150ml of toluene. To the obtained solution, 25.048g of δ - Al_2O_3 were added and followed by the same treatment as described in step 1. The obtained catalyst sample has an eggshell distribution of Pd metal from an EPMA analysis as shown in Fig. 2.” [Page 8-9] “Example 2: Thickness of Pd-eggshell and Pd content controlled by impregnation time. Step 1: Several 10g portions of δ-Al_2O_3 spherical support (with diameter of 4mm) were separately impregnated in 200ml of over-saturated $Pd(CH_3COO)_2$/toluene solution under stirring of a shaker at 100 rpm. The impregnation time for each portion of the support was controlled respectively to be 0.5, 1, 6, 12, and 24 hours. After filtration, the obtained solid was

’864 patent	UK Patent Application No. GB 2,310,384 (“*Lin*”)
	dried at 100°C for 1 hour and then treated by raising temperature from ambient temperature to 350°C at a rate of 10°C/min and maintained at 350°C for 6 hours. Step 2: The obtained catalyst samples were analyzed by EPMA after different impregnation time. The measured Pd-metal distribution is shown in Fig. 11. Fig. 12 shows a plot of Pd-eggshell thickness (r/R) adopted from Fig. 11 vs. the impregnation time, in which the longer impregnation time causes the thicker Pd-eggshell thickness. Fig. 13 shows a plot of Pd wt% vs. the impregnation time and longer impregnation time causes higher Pd content.” [Page 9] “Example 3: Pd dispersion on Pd-eggshell catalyst controlled by calcining temperature Step 1: 0.534g of $Pd(CH_3COO)_2$ having a Pd purity of 47 wt% was dissolved in 750 ml of toluene. To the resulted solution, 125.24g of o-Al_2O_3 (spherical particles with diameter of 2 mm, surface area of 82.4 m^2/g, and pore volume of 0.570 cc/g) were added and then stirred with a shaker at 100 rpm for 3 hours. After filtration, the obtained solid was dried at 100°C for 1 hour. Step 2: The dried catalyst sample was separated into five aliquot portions. Each portion was independently heated from ambient temperature to 300°C, 400°C, 500°C, 600°C, and 700°C at an elevating rate of 10°C/min and then maintained at final temperature for 6 hours. The resulted catalyst sample has 0.2 wt% of Pd content via elementary analysis and obtain a uniform Pd distribution on the outer layer of supports with a Pd-eggshell thickness (r/R) of about 0.05, which is not changed due to a variation of the calcining temperature, via EPMA analysis. The Pd content of catalyst sample calcinated at 500°C was quantitatively determined via 21 points of EPMA data and the result is shown in Fig. 14.”

EXHIBIT H

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'864 patent	**UK Patent Application No. GB 2,310,384 ("*Lin*")**
	[Page 10] "Example 4: Preparation of catalyst with linear decrease of Pd metal distribution from outer layer toward spherical core Step 1: $Ni(NO_2)_2 \cdot 6H_2O$ was dissolved in deionized water to form a solution of 1M concentration. Spherical y-Al_2O_3 (with diameter of 4mm, surface area of 173 m^2/g, and porous volume of 0.760 cc/g) was placed in the solution and treated by wet impregnation for 1 hour. Filtered and dried at 110°C for 1 hour. Step 2: The resulting catalyst sample was placed in a solution of over-saturated $Pd(CH_3COO)_2$/toluene and treated by wet impregnation for 0.5 hour. Then, filtered and dried at 110°C for 1 hour. Step 3: The resulting catalyst sample was subject to an EPMA analysis for determination of Ni and Pd metal distribution. The obtained spectrum is shown in figure 17. Obviously, the Pd metal distribution exhibits a linear decrease from outer layer toward spherical core while the Ni metal distribution is uniform." [Page 11] "It can be seen from the foregoing that a new method is disclosed for preparing supported metal catalysts with eggshell active metal profile. The inorgano- or organometallic compounds, preferably comprising Pd, Ni, Co, Mo, Cu, Pt, Fe, Ag, Ir, Pb, Ti, Sn, V, and Zn, etc. are dissolved in an organic solvent, such as benzene, toluene, xylene, methanol, ethanol, and tetrahydrofuran (THF), etc. and/or an organic solvent mixture thereof. The catalyst precursors (i.e. the reactive metal of the prepared organic or inorganic metal concentrated solution) are carried to the surface of MgO, Al_2O_3, SiO_2, TiO_2, ZrO_2, zeolite, active carbon, and polymer support by wet impregnation and/or spray method. The metal concentration profile and metal loading can be precisely controlled by choosing suitable solvent and/or operation conditions."

EXHIBIT H

Invalidity of U.S. Patent No. 11,224,864 in View of UK Patent Application No. GB 2,310,384 ("*Lin*")

'864 patent	**UK Patent Application No. GB 2,310,384 ("*Lin*")**
	To the extent that this limitation is not disclosed, either explicitly or inherently, by *Lin*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[3] The combustion promoter according to claim 1, comprising a plurality of microsphere sized porous particles comprising alumina.	*Lin* discloses a combustion promoter comprising a plurality of microsphere sized porous particles comprising alumina. *See, e.g.*, The support provided above for Claim 1[b]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Lin*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[6] The combustion promoter according to claim 1, wherein the microsphere sized porous particles collectively have an average (D50) size of between 60 and 90 microns as measured by laser diffraction.	*Lin* discloses a combustion promoter comprising microsphere sized porous particles, wherein the microsphere sized porous particles collectively have an average (D50) size of between 60 and 90 microns as measured by laser diffraction. *See, e.g.*, The support provided above for Claim 1[a]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Lin*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from

EXHIBIT H

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'864 patent	**UK Patent Application No. GB 2,310,384 ("*Lin*")**
	known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[12] The combustion promoter according to claim 1, wherein the one or more Group VIII noble metals comprises one or more of platinum, palladium, iridium, ruthenium, and rhodium.	*Lin* discloses a combustion promoter comprising one or more Group VIII noble metals, wherein the one or more Group VIII noble metals comprises one or more of platinum, palladium, iridium, ruthenium, and rhodium. *See, e.g.*, The support provided above for Claim 1[c]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Lin*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[15] The combustion promoter according to claim 12, wherein the one or more Group VIII noble metals comprises palladium.	*Lin* discloses a combustion promoter comprising one or more Group VIII noble metals, wherein the one or more Group VIII noble metals comprises palladium. *See, e.g.*, The support provided above for Claim 1[c]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Lin*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.

EXHIBIT I

EXHIBIT I

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2007/0197379 ("*Miura*")

U.S. Patent Application No. 2007/0197379 ("*Miura*"), filed on April 27, 2004 and published on August 23, 2007, qualifies as prior art under at least 35 U.S.C. §§ 102 and/or 103 for the asserted claims of U.S. Patent No. 11,224,864 ("the '864 patent"), as discussed in detail in the claim chart below.

The Court has not yet construed the claims and therefore the meaning of the terms in the claims has yet to be resolved by the Court. The support identified here for these limitations is responsive to Plaintiffs' infringement contentions, which Defendant disagrees with. As such, nothing in Defendant's claim charts should be construed as an admission regarding infringement, either literally or under the doctrine of equivalents, or as an admission regarding Defendant's understanding of the proper scope of the asserted claims. Defendant reserves the right to rely on additional citations or sources of evidence that also may be applicable, or that may become applicable in light of claim construction, changes in Plaintiffs' infringement contentions, and/or information obtained during discovery as the case progresses.

To the extent Plaintiffs allege that *Miura* does not disclose any particular limitation of the asserted claims of the '864 patent, either expressly or inherently, it would have been obvious to a person of ordinary skill in the art as of the priority date of the '864 patent to modify *Miura* and/or to combine the teachings of *Miura* with other prior art references, including but not limited to the present prior art references cited in the Cover Pleading and the relevant section(s) of claim charts for other prior art references for the '864 patent in a manner that would have rendered the asserted claims invalid as obvious.

Defendant reserves the right to amend or supplement this claim chart at a later date as more fully set forth in the Cover Pleading.

EXHIBIT I

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2007/0197379 ("*Miura*")

'864 patent	**U.S. Patent Application No. 2007/0197379 ("*Miura*")**
1[a] A CO to CO_2 combustion promoter comprising microsphere sized porous particles, each microsphere sized porous particle having a diameter of less than 1mm, and independently comprising	*Miura* discloses a CO to CO_2 combustion promoter comprising microsphere sized porous particles, each microsphere sized porous particle having a diameter of less than 1mm, and independently comprising the limitations discussed in the following sections. *See, e.g.*, [Abstract] "The present invention provides an exhaust gas purifying catalyst and a process for producing a metal oxide particle comprising multiple species of metal oxides and capable of satisfactorily exerting the properties of respective metal oxides. A process of the present invention comprises providing a sol containing at least a population of first colloid particles and a population of second colloid particles differing in the isoelectric point with each other; adjusting the pH of the sol to be closer to the isoelectric point of the population of first colloid particles than to the isoelectric point of the population of second colloid particles, thereby aggregating the population of first colloid particles; adjusting the pH of the sol, thereby aggregating the population of second colloid particles onto the population of first colloid particles aggregated; and drying and firing the obtained aggregate." [0001] "The present invention relates to a process for producing a metal oxide particle and, more specifically, the present invention relates to a process for producing a metal oxide particle preferably used as an exhaust gas purifying catalyst by loading a noble metal thereon. Further, present invention relates to an exhaust gas purifying catalyst for an internal combustion engine." [0002]-[0005] "The exhaust gas from internal combustion engines, such as automobile engines, contains nitrogen oxide (NO_x), carbon monoxide (CO), hydrocarbon (HC) and the like. These substances can be purified by an exhaust gas purifying catalyst capable of oxidizing CO and HC and, at the same time, reducing NO_x. As for representative

EXHIBIT I

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2007/0197379 ("*Miura*")

'864 patent	U.S. Patent Application No. 2007/0197379 ("*Miura*")
	exhaust gas purifying catalysts, three-way catalysts where a noble metal such as platinum (Pt), rhodium (Rh) and palladium (Pd) is supported on a porous metal oxide support such as γ-alumina are known. The metal oxide support may be formed of various materials but, in order to obtain a large surface area, alumina (Al_2O_3) has been heretofore generally used. However, in recent years, for accelerating purification of the exhaust gas by using chemical properties of the support, it has been proposed to use various other materials such as ceria (CeO_2), zirconia (ZrO_2) and titanium (TiO_2) in combination with, or not in combination with, alumina. For example, in order to alleviate the fluctuation of oxygen concentration in the exhaust gas and thereby increase the exhaust gas purifying ability of the three-way catalyst, a material having an oxygen storage capacity (OSC) for storing oxygen when the oxygen concentration in the exhaust gas is high, and releasing oxygen when the oxygen concentration in the exhaust gas is low, is used as a support of the exhaust gas purifying catalyst. A representative material having OSC is ceria. In order to allow for efficient progress of oxidation of CO and HC and reduction of NO_x by the activity of the three-way catalyst, the air-fuel ratio in the internal combustion engine must be a theoretical air-fuel ratio (stoichiometric air-fuel ratio). Therefore, the fluctuation of oxygen concentration in the exhaust gas is preferably alleviated by a material having OSC to maintain the oxygen concentration in the vicinity of the theoretical air-fuel ratio, so that the three-way catalyst can exert its exhaust gas purifying ability. Furthermore, according to recent studies, it has been found that ceria not only has OSC but also, by virtue of its strong affinity for a noble metal, particularly platinum, can prevent particle growth (sintering) of the noble metal supported thereon."

EXHIBIT I

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2007/0197379 ("*Miura*")

'864 patent	U.S. Patent Application No. 2007/0197379 ("*Miura*")
	[0014]-[0015] "According to the process of the present invention, a metal oxide particle comprising a core part relatively rich in a component originated in the population of first colloid particles and a surface layer relatively rich in a component originated in the population of second colloid particles can be obtained. Furthermore, according to the process of the present invention, a metal oxide particle having any particle diameter and having a structure comprising a core part and a surface layer can be obtained. For example, when the population of colloid particles used as a raw material in practice has an average particle diameter of about 5 nm, the average particle diameter of the metal oxide particle produced by the process of the present invention can be made to be 50 nm or less. Accordingly, this metal oxide particle can have an average particle diameter of, for example, less than 10 μm, less than 5 μm, less than 1 μm, less than 500 nm, less than 200 nm, less than 100 nm or less than 50 nm." [0009] "Also, a substantially uniform metal oxide particle may be obtained from a sol in which multiple different species of colloid particles are mixed, but a uniform mixture does not always yield the best result." [0068]-[0070] "As shown in FIG. 2, in an optimum aspect of the exhaust gas purifying catalyst of the present invention, CeO_2 constituting a surface layer 12 of particulate support covers an entire core part 22 formed of ZrO_2 in a proper thickness, and a noble metal 13 (Pt) is supported on the particulate support. Furthermore, the particulate support preferably has a small particle diameter. It was confirmed that the exhaust gas purifying catalyst comprising the particulate support and the noble metal 3 supported thereon had an excellent exhaust gas purifying performance. The

EXHIBIT I

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2007/0197379 ("*Miura*")

'864 patent	U.S. Patent Application No. 2007/0197379 ("*Miura*")
	same effect is exerted in a monolith catalyst obtained by coating the particulate support on a honeycomb substrate. The particulate support used in the exhaust gas purifying catalyst of the present invention can be produced by the present process for producing metal oxide particle comprising a core part and surface layer differing in composition. In the process for producing a particulate support used in the exhaust gas purifying catalyst of the present invention, it is preferable to use CeO_2 and ZrO_2 sols comprising CeO_2 and ZrO_2 colloid particles having a particle diameter as small as possible such that the particle diameter of the resulting particulate support is small. The particulate support having a smaller particle diameter has a larger specific surface area. Also, the particle diameter of the noble metal to be supported on the particulate support is preferably controlled to be small, thereby increasing the specific surface area thereof." [0098] "The optimum contents of CeO_2 and ZrO_2 are examined in the following example. Both CeO_2 and ZrO_2 colloid particles in CeO_2 and ZrO_2 sols used have a particle diameter of 100 nm or less. The particle diameter of the particulate support produced is 6 μm or less. The Pt particle diameter supported was 3 nm or less." To the extent that this limitation is not disclosed, either explicitly or inherently, by *Miura*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.

EXHIBIT I

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2007/0197379 ("*Miura*")

'864 patent	**U.S. Patent Application No. 2007/0197379 ("*Miura*")**
1[b] silica, alumina, or mixtures thereof; and	*Miura* discloses a combustion promoter comprising silica, alumina, or mixtures thereof. *See, e.g.*, [0002]-[0003] "The exhaust gas from internal combustion engines, such as automobile engines, contains nitrogen oxide (NO_x), carbon monoxide (CO), hydrocarbon (HC) and the like. These substances can be purified by an exhaust gas purifying catalyst capable of oxidizing CO and HC and, at the same time, reducing NO_x. As for representative exhaust gas purifying catalysts, three-way catalysts where a noble metal such as platinum (Pt), rhodium (Rh) and palladium (Pd) is supported on a porous metal oxide support such as γ-alumina are known. The metal oxide support may be formed of various materials but, in order to obtain a large surface area, alumina (Al_2O_3) has been heretofore generally used. However, in recent years, for accelerating purification of the exhaust gas by using chemical properties of the support, it has been proposed to use various other materials such as ceria (CeO_2), zirconia (ZrO_2) and titanium (TiO_2) in combination with, or not in combination with, alumina." [0014]-[0015] "According to the process of the present invention, a metal oxide particle comprising a core part relatively rich in a component originated in the population of first colloid particles and a surface layer relatively rich in a component originated in the population of second colloid particles can be obtained. Furthermore, according to the process of the present invention, a metal oxide particle having any particle diameter and having a structure comprising a core part and a surface layer can be obtained. For example, when the population of colloid particles used as a raw material in practice has an average particle diameter of about 5 nm, the average particle diameter of the metal oxide particle produced by the

EXHIBIT I

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2007/0197379 ("*Miura*")

'864 patent	U.S. Patent Application No. 2007/0197379 ("*Miura*")
	process of the present invention can be made to be 50 nm or less. Accordingly, this metal oxide particle can have an average particle diameter of, for example, less than 10 μm, less than 5 μm, less than 1 μm, less than 500 nm, less than 200 nm, less than 100 nm or less than 50 nm." [0023]-[0027] "In another embodiment of the process of the present invention, the population of first colloid particles and a population of second colloid particles each is independently selected from the group consisting of alumina, ceria, zirconia and titania colloid particles. In another embodiment of the process of the present invention, the population of first colloid particles is zirconia, alumina or titania, particularly zirconia, and the population of second colloid particles is ceria. According to this embodiment, a metal oxide particle comprising a core part relatively rich in zirconia, alumina or titania and a surface layer relatively rich in ceria can be obtained. When platinum is supported on such a metal oxide particle, good heat resistance attributable to zirconia, alumina or titania, particularly zirconia, can be realized, and an effect of preventing sintering of platinum can be achieved by virtue of ceria. In this case, the total molar fraction of cerium and zirconium, aluminum or titanium may be at least 85 mol %, particularly at least 90 mol %, more particularly at least 95 mol %, based on the total molar number of metals in the metal oxide particle." [0046]-[0047] "As for the metal oxides constituting the metal oxide particle produced by the process of the present invention, any metal oxide can be selected and a metal oxide

EXHIBIT I

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2007/0197379 ("*Miura*")

'864 patent	U.S. Patent Application No. 2007/0197379 ("*Miura*")
	which is preferably held in the core part of the metal oxide particle may be selected as a first metal oxide, while selecting, as a second metal oxide, a metal oxide which is preferably exposed to the surface layer of the metal oxide particle. For example, the first metal oxide is preferably zirconia and the second metal oxide is preferably ceria. The zirconia has high heat resistance and the ceria can prevent sintering of platinum when platinum is supported. When the surface layer or core part of the metal oxide particle of the present invention contains zirconia or ceria, the core or surface layer may contain a metal other than cerium (Ce) and zirconium (Zr), for example, a metal selected from the group consisting of alkaline earth metals and rare earth elements, particularly yttrium (Y). These alkaline earth metals and rare earth elements, particularly yttrium, tend to provide excellent heat resistance to zirconia and ceria." [0080-0081] "In this Example, a metal oxide particle comprising a core part relatively rich in alumina and a surface layer relatively rich in ceria is obtained from an alkali-stabilized alumina aqueous sol and an acid-stabilized ceria aqueous sol. An alkali-stabilized alumina aqueous sol (isoelectric point: pH 4.8) and an acid-stabilized ceria aqueous sol (isoelectric point: pH 8.5) were mixed to give a molar ratio of 1:2 between alumina (Al_2O_3) and ceria (CeO_2). To this sol mixture, an aqueous nitric acid solution was added dropwise with stirring to adjust the pH to 3.0, thereby aggregating alumina. Thereafter, an aqueous ammonia solution was added dropwise to this mixed sol with stirring to adjust the pH to 10, thereby aggregating ceria. Subsequently, drying and firing were performed in the same manner as in Example 1 to obtain a metal oxide particle."

EXHIBIT I

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2007/0197379 ("*Miura*")

'864 patent	**U.S. Patent Application No. 2007/0197379 ("*Miura*")**
	[0086]-[0087] "In this Example, a metal oxide particle comprising a core part relatively rich in alumina and a surface layer relatively rich in ceria is obtained from an acid-stabilized alumina aqueous sol and an alkali-stabilized ceria aqueous sol. An acid-stabilized alumina aqueous sol (isoelectric point: pH 7.6) and an alkali-stabilized ceria aqueous sol (isoelectric point: pH 4.0) were mixed to give a molar ratio of 1:2 between alumina (Al_2O_3) and ceria (CeO_2). To this mixed sol, an aqueous ammonia solution was added dropwise with stirring to adjust the pH to 10, thereby aggregating alumina. Thereafter, an aqueous nitric acid solution was added dropwise to this mixed sol with stirring to adjust the pH to 3.0, thereby aggregating ceria. Subsequently, drying and firing were performed in the same manner as in Example 1 to obtain a metal oxide particle." To the extent that this limitation is not disclosed, either explicitly or inherently, by *Miura*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
1[c] one or more Group VIII noble metals distributed in the particle as an eggshell such that a higher concentration of the one or more Group VIII noble metals is present in the outer region of the microsphere sized porous particle as compared to the concentration of the one or more Group VIII	*Miura* discloses a combustion promoter comprising one or more Group VIII noble metals, the metals distributed in the particle as an eggshell such that a higher concentration of the one or more Group VIII noble metals is present in the outer region of the microsphere sized porous particle as compared to the concentration of the one or more Group VIII noble metals in the centre of the microsphere sized porous particle. *See, e.g.*,

EXHIBIT I

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2007/0197379 ("*Miura*")

'864 patent	**U.S. Patent Application No. 2007/0197379 ("*Miura*")**
noble metals in the centre of the microsphere sized porous particle.	[Abstract] "The present invention provides an exhaust gas purifying catalyst and a process for producing a metal oxide particle comprising multiple species of metal oxides and capable of satisfactorily exerting the properties of respective metal oxides. A process of the present invention comprises providing a sol containing at least a population of first colloid particles and a population of second colloid particles differing in the isoelectric point with each other; adjusting the pH of the sol to be closer to the isoelectric point of the population of first colloid particles than to the isoelectric point of the population of second colloid particles, thereby aggregating the population of first colloid particles; adjusting the pH of the sol, thereby aggregating the population of second colloid particles onto the population of first colloid particles aggregated; and drying and firing the obtained aggregate." [0001] "The present invention relates to a process for producing a metal oxide particle and, more specifically, the present invention relates to a process for producing a metal oxide particle preferably used as an exhaust gas purifying catalyst by loading a noble metal thereon. Further, present invention relates to an exhaust gas purifying catalyst for an internal combustion engine." [0002] "The exhaust gas from internal combustion engines, such as automobile engines, contains nitrogen oxide (NO_x), carbon monoxide (CO), hydrocarbon (HC) and the like. These substances can be purified by an exhaust gas purifying catalyst capable of oxidizing CO and HC and, at the same time, reducing NO_x. As for representative exhaust gas purifying catalysts, three-way catalysts where a noble metal such as platinum (Pt), rhodium (Rh) and palladium (Pd) is supported on a porous metal oxide support such as γ-alumina are known."

EXHIBIT I

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2007/0197379 ("*Miura*")

'864 patent	U.S. Patent Application No. 2007/0197379 ("*Miura*")
	[0005] "In order to allow for efficient progress of oxidation of CO and HC and reduction of NO_x by the activity of the three-way catalyst, the air-fuel ratio in the internal combustion engine must be a theoretical air-fuel ratio (stoichiometric air-fuel ratio). Therefore, the fluctuation of oxygen concentration in the exhaust gas is preferably alleviated by a material having OSC to maintain the oxygen concentration in the vicinity of the theoretical air-fuel ratio, so that the three-way catalyst can exert its exhaust gas purifying ability. Furthermore, according to recent studies, it has been found that ceria not only has OSC but also, by virtue of its strong affinity for a noble metal, particularly platinum, can prevent particle growth (sintering) of the noble metal supported thereon." [0009]-[0012] "Also, a substantially uniform metal oxide particle may be obtained from a sol in which multiple different species of colloid particles are mixed, but a uniform mixture does not always yield the best result. For example, a composite metal oxide obtained by uniformly mixing ceria and zirconia is known to have good OSC and heat resistance, but sometimes does not allow ceria to satisfactorily bring out its property of preventing sintering of noble metal such as platinum. That is, ceria and zirconia both are present on the surface of this composite metal oxide and, therefore, a part of the noble metal is supported on the zirconia portion rather than on the ceria portion and cannot be prevented from sintering, in some cases. Accordingly, the present invention provides a process for producing a metal oxide particle comprising multiple species of metal oxides and capable of satisfactorily exerting the properties of respective metal oxides.

EXHIBIT I

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2007/0197379 ("*Miura*")

'864 patent	U.S. Patent Application No. 2007/0197379 ("*Miura*")
	Further, the present invention provides an exhaust gas purifying catalyst, which can exert a heat resistance improving effect due to ZrO_2, and an effect of preventing sintering of a noble metal supported on CeO_2 at the same time, by supporting the noble metal on a particulate support comprising a core relatively rich in ZrO_2 and a surface layer relatively rich in CeO_2." [0013]-[0016] "The present process for producing a metal oxide particle comprising a core part and a surface layer differing in the composition comprises providing a sol containing at least a population of first colloid particles and a population of second colloid particles differing in the isoelectric point with each other; adjusting the pH of the sol to be closer to the isoelectric point of the population of first colloid particles than to the isoelectric point of the population of second colloid particles, particularly closer to the range of ±1.0, more particularly ±0.5, of the isoelectric point of the population of first colloid particles, thereby aggregating the population of first colloid particles; adjusting the pH of the sol to be closer to the isoelectric point of the population of second colloid particles than to the isoelectric point of the population of first colloid particles, particularly be into the range of ±1.0, more particularly ±0.5, of the isoelectric point of the population of second colloid particles, thereby aggregating the population of second colloid particles onto the population of first colloid particles aggregated; and drying and firing the obtained aggregate. According to the process of the present invention, a metal oxide particle comprising a core part relatively rich in a component originated in the population of first colloid particles and a surface layer relatively rich in a component originated in the population of second colloid particles can be obtained. Furthermore, according to the process of the present invention, a metal oxide particle having any particle diameter and having a structure comprising a core part and a surface layer can be obtained. For example, when the population of colloid

EXHIBIT I

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2007/0197379 ("*Miura*")

'864 patent	U.S. Patent Application No. 2007/0197379 ("*Miura*")
	particles used as a raw material in practice has an average particle diameter of about 5 nm, the average particle diameter of the metal oxide particle produced by the process of the present invention can be made to be 50 nm or less. Accordingly, this metal oxide particle can have an average particle diameter of, for example, less than 10 μm, less than 5 μm, less than 1 μm, less than 500 nm, less than 200 nm, less than 100 nm or less than 50 nm The term "relatively rich in" as used herein for the metal oxide comprising a core part and a surface layer is used with respect to the molar fraction based on the total molar number of metals in each of the core part and the surface layer. Accordingly, for example, the "core part relatively rich in a component originated in the population of first colloid particles" means that the molar fraction of a metal constituting this component in the core part is higher than the molar fraction of the same metal in the surface layer." [0023]-[0030] "In another embodiment of the process of the present invention, the population of first colloid particles and a population of second colloid particles each is independently selected from the group consisting of alumina, ceria, zirconia and titania colloid particles. In another embodiment of the process of the present invention, the population of first colloid particles is zirconia, alumina or titania, particularly zirconia, and the population of second colloid particles is ceria. According to this embodiment, a metal oxide particle comprising a core part relatively rich in zirconia, alumina or titania and a surface layer relatively rich in ceria can be obtained.

EXHIBIT I

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2007/0197379 ("*Miura*")

'864 patent	U.S. Patent Application No. 2007/0197379 ("*Miura*")
	When platinum is supported on such a metal oxide particle, good heat resistance attributable to zirconia, alumina or titania, particularly zirconia, can be realized, and an effect of preventing sintering of platinum can be achieved by virtue of ceria. In this case, the total molar fraction of cerium and zirconium, aluminum or titanium may be at least 85 mol %, particularly at least 90 mol %, more particularly at least 95 mol %, based on the total molar number of metals in the metal oxide particle. The present exhaust gas purifying catalyst for internal combustion engine comprises a particulate support and a noble metal supported thereon, the particulate support comprising a core part relatively rich in zirconia (ZrO_2) and a surface layer relatively rich in ceria (CeO_2), the content of CeO_2 in the particulate support being 40 to 65 mol %, particularly 45 to 55 mol %. According to the exhaust gas purifying catalyst of the present invention, in the particulate support, the core part relatively rich in ZrO_2 has a suitable volume, and the surface layer relatively rich in CeO_2 covers the entire core part to form a layer having a suitable thickness. As a result, the effect of improving heat resistance due to ZrO_2 and the effect of preventing sintering of the noble metal due to CeO_2 both are preferably exerted at the same time. In one embodiment of the exhaust gas purifying catalyst of the present invention, the surface layer comprises at least one element selected from the group consisting of alkaline earth metals and rare earths, particularly at least one of Y and Nd."

EXHIBIT I

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2007/0197379 ("*Miura*")

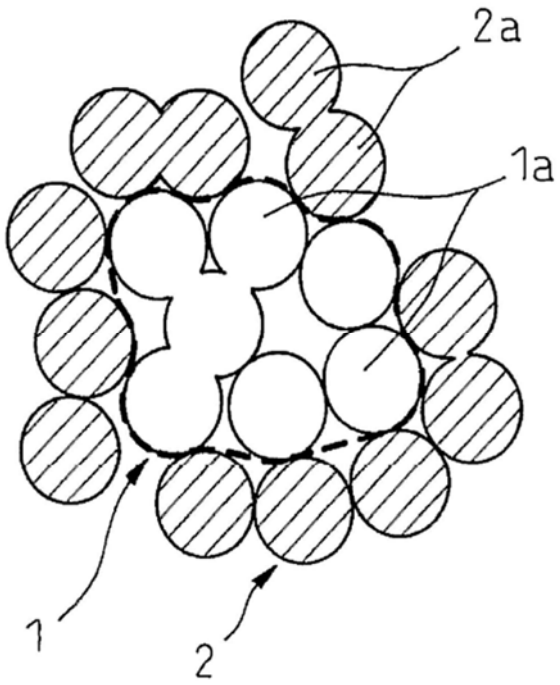
'864 patent	**U.S. Patent Application No. 2007/0197379 ("*Miura*")**
	[Fig. 1] 

EXHIBIT I

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2007/0197379 ("*Miura*")

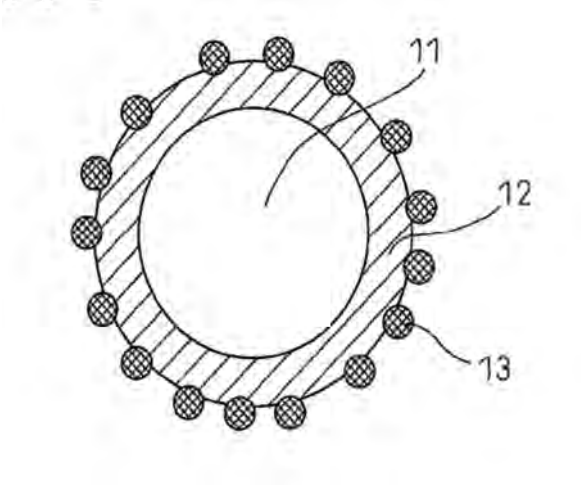
'864 patent	U.S. Patent Application No. 2007/0197379 ("*Miura*")
	[Fig. 2]  [0034]-[0035] "FIG. 1 is a cross-sectional view showing the metal oxide particle produced by the method of the present invention. FIG. 2 is a cross-sectional view showing a support of the present invention and a noble metal supported thereon." [0044]-[0049] "As shown in FIG. 1, according to the process of the present invention, a metal oxide particle comprising a core part 1 and a surface layer 2 differing in the composition can be produced. More specifically, according to the process of the present invention, in a sol containing at least two populations of colloid particles differing in the isoelectric point from each other, the population of first colloid particles is aggregated and then the population of second colloid particles is aggregated onto the periphery of the population of first aggregated colloid particles, whereby a metal oxide particle comprising a core part 1 mainly composed of a

EXHIBIT I

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2007/0197379 ("*Miura*")

'864 patent	U.S. Patent Application No. 2007/0197379 ("*Miura*")
	component originated in the population of first colloid particles and a surface layer 2 mainly composed of a component originated in the population of second colloid particles is produced. In the metal oxide particle shown, the core part 1 and the surface layer 2 each comprises a plurality of primary particles (1 *a*, 2 *a*) originated in the population of first colloid particles and a population of second colloid particles. However, a distinct boundary may or may not be present between respective primary particles. Also, the boundary between the core part 1 and the surface layer 2 may not be necessarily distinct and may appear as a portion where the composition is gradually changing. Furthermore, the boundary part between the core part 1 and the surface layer 2 may be a mixture, particularly a solid solution, of a component originated in the population of first colloid particles and a component originated in the population of second colloid particles. In FIG. 1, the surface layer 2 is shown as if it is discontinuous, but the surface layer may be continuous. As for the metal oxides constituting the metal oxide particle produced by the process of the present invention, any metal oxide can be selected and a metal oxide which is preferably held in the core part of the metal oxide particle may be selected as a first metal oxide, while selecting, as a second metal oxide, a metal oxide which is preferably exposed to the surface layer of the metal oxide particle. For example, the first metal oxide is preferably zirconia and the second metal oxide is preferably ceria. The zirconia has high heat resistance and the ceria can prevent sintering of platinum when platinum is supported. When the surface layer or core part of the metal oxide particle of the present invention contains zirconia or ceria, the core or surface layer may contain a metal other than cerium (Ce) and zirconium (Zr), for example, a metal selected from the group consisting of alkaline earth metals and rare earth elements, particularly

EXHIBIT I

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2007/0197379 ("*Miura*")

'864 patent	U.S. Patent Application No. 2007/0197379 ("*Miura*")
	yttrium (Y). These alkaline earth metals and rare earth elements, particularly yttrium, tend to provide excellent heat resistance to zirconia and ceria. A noble metal such as platinum, rhodium and palladium is supported on the metal oxide particle obtained by the process of the present invention, and thereby an exhaust gas purifying catalyst can be produced. In the exhaust gas purifying catalyst produced, the noble metal can be supported mainly on the surface layer, because the metal oxide particle obtained by the process of the present invention can have a core part and a surface layer. The noble metal may be loaded on the metal oxide particle by any known method, for example, by a method of impregnating metal oxide particles with a solution containing a salt and/or a complex salt of noble metal, and drying and then firing it. The amount of the noble metal supported on the metal oxide particle may be from 0.01 to 5 mass %, particularly from 0.1 to 2 mass %, based on the metal oxide particle." [0066] "The thus-obtained aggregate is dried and fired, whereby a metal oxide particle comprising a core part mainly composed of a component originated in the population of first colloid particles and a surface layer mainly composed of a component originated in the population of second colloid particles can be produced." [0068]-[0072] "As shown in FIG. 2, in an optimum aspect of the exhaust gas purifying catalyst of the present invention, CeO_2 constituting a surface layer 12 of particulate support covers an entire core part 22 formed of ZrO_2 in a proper thickness, and a noble metal 13 (Pt) is supported on the particulate support. Furthermore, the particulate support preferably has a small particle diameter. It was confirmed that the exhaust

EXHIBIT I

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2007/0197379 ("*Miura*")

'864 patent	U.S. Patent Application No. 2007/0197379 ("*Miura*")
	gas purifying catalyst comprising the particulate support and the noble metal 3 supported thereon had an excellent exhaust gas purifying performance. The same effect is exerted in a monolith catalyst obtained by coating the particulate support on a honeycomb substrate. The particulate support used in the exhaust gas purifying catalyst of the present invention can be produced by the present process for producing metal oxide particle comprising a core part and surface layer differing in composition. In the process for producing a particulate support used in the exhaust gas purifying catalyst of the present invention, it is preferable to use CeO_2 and ZrO_2 sols comprising CeO_2 and ZrO_2 colloid particles having a particle diameter as small as possible such that the particle diameter of the resulting particulate support is small. The particulate support having a smaller particle diameter has a larger specific surface area. Also, the particle diameter of the noble metal to be supported on the particulate support is preferably controlled to be small, thereby increasing the specific surface area thereof. An increase in the specific surface area of the particulate support makes it possible to increase the amount of the noble metal which can be supported, even if the same weight of the particulate support is used. Increase of the specific surface area of the noble metal makes it possible to increase the area of the noble metal which is contacted with the exhaust gas, even if the same weight of the noble metal is used. These increases of the surface area can improve exhaust gas purifying performance, even if a smaller amount of the particulate support, in combination with the noble metal, is used. The noble metal can be supported by dispersing the particulate support in distilled water and adding a noble metal solution, followed by stirring, drying and further firing. The noble metal to be supported is preferably at least one selected from the

EXHIBIT I

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2007/0197379 ("*Miura*")

'864 patent	U.S. Patent Application No. 2007/0197379 ("*Miura*")
	group consisting of Pt, Pd, Rh, Ir and Au, more preferably at least one selected from the group consisting of Pt, Pd and Rh, and Pt is most preferable." [0101] "The thus-obtained particulate supports were dispersed in distilled water which was in an amount of 6-fold weight, an aqueous dinitrodiamine platinum solution (Pt content: 4.4% by weight) was added thereto so that platinum is in an amount of 1% by weight based on the particulate support, and the resulting dispersion was stirred for 1 hour. Thereafter, the water content was removed by drying at 120° C. for 24 hours and the residue was fired at 500° C. for 2 hours. The obtained catalyst was shaped into a 1 mm-square pellet and used for the evaluation of performance." [0104] "The thus-obtained particulate supports were dispersed in distilled water which was in an amount of 6 times by weight, an aqueous dinitrodiamine platinum solution (Pt content: 4.4% by weight) was added thereto so that platinum is in an amount of 1% by weight based on the particulate support, and the resulting dispersion was stirred for 1 hour. Thereafter, the water content was removed by drying at 120° C. for 24 hours and the residue was fired at 500° C. for 2 hours. The obtained catalyst was shaped into a 1 mm-square pellet and used for the evaluation of performance." To the extent that this limitation is not disclosed, either explicitly or inherently, by *Miura*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.

EXHIBIT I

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2007/0197379 ("*Miura*")

'864 patent	**U.S. Patent Application No. 2007/0197379 ("*Miura*")**
[3] The combustion promoter according to claim 1, comprising a plurality of microsphere sized porous particles comprising alumina.	*Miura* discloses a combustion promoter comprising a plurality of microsphere sized porous particles comprising alumina. *See, e.g.*, The support provided above for Claim 1[b]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Miura*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[6] The combustion promoter according to claim 1, wherein the microsphere sized porous particles collectively have an average (D50) size of between 60 and 90 microns as measured by laser diffraction.	*Miura* discloses a combustion promoter comprising microsphere sized porous particles, wherein the microsphere sized porous particles collectively have an average (D50) size of between 60 and 90 microns as measured by laser diffraction. *See, e.g.*, The support provided above for Claim 1[a]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Miura*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.

EXHIBIT I

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2007/0197379 ("*Miura*")

'864 patent	**U.S. Patent Application No. 2007/0197379 ("*Miura*")**
[12] The combustion promoter according to claim 1, wherein the one or more Group VIII noble metals comprises one or more of platinum, palladium, iridium, ruthenium, and rhodium.	*Miura* discloses a combustion promoter comprising one or more Group VIII noble metals, wherein the one or more Group VIII noble metals comprises one or more of platinum, palladium, iridium, ruthenium, and rhodium. *See, e.g.*, The support provided above for Claim 1[c]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Miura*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[15] The combustion promoter according to claim 12, wherein the one or more Group VIII noble metals comprises palladium.	*Miura* discloses a combustion promoter comprising one or more Group VIII noble metals, wherein the one or more Group VIII noble metals comprises palladium. *See, e.g.*, The support provided above for Claim 1[c]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Miura*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.

EXHIBIT J

EXHIBIT J

Invalidity of U.S. Patent No. 11,224,864 in View of *Munnik Article*

"*Recent Developments in the Synthesis of Supported Catalysts*" ("*Munnik Artic*le"), authored by Peter Munnik et al. and published in 2015, qualifies as prior art of U.S. Patent No. 11,224,864 ("the '864 patent") under at least 35 U.S.C. §§ 102 and/or 103 for the asserted claims of the '864 patent, as discussed in detail in the claim chart below.

The Court has not yet construed the claims and therefore the meaning of the terms in the claims has yet to be resolved by the Court. The support identified here for these limitations is responsive to Plaintiffs' infringement contentions, which Defendant disagrees with. As such, nothing in Defendant's claim charts should be construed as an admission regarding infringement, either literally or under the doctrine of equivalents, or as an admission regarding Defendant's understanding of the proper scope of the asserted claims. Defendant reserves the right to rely on additional citations or sources of evidence that also may be applicable, or that may become applicable in light of claim construction, changes in Plaintiffs' infringement contentions, and/or information obtained during discovery as the case progresses.

To the extent Plaintiffs allege that *Munnik Article* does not disclose any particular limitation of the asserted claims of the '864 patent, either expressly or inherently, it would have been obvious to a person of ordinary skill in the art as of the priority date of the '864 patent to modify *Munnik Article* and/or to combine the teachings of *Munnik Article* with other prior art references, including but not limited to the present prior art references cited in the Cover Pleading and the relevant section(s) of claim charts for other prior art references for the '864 patent in a manner that would have rendered the asserted claims invalid as obvious.

Defendant reserves the right to amend or supplement this claim chart at a later date as more fully set forth in the Cover Pleading.

EXHIBIT J

Invalidity of U.S. Patent No. 11,224,864 in View of *Munnik Article*

'864 patent	**"*Recent Developments in the Synthesis of Supported Catalysts*" ("*Munnik Article*")**
1[a] A CO to CO_2 combustion promoter comprising microsphere sized porous particles, each microsphere sized porous particle having a diameter of less than 1mm, and independently comprising	*Munnik Article* discloses a CO to CO_2 combustion promoter comprising microsphere sized porous particles, each microsphere sized porous particle having a diameter of less than 1mm, and independently comprising the limitations discussed in the following sections. *See, e.g.*, [Page 6687] "Heterogeneous catalysis, whereby a gas- or liquid-phase reaction is performed over a solid catalyst, is at the heart of the modern energy and chemical industries. Most chemical processes, both established and emerging, are performed using functional nanomaterials as catalysts. Moreover, upcoming fields such as nanoelectronics and energy conversion and storage are based upon the application of similar nanomaterials. The catalytic activity for these materials often resides with metal (oxide, sulfide) surface sites, and efficient use of the metals and space available requires small particles, located on a mostly inert support to enhance the thermal stability of the catalyst. The field of catalyst synthesis, also known as catalyst preparation or catalyst manufacturing, aims at establishing the desired composition and structure of these materials." [Page 6688] "Catalysts come in a wide variety to accommodate the large differences in operating conditions, feedstocks, and products, and the catalytically active metals can be deposited on a range of supports, which are generally highly porous and can vary in size from small powders to centimeter-sized pellets. In each case, high activities and selectivities toward the desired products are required. Moreover, the stability of the catalyst is highly important, and processes such as the irreversible growth of particles or poisoning of the active sites have to be prevented. The performance and stability of the catalyst are determined by what we refer to as the 'DNA' of the catalyst, that is, the density, nature, and accessibility of the active sites, with the accessibility largely determined by the support pore architecture."

EXHIBIT J

Invalidity of U.S. Patent No. 11,224,864 in View of *Munnik Article*

'864 patent	***"Recent Developments in the Synthesis of Supported Catalysts"* ("*Munnik Article*")**
	[Page 6689] "Common support materials consist of refractory oxides such as SiO_2, Al_2O_3, or TiO_2. These materials exhibit high specific surface areas, high porosities, and high thermal and mechanical stability and come in a variety of pore sizes, while they are mostly chemically inert. Carbon is also widely used, while zeolites are often applied in many oil-refining and petrochemical applications. Recently, an increasing variety of ordered mesoporous materials have been used as model supports. Finally, metal organic frameworks are showing an increased use as catalysts or as support for metal (oxide) nanoparticles. To maintain activity as well as for practical reasons, the support requires a high mechanical strength and the macroscopic bodies should be resistant to attrition if the catalyst is vigorously agitated such as in fluid bed or slurry bed reactors." [Page 6693] "Impregnation and drying is a frequently used preparation method among others due to its simple execution and low waste streams. The first step is contacting a metal precursor solution with a porous support. Common precursors include inorganic metal salts, such as metal sulfates, carbonates, chlorides, nitrates, or acetates, and organic metal complexes, such as metal acetylacetonates. The most commonly used solvent for inorganic salts is water because of the high solubility of many precursors, whereas organic solvents are mainly used for organometallic precursors. To prevent premature deposition of the metal precursor in bulk solution, concentrations below (super)saturation are required." [Page 6694] "However, it is also possible N_2 physisorption underestimated the total pore volume of the SBA-15, so that there was not enough liquid to fill all the pores. A more detailed cryo-TEM tomography study was performed by Eggenhuisen et al. on

EXHIBIT J

Invalidity of U.S. Patent No. 11,224,864 in View of *Munnik Article*

'864 patent	**"*Recent Developments in the Synthesis of Supported Catalysts*" ("*Munnik Article*")**
	SBA-15 impregnated with cobalt nitrate or nickel nitrate solutions. Although a few empty pores were found mainly on the external edge of the support particles, the majority were filled, and it was concluded that impregnation does lead to an almost homogeneous and complete pore filling. More quantitative measurements on larger samples by differential scanning calorimetry (DSC) have also provided evidence on the complete filling of porous supports (Figure 8). Due to the downward shift in the melting point of liquid confined in nanopores, a distinction was possible between extraporous and intraporous liquid in the material. Incipient wetness impregnation of several ordered mesoporous silicas and silica gels with several solutions, including a eutectic nickel nitrate solution, resulted in fillings between 90% and 100%." [Page 6695] "Although short contact times (1h) resulted in outer sphere complex formation, a longer contact time between platinum tetrammine and oxidized carbon at low pH gave rise to reduction of Pt concurrent with oxidation of the carbon surface. Similarly, when trying to deposit cobalt on a carbon support at high pH, it was found that the cobalt ammine complex was completely converted to Co_3O_4 after 1h, which did not occur on silica. Reductive ammination of the precursor took place, forming different precipitates depending on the pH (CoO at pH 9.2, Co_3O_4 at pH 9.8, and $Co(OH)_4^{2-}$ at pH > 11 when NH_4OH was used as the base), so that the process is better described as a deposition precipitation. Moreover, at higher pH the cobalt ammine reacted to $[Co(NH_3)_5Cl]Cl_2$, which did not adsorb at all. Nonetheless, particles of 5 nm size were obtained at a Co loading of 6.7 wt % at a pH of 10." [Page 6699] "For weakly adsorbing species and high concentrations, convection and back-diffusion during the drying step become very important. In the model of Liu et al., when the adsorption strength of nickel nitrate was reduced to a moderate value and

EXHIBIT J

Invalidity of U.S. Patent No. 11,224,864 in View of *Munnik Article*

'864 patent	***"Recent Developments in the Synthesis of Supported Catalysts"* ("*Munnik Article*")**
	assuming a homogeneous distribution after impregnation, drying could result in severe redistribution of the 0.25 M nickel nitrate solution on y-alumina depending on the conditions. Drying at 70 °C resulted in an almost homogeneous distribution. However, increasing the drying temperature to 300 °C resulted in eggshell distributions, because back-diffusion of nickel nitrate was not fast enough to compensate for the high drying rate. Similar results were obtained upon alteration of the humidity of the drying air; at 0.5% humidity, drying was relatively fast, resulting in an eggshell distribution, while drying in 10% humidity resulted in an almost homogeneous distribution. Finally, the size of the support pellet had a great influence on the distribution. A pellet diameter of 1 mm resulted in a moderate eggshell distribution whereby the concentration of nickel nitrate in the shell was 1.5 times higher than in the center of the pellet. For a 10 mm pellet, the concentration in the shell was almost 3 times higher compared to that in the center. The model was recently extended to a 3 mm γ-alumina pellet impregnated with nickel nitrate concentrations ranging from 0.005 to 4.0 M (Figure 13), and many of the results were compared to experimental data." [Page 6708] "Wet impregnation is commonly applied to deposit the colloids onto a support due to the relatively low nanoparticle concentrations in colloidal solutions necessary to prevent aggregation and precipitation. It can take a long time for all particles to migrate into the pores of the support because diffusivity of colloidal nanoparticles is relatively slow, which can be stimulated through use of sonication. The drying step can have a large influence on the distribution of the nanoparticles when particles are not strongly bound to the support, as has been demonstrated for the drying of colloidal wet films. Significant redistribution toward the external surface of the grains is possible since evaporation of the solvent in porous bodies usually occurs on the external surface (see section 4.3)."

EXHIBIT J

Invalidity of U.S. Patent No. 11,224,864 in View of *Munnik Article*

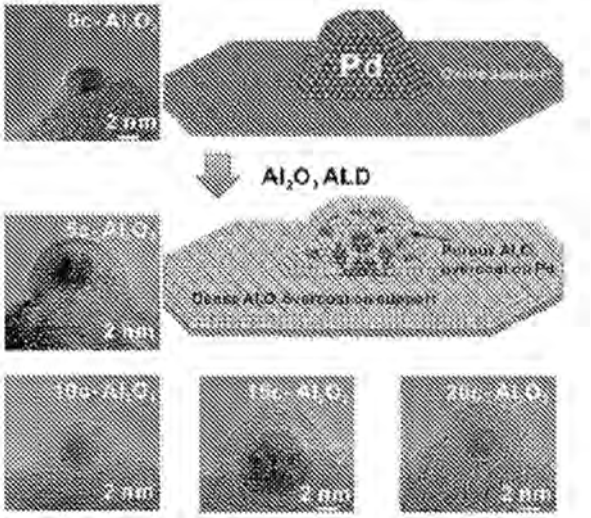
'864 patent	***"Recent Developments in the Synthesis of Supported Catalysts" ("Munnik Article")***
	[Page 6710] "Finally, it is noted that in recent years colloidal techniques have been used for synthesizing a porous support shell around preformed nanoparticles. While these are relevant for catalysis and catalyst preparation, their discussion is beyond the scope of the present review, and they have recently been reviewed elsewhere." [Page 6712]  "Figure 23. Schematic representation of the deposition of a porous alumina layer on Pd particles supported on an oxidic support, and TEM images of spherical alumina-supported Pd particles with different numbers of $A1_2O_3$ layers deposited on top, represented by the number of cycles (xc). An increasingly thick layer was observed with increasing cycles; however, the particles remained accessible for catalysis. Adapted from ref 252. Copyright 2012 American Chemical Society."

EXHIBIT J

Invalidity of U.S. Patent No. 11,224,864 in View of *Munnik Article*

'864 patent	***"Recent Developments in the Synthesis of Supported Catalysts"* ("*Munnik Article*")**
	To the extent that this limitation is not disclosed, either explicitly or inherently, by *Munnik Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
1[b] silica, alumina, or mixtures thereof; and	*Munnik Article* discloses a combustion promoter comprising silica, alumina, or mixtures thereof. *See, e.g.*, [Page 6689] "Common support materials consist of refractory oxides such as SiO_2, Al_2O_3, or TiO_2. These materials exhibit high specific surface areas, high porosities, and high thermal and mechanical stability and come in a variety of pore sizes, while they are mostly chemically inert. Carbon is also widely used, while zeolites are often applied in many oil-refining and petrochemical applications. Recently, an increasing variety of ordered mesoporous materials have been used as model supports. Finally, metal organic frameworks are showing an increased use as catalysts or as support for metal (oxide) nanoparticles. To maintain activity as well as for practical reasons, the support requires a high mechanical strength and the macroscopic bodies should be resistant to attrition if the catalyst is vigorously agitated such as in fluid bed or slurry bed reactors."

EXHIBIT J

Invalidity of U.S. Patent No. 11,224,864 in View of *Munnik Article*

'864 patent	***"Recent Developments in the Synthesis of Supported Catalysts"* ("*Munnik Article*")**
	[Page 6689] **Table 1. Point of Zero Charge for Several Important Supports in Aqueous Solution[29]** support / PZC range / support / PZC range SiO_2 / 2–4 / γ-Al_2O_3 / 8–9 oxidized activated carbon / 2–4 / activated carbon / 8–10 TiO_2 / 5–7 [Page 6695] "Recently, Regalbuto et al. used palladium, copper, cobalt, ruthenium, and nickel ammine complexes to obtain silica- supported catalysts at loadings between 1 and 3 wt %. After deposition at a pH of 12, the catalysts were dried and directly reduced. Small particles between 1 and 3 nm in size were found, and calculations showed that these metal loadings were close to the maximum obtainable by electrostatic adsorption for the used silica with a surface area of 180 m^2/ g and a PZC of 4.0. It was postulated that the maximum loading mainly depends on the surface area of the support. A follow-up study focused on SBA-15 supports with different surface areas.[91] Ammine complexes were used to deposit Pd nanoparticles of 1.3 nm size at a relatively high loading of 8.7 wt % on 1000 m^2/ g SBA- 15, and 1.9 nm Pt particles were synthesized on SBA-15 with a 600 m^2/ g surface area with a loading of 9.2 wt %, showing high surface areas were indeed beneficial for adsorption. Co particles were also synthesized using this method, but larger particles of 7 nm size at a loading of 5.1 wt % resulted. This was attributed to particle growth that occurred during the high reduction temperature of 800 °C needed to reduce the strongly adsorbing cobalt complexes, whereas the Pt and Pd catalysts were reduced at 350 °C or lower. Moreover, dissolution of the silica support was already observed at pH 9,

EXHIBIT J

Invalidity of U.S. Patent No. 11,224,864 in View of *Munnik Article*

'864 patent	***"Recent Developments in the Synthesis of Supported Catalysts"* ("*Munnik Article*")**
	and 25% of the SBA-15 silica was dissolved after 1h at pH 10, the optimum pH for the uptake of metal cations. This led to a large increase in pore diameter and pore volume; however, this was not considered a problem due to the relatively thick walls of SBA-15. It was suggested to minimize the contact time to reduce this effect." [Page 6695] "Although short contact times (1h) resulted in outer sphere complex formation, a longer contact time between platinum tetrammine and oxidized carbon at low pH gave rise to reduction of Pt concurrent with oxidation of the carbon surface. Similarly, when trying to deposit cobalt on a carbon support at high pH, it was found that the cobalt ammine complex was completely converted to Co_3O_4 after 1h, which did not occur on silica. Reductive ammination of the precursor took place, forming different precipitates depending on the pH (CoO at pH 9.2, Co_3O_4 at pH 9.8, and $Co(OH)_4^{2-}$ at pH > 11 when NH_4OH was used as the base), so that the process is better described as a deposition precipitation. Moreover, at higher pH the cobalt ammine reacted to $[Co(NH_3)_5Cl]Cl_2$, which did not adsorb at all. Nonetheless, particles of 5 nm size were obtained at a Co loading of 6.7 wt % at a pH of 10." [Page 6695-6696] "While in each of the previously discussed studies diluted slurries of a powdered support in a large excess of solvent were used, it was recently shown that significant adsorption is also possible during incipient wetness impregnation, in a modified method called charge-enhanced dry impregnation (CEDI). To overcome the buffering effect and charge of the support, very high or low pH values were required (Figure 9). While dissolution of the support could be an issue at such extreme pH, it was reasoned that the buffering effect of the support would result in relatively fast pH equilibration, so that the pH drops to a value where the surface is charged but dissolution is less problematic. Using this method, 2 wt % Pt was deposited on carbon (PZC = 2, pH 13), silica (PZC = 4, pH 13), and alumina (PZC = 8, pH 1).

EXHIBIT J

Invalidity of U.S. Patent No. 11,224,864 in View of *Munnik Article*

'864 patent	***"Recent Developments in the Synthesis of Supported Catalysts"* ("*Munnik Article*")**
	Small particles between 1.5 and 3 nm in size were obtained, much smaller compared to those from a conventional dry impregnation whereby the pH of the impregnating solution was not optimized. Thus, CEDI provides the advantage of obtaining high dispersions without high amounts of wastewater, albeit at relatively low metal loadings." [Page 6699] "Recent improvements in spectroscopic techniques have enabled imaging of distributions after impregnation and drying, providing experimental evidence of the limited effect of drying on strongly adsorbing species. UV-vis and IR microspectroscopy showed nickel ethylenediamine complexes (0.5 M) with one or two en ligands grafted on γ-alumina did not redistribute during drying, whereas nonadsorbing $Ni(en)_3^{2+}$ migrated toward the outside of the support. Strongly adsorbing Pd species (0.1 M $PdCl_4^-$ solution) on alumina were studied with UV-vis microspectroscopy, and were also found to retain the distribution they had obtained during impregnation after drying. Using magnetic resonance imaging, cobalt nitrate at low concentration (0.2 M, pH 4) was found to adsorb sufficiently strongly to a γ-alumina surface to retain its eggshell distribution obtained during impregnation after drying. Thus, these studies confirm that, for strongly adsorbing species, the drying step does not alter the macroscopic distribution of the precursor to a large degree. For weakly adsorbing species and high concentrations, convection and back-diffusion during the drying step become very important. In the model of Liu et al., when the adsorption strength of nickel nitrate was reduced to a moderate value and assuming a homogeneous distribution after impregnation, drying could result in severe redistribution of the 0.25 M nickel nitrate solution on γ-alumina depending on the conditions. Drying at 70 °C resulted in an almost homogeneous distribution. However, increasing the drying temperature to 300 °C resulted in eggshell distributions, because back-diffusion of nickel nitrate was not fast enough to

EXHIBIT J

Invalidity of U.S. Patent No. 11,224,864 in View of *Munnik Article*

'864 patent	***"Recent Developments in the Synthesis of Supported Catalysts"* ("*Munnik Article*")**
	compensate for the high drying rate. Similar results were obtained upon alteration of the humidity of the drying air; at 0.5% humidity, drying was relatively fast, resulting in an eggshell distribution, while drying in 10% humidity resulted in an almost homogeneous distribution. Finally, the size of the support pellet had a great influence on the distribution. A pellet diameter of 1 mm resulted in a moderate eggshell distribution whereby the concentration of nickel nitrate in the shell was 1.5 times higher than in the center of the pellet. For a 10 mm pellet, the concentration in the shell was almost 3 times higher compared to that in the center. The model was recently extended to a 3 mm γ-alumina pellet impregnated with nickel nitrate concentrations ranging from 0.005 to 4.0 M (Figure 13), and many of the results were compared to experimental data." [Page 6700] "Other experimental evidence on redistribution of the precursor during drying has been reported in recent years. Impregnation of a basic AHM solution (1.8 M, pH 9) onto γ-alumina resulted in little interaction with the support, so that redistribution toward the external portion of the pellet resulted in eggshell distributions. Alternatively, addition of H_3PO_4 to an impregnation of γ-alumina with AHM resulted in surface adsorption of PO_4^{3-}, so that the support surface was not significantly charged even at pH 2, together with complex formation of the form $H_xP_2Mo_5O_{23}^{6-x-}$. While normally AHM would adsorb to the alumina surface at this low pH, the negatively charged complex only weakly interacted with the PO_4 surface groups, so that redistribution of the complexes also resulted in eggshell distributions. As mentioned earlier, weakly interacting $Ni(en)_3$ complexes redistributed during drying, resulting in eggshell pro files. Clearly, weakly interacting species can migrate over macroscopic distances during the drying step."

EXHIBIT J

Invalidity of U.S. Patent No. 11,224,864 in View of *Munnik Article*

'864 patent	***"Recent Developments in the Synthesis of Supported Catalysts"* ("*Munnik Article*")**
	[Page 6701] "The effect of precursor-support interactions was also investigated for a y-alumina support. While stagnant drying in a muffle oven between 60 to 120 °C always resulted in 8 nm cobalt particles located in 30-60 nm aggregates after calcination in a N_2 flow, drying in a fluidized bed between 25 and 100 °C resulted in the 8 nm particles being uniformly distributed throughout the support after calcination. At 150°C, aggregates of 30-40 nm size were found, somewhat smaller than on silica. Thus, it appeared that the increased support-precursor interactions of the γ-alumina inhibited aggregate formation. Moreover, although drying in stagnant air always resulted in aggregates, drying using a N_2 flow resulted in uniform distributions, showing the importance of rapid removal of water during the drying process. It should be noted that fast drying can have a negative effect on the macroscopic distribution across catalyst grains when precipitation occurs without sufficient time for back-diffusion, as mentioned in the previous section and observed for silica with larger pores of 23 nm." [Page 6703] "Furthermore, temperature-programmed calcination on cobalt nitrate melt infiltrated (see section 5.1) on γ-Al_2O_3, followed by drying at 85 °C in air, revealed the hydrolysis of cobalt nitrate during a NO thermal treatment and subsequent improved particle size after decomposition also occurred on γ-Al_2O_3. However, the resulting increase in cobalt specific surface area of up to 33% for catalysts prepared using NO was less compared to the increase with silica. Using temperature-programmed reduction-X-ray absorption fine structure (TPR-XAFS) spectroscopy, Jacobs et al. also observed that the effect of NO on the dispersion is larger on silica compared to γ-alumina, and attributed this to the strong interaction between γ-alumina and cobalt nitrate, which already results in relatively small particles. Therefore, it was postulated that the effect of NO would work best on supports such as silica that exhibit a weak interaction with the precursor."

EXHIBIT J

Invalidity of U.S. Patent No. 11,224,864 in View of *Munnik Article*

'864 patent	***"Recent Developments in the Synthesis of Supported Catalysts"* ("*Munnik Article*")**
	[Page 6708] "Similarly, iron oxide particles of 2-10 nm size deposited by wet impregnation on γ-Al_2O_3 showed similarly inhomogeneous distributions with particles aggregated and preferentially deposited on the external surface of the support. The same particles deposited on MCM-41 with a pore size of 3 nm exhibited homogeneous distributions, but were exclusively deposited on the external surface, not surprising considering the relatively small pore size in relation to the size of the nanoparticles. However, eggshell catalysts have also been observed on carbon model supports with pore sizes larger than the colloidal particles. Focused ion beam scanning electron microscopy (FIB-SEM) on a model carbon support with pore sizes between 75 and 250 nm impregnated with 15-30 nm Pd particles revealed almost exclusively eggshell distributions. Penetration of the pores was possible when a vacuum impregnation was performed; however, the resulting metal distribution remained inhomogeneous." [Page 6711] "Feng et al. used silica with a grain size between 75-200 μm as a support for deposition of Pd.[242] Prior to Pd deposition, the silica was first coated by a 0.5 nm thick layer of Al_2O_3 using trimethylaluminum (TMA), or ZnO using diethyl zinc at 200°C, while water was used to decompose the precursors. Subsequently, 1.5-2.5 wt % Pd was deposited by one or two cycles of palladium hexafluoroacetylacetonate ($Pd(hfac)_2$) and formalin (formaldehyde and methanol in water). On the basis of a time-dependent deposition study, it was concluded that diffusion of the precursor into the support was the limiting step for these materials and conditions. On the Al_2O_3-coated silica, 1-2 nm Pd particles were found distributed uniformly across the support particles by scanning electron microscopy-energy- dispersive X-ray (SEM-EDX) spectroscopy, while no particles were detected on ZnO, suggesting an alloy formation. Using up to 25 cycles, it was found that the Pd particle size increased

'864 patent	***"Recent Developments in the Synthesis of Supported Catalysts"* ("*Munnik Article*")**
	almost linearly from 1.1 to 2.9 nm on Al_2O_3-coated silica. Moreover, an induction time was observed for the uptake of Pd, which increased after several cycles. It was postulated that when $Pd(hfac)_2$ is deposited, Al(hfac)* species are formed which are not removed during subsequent formalin exposure, and thus are poisoning the surface.[244] The formalin treatment only removed hfac ligands on the Pd and replaced them with hydrogen atoms, which formed new nucleation sites for further Pd cycles, resulting in an increased Pd uptake as particles became larger due to an increasing amount of nucleation sites for additional $Pd(hfac)_2$. A similar growth behavior was observed on MgO and TiO_2 layers; however, no Pd particles were observed on pure silica, suggesting a limited reactivity of the Si-OH hydroxyl groups with $Pd(hfac)_2$. A study of the deposition of Pt on γ-Al_2O_3, TiO_2 and $SrTiO_3$ using $MeCpPtMe_3$ and H_2 or N_2 cycles showed the Pt precursor can be adsorbed on oxidic supports at temperatures as low as 100 °C, and is decomposed at 200 °C in H_2 or 300 °C in N_2 to form metallic PT particles."

EXHIBIT J

Invalidity of U.S. Patent No. 11,224,864 in View of *Munnik Article*

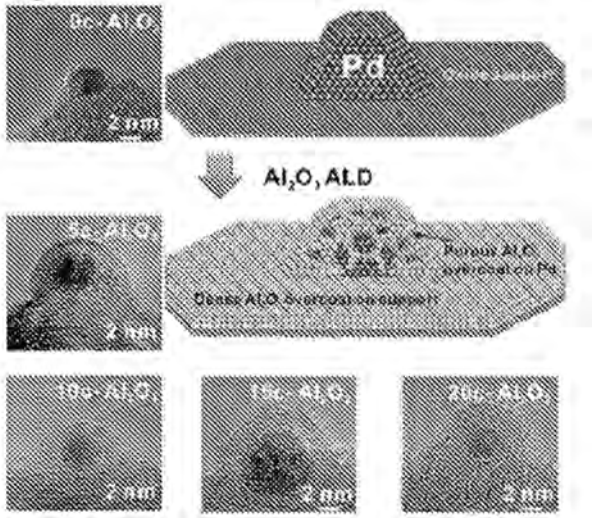
'864 patent	***"Recent Developments in the Synthesis of Supported Catalysts"* ("*Munnik Article*")**
	[Page 6712]  "Figure 23. Schematic representation of the deposition of a porous alumina layer on Pd particles supported on an oxidic support, and TEM images of spherical alumina-supported Pd particles with different numbers of $A1_2O_3$ layers deposited on top, represented by the number of cycles *(xc).* An increasingly thick layer was observed with increasing cycles; however, the particles remained accessible for catalysis. Adapted from ref 252. Copyright 2012 American Chemical Society." To the extent that this limitation is not disclosed, either explicitly or inherently, by *Munnik Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and

EXHIBIT J

Invalidity of U.S. Patent No. 11,224,864 in View of *Munnik Article*

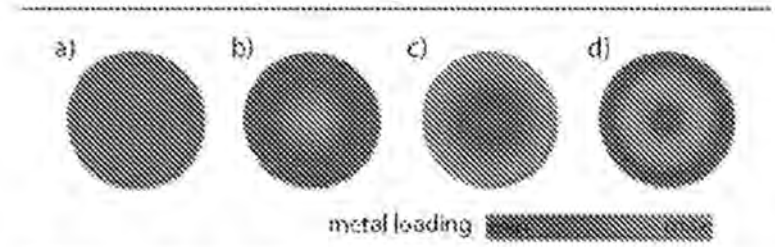
'864 patent	***"Recent Developments in the Synthesis of Supported Catalysts"* ("*Munnik Article*")**
	predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
1[c] one or more Group VIII noble metals distributed in the particle as an eggshell such that a higher concentration of the one or more Group VIII noble metals is present in the outer region of the microsphere sized porous particle as compared to the concentration of the one or more Group VIII noble metals in the centre of the microsphere sized porous particle.	*Munnik Article* discloses a combustion promoter comprising one or more Group VIII noble metals, the metals distributed in the particle as an eggshell such that a higher concentration of the one or more Group VIII noble metals is present in the outer region of the microsphere sized porous particle as compared to the concentration of the one or more Group VIII noble metals in the centre of the microsphere sized porous particle. *See, e.g.*, [Page 6688-6689] "Thus, for large catalyst bodies, a homogeneous distribution of the active phase is only preferential for relatively slow catalytic reactions, providing reactants time to diffuse to the active sites. If the reaction is fast or if secondary reactions are problematic, a different macroscopic distribution of the active phase might be preferred, such as an eggshell distribution or an egg-white distribution if the catalyst is prone to attrition. If the reactants contain poisons, an egg-yolk distribution may be favorable so that the poison can be captured at the edge of the catalyst body, where few active sites are present (Figure 1)."  **Figure 1.** Different types of macroscopic metal distributions across a support body: (a) homogeneous distribution, (b) egg-yolk distribution, (c) eggshell distribution, (d) egg-white distribution (red, high concentration of metal; blue, low concentration of metal).

EXHIBIT J

Invalidity of U.S. Patent No. 11,224,864 in View of *Munnik Article*

'864 patent	***"Recent Developments in the Synthesis of Supported Catalysts"* ("*Munnik Article*")**
	[Page 6695] "Recently, Regalbuto et al. used palladium, copper, cobalt, ruthenium, and nickel ammine complexes to obtain silica- supported catalysts at loadings between 1 and 3 wt %. After deposition at a pH of 12, the catalysts were dried and directly reduced. Small particles between 1 and 3 nm in size were found, and calculations showed that these metal loadings were close to the maximum obtainable by electrostatic adsorption for the used silica with a surface area of 180 m^2/g and a PZC of 4.0. It was postulated that the maximum loading mainly depends on the surface area of the support. A follow-up study focused on SBA-15 supports with different surface areas.[91] Ammine complexes were used to deposit Pd nanoparticles of 1.3 nm size at a relatively high loading of 8.7 wt % on 1000 m^2/g SBA- 15, and 1.9 nm Pt particles were synthesized on SBA-15 with a 600 m^2/g surface area with a loading of 9.2 wt %, showing high surface areas were indeed beneficial for adsorption. Co particles were also synthesized using this method, but larger particles of 7 nm size at a loading of 5.1 wt % resulted. This was attributed to particle growth that occurred during the high reduction temperature of 800 °C needed to reduce the strongly adsorbing cobalt complexes, whereas the Pt and Pd catalysts were reduced at 350 °C or lower. Moreover, dissolution of the silica support was already observed at pH 9, and 25% of the SBA-15 silica was dissolved after 1h at pH 10, the optimum pH for the uptake of metal cations. This led to a large increase in pore diameter and pore volume; however, this was not considered a problem due to the relatively thick walls of SBA-15. It was suggested to minimize the contact time to reduce this effect." [Page 6697] "Adsorption also plays a significant role in the metal distribution across macroscopic support grains or pellets. Although the pore filling with liquids of larger bodies occurs within a few seconds or minutes, adsorption on support sites often slows penetration of the precursor significantly. After short equilibration

'864 patent	***"Recent Developments in the Synthesis of Supported Catalysts"* ("*Munnik Article*")**
	times, it is possible that metal precursor complexes are mostly adsorbed in the outer shell of the support body, resulting in an eggshell distribution. The details of an eggshell distribution will strongly depend on the equilibrium between adsorbed and solvated species. A longer equilibration time leads to diffusion of the metal complex toward the center of the catalyst body due to concentration gradients if the adsorption is at least to some extent reversible, so that eventually the system will converge to a homogeneous distribution. Addition of a second component that adsorbs more strongly than the precursor can result in egg- white or egg-yolk distributions (Figure 1). The additional component adsorbs in the outer shell, leaving no sites available for the metal precursor complex, which diffuses inward where more adsorption sites are available. Although this has been known for a long time, the subsequent drying step is also reported to have a large impact on the distribution, and direct monitoring of the distribution during impregnation is now possible by advanced spectroscopic techniques that have recently been developed." [Page 6698] "In the previous study, competitive adsorption was also studied using H3PO4. This led to egg-yolk catalysts upon co-impregnation with Cr^{6+} species. Another example of competitive adsorption is Cl^- and $PdCl_4^{2-}$. Five minutes after impregnation, all $PdCl_4^{2-}$ had hydrolyzed to $PdCl_2(OH)_2^{2-}$ at the edge of the γ-Al_2O_3 pellet due to the basic surface OH groups, regardless of the Cl^- concentration and pH. After 2h, egg-white distributions were obtained at pH 1 because many sites at the edge of the pellet were protonated followed by adsorption of Cl^-, so that the $PdCl_2(OH)_2^{2-}$moved deeper into the pellet for adsorption on free sites. At pH 5, Cl^- adsorption was less favorable so that eggshell distributions were still detected. After 96 h each of the four main distributions (Figure 1) could be obtained depending on the conditions used. An egg-white distribution was obtained by strong and competitive Cl^- adsorption at low pH, which was limited to the outer shell of the pellet at low Cl^- concentration, while at higher Cl^- concentration more competitive adsorption

EXHIBIT J

Invalidity of U.S. Patent No. 11,224,864 in View of *Munnik Article*

'864 patent	***"Recent Developments in the Synthesis of Supported Catalysts"* ("*Munnik Article*")**
	resulted in an egg-yolk profile. At higher pH and low Cl^- concentration, limited competitive adsorption resulted in an eggshell catalyst, while increasing the Cl^- concentration resulted in a more homogeneous distribution." [Page 6699] "Recent improvements in spectroscopic techniques have enabled imaging of distributions after impregnation and drying, providing experimental evidence of the limited effect of drying on strongly adsorbing species. UV-vis and IR microspectroscopy showed nickel ethylenediamine complexes (0.5 M) with one or two en ligands grafted on γ-alumina did not redistribute during drying, whereas nonadsorbing $Ni(en)_3^{2+}$ migrated toward the outside of the support. Strongly adsorbing Pd species (0.1 M $PdCl_4^-$ solution) on alumina were studied with UV-vis microspectroscopy, and were also found to retain the distribution they had obtained during impregnation after drying. Using magnetic resonance imaging, cobalt nitrate at low concentration (0.2 M, pH 4) was found to adsorb sufficiently strongly to a γ-alumina surface to retain its eggshell distribution obtained during impregnation after drying. Thus, these studies confirm that, for strongly adsorbing species, the drying step does not alter the macroscopic distribution of the precursor to a large degree. For weakly adsorbing species and high concentrations, convection and back-diffusion during the drying step become very important. In the model of Liu et al., when the adsorption strength of nickel nitrate was reduced to a moderate value and assuming a homogeneous distribution after impregnation, drying could result in severe redistribution of the 0.25 M nickel nitrate solution on γ-alumina depending on the conditions. Drying at 70 °C resulted in an almost homogeneous distribution. However, increasing the drying temperature to 300 °C resulted in eggshell distributions, because back-diffusion of nickel nitrate was not fast enough to compensate for the high drying rate. Similar results were obtained upon alteration of the humidity of the drying air; at 0.5% humidity, drying was relatively fast, resulting

'864 patent	***"Recent Developments in the Synthesis of Supported Catalysts"* ("*Munnik Article*")**
	in an eggshell distribution, while drying in 10% humidity resulted in an almost homogeneous distribution. Finally, the size of the support pellet had a great influence on the distribution. A pellet diameter of 1 mm resulted in a moderate eggshell distribution whereby the concentration of nickel nitrate in the shell was 1.5 times higher than in the center of the pellet. For a 10 mm pellet, the concentration in the shell was almost 3 times higher compared to that in the center. The model was recently extended to a 3 mm γ-alumina pellet impregnated with nickel nitrate concentrations ranging from 0.005 to 4.0 M (Figure 13), and many of the results were compared to experimental data." [Page 6708] "Wet impregnation is commonly applied to deposit the colloids onto a support due to the relatively low nanoparticle concentrations in colloidal solutions necessary to prevent aggregation and precipitation. It can take a long time for all particles to migrate into the pores of the support because diffusivity of colloidal nanoparticles is relatively slow, which can be stimulated through use of sonication. The drying step can have a large influence on the distribution of the nanoparticles when particles are not strongly bound to the support, as has been demonstrated for the drying of colloidal wet films. Significant redistribution toward the external surface of the grains is possible since evaporation of the solvent in porous bodies usually occurs on the external surface (see section 4.3). Even so, few studies report the distribution of nanoparticles at the nanoscale and even less the macroscopic distribution across support grains. Co nanoparticles deposited on SBA-15 by wet impregnation followed by extensive washing resulted in aggregation of particles in some areas, while other areas of the support remained empty. Similarly, iron oxide particles of 2-10 nm size deposited by wet impregnation on γ-Al_2O_3 showed similarly inhomogeneous distributions with particles aggregated and preferentially deposited on the external surface of the support. The same particles deposited on MCM-41 with a pore size of 3 nm exhibited homogeneous distributions, but were exclusively

'864 patent	***"Recent Developments in the Synthesis of Supported Catalysts"* ("*Munnik Article*")**
	deposited on the external surface, not surprising considering the relatively small pore size in relation to the size of the nanoparticles. However, eggshell catalysts have also been observed on carbon model supports with pore sizes larger than the colloidal particles. Focused ion beam scanning electron microscopy (FIB-SEM) on a model carbon support with pore sizes between 75 and 250 nm impregnated with 15-30 nm Pd particles revealed almost exclusively eggshell distributions. Penetration of the pores was possible when a vacuum impregnation was performed; however, the resulting metal distribution remained inhomogeneous." [Page 6711] "Feng et al. used silica with a grain size between 75-200 μm as a support for deposition of Pd. Prior to Pd deposition, the silica was first coated by a 0.5 nm thick layer of Al_2O_3 using trimethylaluminum (TMA), or ZnO using diethyl zinc at 200°C, while water was used to decompose the precursors. Subsequently, 1.5-2.5 wt % Pd was deposited by one or two cycles of palladium hexafluoroacetylacetonate ($Pd(hfac)_2$) and formalin (formaldehyde and methanol in water). On the basis of a time-dependent deposition study, it was concluded that diffusion of the precursor into the support was the limiting step for these materials and conditions. On the Al_2O_3-coated silica, 1-2 nm Pd particles were found distributed uniformly across the support particles by scanning electron microscopy-energy- dispersive X-ray (SEM-EDX) spectroscopy, while no particles were detected on ZnO, suggesting an alloy formation. Using up to 25 cycles, it was found that the Pd particle size increased almost linearly from 1.1 to 2.9 nm on Al_2O_3-coated silica. Moreover, an induction time was observed for the uptake of Pd, which increased after several cycles. It was postulated that when $Pd(hfac)_2$ is deposited, Al(hfac)* species are formed which are not removed during subsequent formalin exposure, and thus are poisoning the surface.[244] The formalin treatment only removed hfac ligands on the Pd and replaced them with hydrogen atoms, which formed new nucleation sites for further Pd cycles, resulting in an increased Pd uptake as particles became larger due to an increasing

'864 patent	***"Recent Developments in the Synthesis of Supported Catalysts"* ("*Munnik Article*")**
	amount of nucleation sites for additional $Pd(hfac)_2$. A similar growth behavior was observed on MgO and TiO_2 layers; however, no Pd particles were observed on pure silica, suggesting a limited reactivity of the Si-OH hydroxyl groups with $Pd(hfac)_2$. A study of the deposition of Pt on γ-Al_2O_3, TiO_2 and $SrTiO_3$ using $MeCpPtMe_3$ and H_2 or N_2 cycles showed the Pt precursor can be adsorbed on oxidic supports at temperatures as low as 100 °C, and is decomposed at 200 °C in H_2 or 300 °C in N_2 to form metallic PT particles." [Page 6711] "Mixed PtRu particles on Al_2O_3 were synthesized using alternating cycles of $MeCpPtMe_3$ and (2, 4-dimethylpentadienyl) (ethylcyclopentadienyl)ruthenium (Ru-(DER)), decomposing the precursor with oxygen in between. Two cycles of $Ru(DER)/O_2$ were followed by one cycle of $MeCpPtMe_3/O_2$ and one cycle of $Ru(DER)/O_2$ for a final 1:1 molar ratio, and resulted in 1- 2 nm particles with good mixing of Ru and Pt as determined by EXAFS analysis. PtPd bimetallic particles have also been synthesized using ALD on SiO_2 covered with Al_2O_3 or TiO_2 supports, creating 1-2 nm particles. However, as soon as a reduction treatment was performed, the particles rearranged to a Pt core/Pd shell nanostructure independent of the deposition sequence. Although most studies have been performed on noble-metal nanoparticles, non-noble-metal particles can also be synthesized. Co_3O_4 was deposited on Al2O3-coated and uncoated silica gel to 25 wt % using bis(cyclopentadienyl)cobalt and ozone. Using ALD, the particle size was reduced 2-fold compared to that obtained with incipient wetness impregnation, from 10-11 to 5-6 nm. However, IWI-prepared catalysts were more active for the Fischer-Tropsch reaction. This was attributed to retarded reduction of Co_3O_4 to Co for catalysts prepared by ALD, which was mitigated somewhat by Pt addition."

EXHIBIT J

Invalidity of U.S. Patent No. 11,224,864 in View of *Munnik Article*

'864 patent	***"Recent Developments in the Synthesis of Supported Catalysts"* ("*Munnik Article*")**
	To the extent that this limitation is not disclosed, either explicitly or inherently, by *Munnik Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[3] The combustion promoter according to claim 1, comprising a plurality of microsphere sized porous particles comprising alumina.	*Munnik Article* discloses a combustion promoter comprising a plurality of microsphere sized porous particles comprising alumina. *See, e.g.*, The support provided above for Claim 1[b]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Munnik Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[6] The combustion promoter according to claim 1, wherein the microsphere sized porous particles collectively have an average (D50) size of between 60 and 90 microns as measured by laser diffraction.	*Munnik Article* discloses a combustion promoter comprising microsphere sized porous particles, wherein the microsphere sized porous particles collectively have an average (D50) size of between 60 and 90 microns as measured by laser diffraction. *See, e.g.*, The support provided above for Claim 1[a].

EXHIBIT J

Invalidity of U.S. Patent No. 11,224,864 in View of *Munnik Article*

'864 patent	***"Recent Developments in the Synthesis of Supported Catalysts"* ("*Munnik Article*")**
	To the extent that this limitation is not disclosed, either explicitly or inherently, by *Munnik Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[12] The combustion promoter according to claim 1, wherein the one or more Group VIII noble metals comprises one or more of platinum, palladium, iridium, ruthenium, and rhodium.	*Munnik Article* discloses a combustion promoter comprising one or more Group VIII noble metals, wherein the one or more Group VIII noble metals comprises one or more of platinum, palladium, iridium, ruthenium, and rhodium. *See, e.g.*, The support provided above for Claim 1[c]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Munnik Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[15] The combustion promoter according to claim 12, wherein the one or more Group VIII noble metals comprises palladium.	*Munnik Article* discloses a combustion promoter comprising one or more Group VIII noble metals, wherein the one or more Group VIII noble metals comprises palladium. *See, e.g.*, The support provided above for Claim 1[c].

EXHIBIT J

Invalidity of U.S. Patent No. 11,224,864 in View of *Munnik Article*

'864 patent	***"Recent Developments in the Synthesis of Supported Catalysts"* ("*Munnik Article*")**
	To the extent that this limitation is not disclosed, either explicitly or inherently, by *Munnik Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.

EXHIBIT K

EXHIBIT K

Invalidity of U.S. Patent No. 11,224,864 in View of *Papageorgiou Article*

"*Preparation of Pt/γ-Al_2O_3 Pellets with Internal Step-Distribution of Catalyst: Experiments and Theory*" ("*Papageorgiou Article*"), authored by Panayiotis Papageorgiou et al. and published in 1996, qualifies as prior art of U.S. Patent No. 11,224,864 ("the '864 patent") under at least 35 U.S.C. §§ 102 and/or 103 for the asserted claims of the '864 patent, as discussed in detail in the claim chart below.

The Court has not yet construed the claims and therefore the meaning of the terms in the claims has yet to be resolved by the Court. The support identified here for these limitations is responsive to Plaintiffs' infringement contentions, which Defendant disagrees with. As such, nothing in Defendant's claim charts should be construed as an admission regarding infringement, either literally or under the doctrine of equivalents, or as an admission regarding Defendant's understanding of the proper scope of the asserted claims. Defendant reserves the right to rely on additional citations or sources of evidence that also may be applicable, or that may become applicable in light of claim construction, changes in Plaintiffs' infringement contentions, and/or information obtained during discovery as the case progresses.

To the extent Plaintiffs allege that *Papageorgiou Article* does not disclose any particular limitation of the asserted claims of the '864 patent, either expressly or inherently, it would have been obvious to a person of ordinary skill in the art as of the priority date of the '864 patent to modify *Papageorgiou Article* and/or to combine the teachings of *Papageorgiou Article* with other prior art references, including but not limited to the present prior art references cited in the Cover Pleading and the relevant section(s) of claim charts for other prior art references for the '864 patent in a manner that would have rendered the asserted claims invalid as obvious.

Defendant reserves the right to amend or supplement this claim chart at a later date as more fully set forth in the Cover Pleading.

EXHIBIT K

Invalidity of U.S. Patent No. 11,224,864 in View of *Papageorgiou Article*

'864 patent	***"Preparation of Pt/γ-Al_2O_3 Pellets with Internal Step-Distribution of Catalyst: Experiments and Theory"* ("*Papageorgiou Article*")**
1[a] A CO to CO_2 combustion promoter comprising microsphere sized porous particles, each microsphere sized porous particle having a diameter of less than 1mm, and independently comprising	*Papageorgiou Article* discloses a CO to CO_2 combustion promoter comprising microsphere sized porous particles, each microsphere sized porous particle having a diameter of less than 1mm, and independently comprising the limitations discussed in the following sections. *See, e.g.*, [Page 1-2] "A common method of catalyst preparation is by impregnation. A high-surface-area porous support is contacted with a liquid solution of active component. The active agent enters the porous support via capillary action and diffusion and adsorbs on the available surface sites. Supported platinum catalysts are usually prepared by contacting hexachloroplatinic acid with either a dry or solvent-filled support. When γ-Al_2O_3 pellets are impregnated with hexachloroplatinic acid, the resulting catalyst profile is of the eggshell type; i.e., the platinum is located in a thin shell at the support surface. Maatman (6) found that the platinum distribution could be made more uniform by the addition of various acids to the impregnating solution. Michalko (7) and others *(cf.* (8)) demonstrated that the presence of citric acid in the impregnation solution results in a subsurface band of platinum in alumina pellets. Shyr and Ernst (9) have shown that a variety of platinum profile shapes can be obtained by using different coimpregnants. In all of the above investigators, the preparation of the supported platinum catalyst was based on the coimpregnation of hexachloroplatinic acid with another acid (usually citric acid). Lee and Aris (10) have provided an extensive review of catalyst preparation and have described models for various phases of single- and multicomponent impregnation." [Page 8-9] "Note that even though during the experiments the pellets were initially dry, thus some of the solution entered by imbibition, Eq. [15] assumes that the pores of the pellet are initially filled with solvent. Lee and Aris (10) showed by numerical calculations that for the impregnation of γ-Al_2O_3 pellets with hexachloroplatinic acid in which the pore-

'864 patent	***"Preparation of Pt/γ-Al_2O_3 Pellets with Internal Step-Distribution of Catalyst: Experiments and Theory"* ("*Papageorgiou Article*")**
	filling time was approximately 20 s, there was no noticeable difference between the platinum distributions in solvent-filled and dry supports when the impregnation period lasted 5 min or more. The reason for this is the short pore-filling time of the pellets. During the porn-filling stage, hexachloroplatinic acid adsorbs near the external surface of the pellet; thus the pores become filled with pure solvent. In our experiments the average pore-filling time was 12 s, while the impregnation periods varied from 15 min to 2.5 h; thus using a solvent-filled pellet to describe the impregnation of dry pellets does not induce any significant error." To the extent that this limitation is not disclosed, either explicitly or inherently, by *Papageorgiou Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
1[b] silica, alumina, or mixtures thereof; and	*Papageorgiou Article* discloses a combustion promoter comprising silica, alumina, or mixtures thereof. *See, e.g.*, [Page 1-2] "A common method of catalyst preparation is by impregnation. A high-surface-area porous support is contacted with a liquid solution of active component. The active agent enters the porous support via capillary action and diffusion and adsorbs on the available surface sites. Supported platinum catalysts are usually prepared by contacting hexachloroplatinic acid with either a dry or solvent-filled support. When γ-Al_2O_3 pellets are impregnated with hexachloroplatinic acid, the resulting catalyst profile is of the eggshell type; i.e., the platinum is located in a thin shell at the support surface. Maatman (6) found that the platinum distribution could be made more uniform by the

EXHIBIT K

Invalidity of U.S. Patent No. 11,224,864 in View of *Papageorgiou Article*

'864 patent	***"Preparation of Pt/γ-Al_2O_3 Pellets with Internal Step-Distribution of Catalyst: Experiments and Theory"* ("*Papageorgiou Article*")**
	addition of various acids to the impregnating solution. Michalko (7) and others *(cf.* (8)) demonstrated that the presence of citric acid in the impregnation solution results in a subsurface band of platinum in alumina pellets. Shyr and Ernst (9) have shown that a variety of platinum profile shapes can be obtained by using different coimpregnants. In all of the above investigators, the preparation of the supported platinum catalyst was based on the coimpregnation of hexachloroplatinic acid with another acid (usually citric acid). Lee and Aris (10) have provided an extensive review of catalyst preparation and have described models for various phases of single- and multicomponent impregnation." [Page 2] "The present work addresses the preparation of step-type Pt /γ-Al_2O_3 pellets by impregnation. A diffusion-- adsorption model which predicts satisfactorily the catalyst distribution inside the pellet is proposed. The parameters of the model, i.e., adsorption constants and effective diffusivities, were determined from separate experiments. In addition to coimpregnation, sequential impregnation was also used to prepare step-type catalyst pellets and yielded thinner step widths as compared with the former." [Page 2] "Thin step-type Pt /γ-Al_2O_3 catalysts were prepared by coimpregnation and sequential impregnation techniques using citric acid as the competitive adsorbate. Harshaw-Filtrol AL-0104 cylindrical γ-Al_2O_3 pellets were used, and their physical characteristics are given in Table 1." [Page 4] "When comparing the adsorption data from various γ-Al_2O_3 supports, caution should be exercised due to the variances that may be present in the supports. Jiratova (22) demonstrated that the isoelectric point (the pH where no adsorption occurs) of alumina varies strongly with trace impurities in the solid and the value of isoelectric point

EXHIBIT K

Invalidity of U.S. Patent No. 11,224,864 in View of *Papageorgiou Article*

'864 patent	***"Preparation of Pt/γ-Al_2O_3 Pellets with Internal Step-Distribution of Catalyst: Experiments and Theory"* ("*Papageorgiou Article*")**
	influences the amount of adsorption that occurs for a given solution pH. Furthermore, the solution pH also greatly affects the adsorption properties of a given γ-Al_2O_3 sample (23) and in tum depends on the hexachloroplatinic acid concentration. In addition, even for a specific concentration of hexachloroplatinic acid, solely the amount of γ-Al_2O_3 added can alter the solution pH; i.e., the pH depends on the ratio of solution volume to mass of alumina *(V/G)* (24). These investigators have shown that when alumina is added gradually to a solution, its pH changes and approaches the isoelectric point of the oxide. Considering the fact that the data shown in Table 2 were obtained over a large range of *V/G* values (5-80 ml/g) and with different alumina samples, the large range of adsorption parameters obtained may be justifiable." [Page 7] "At the onset of citric acid impregnation, the fluid-phase concentration of citric acid is relatively high, thus causing some desorption of the platinum from the γ-Al_2O_3 support. As the solution concentration of citric acid declines due to adsorption, there is a readsorption of platinum, which decreases the liquid-phase concentration of hexachloroplatinic acid, as also observed in the experimental data." [Page 9] "The diffusion-adsorption model described previously predicted successfully the effects of the various impregnation parameters on the platinum distribution within the pellet. The cross sections of pellets prepared by sequential and coimpregnation techniques are shown in Fig, 5. The dark rings in Figs. 5a-5i are the platinum catalyst, while the lighter portion is the γ-Al_2O_3 support. For comparison, eggshell, egg-yolk, and uniform pellets are also shown in Figs. 5j-5l." To the extent that this limitation is not disclosed, either explicitly or inherently, by *Papageorgiou Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the

EXHIBIT K

Invalidity of U.S. Patent No. 11,224,864 in View of *Papageorgiou Article*

'864 patent	***"Preparation of Pt/γ-Al_2O_3 Pellets with Internal Step-Distribution of Catalyst: Experiments and Theory"* ("*Papageorgiou Article*")**
	time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
1[c] one or more Group VIII noble metals distributed in the particle as an eggshell such that a higher concentration of the one or more Group VIII noble metals is present in the outer region of the microsphere sized porous particle as compared to the concentration of the one or more Group VIII noble metals in the centre of the microsphere sized porous particle.	*Papageorgiou Article* discloses a combustion promoter comprising one or more Group VIII noble metals, the metals distributed in the particle as an eggshell such that a higher concentration of the one or more Group VIII noble metals is present in the outer region of the microsphere sized porous particle as compared to the concentration of the one or more Group VIII noble metals in the centre of the microsphere sized porous particle. *See, e.g.*, [Page 1] "Catalysts involving group VIII metals are usually prepared by dispersing the metal on a high-surface-area support, so that a large fraction of metal atoms becomes avail- able for reaction. The supports used in industrial reactors are generally spherical or cylindrical pellets, with the catalytic agent deposited on the external surface or uniformly within the pellet. The location of the catalytic material in the support can have a significant effect on catalyst performance, especially when diffusional resistances during reaction are important. For an isothermal reaction exhibiting positive-order kinetics, it is desirable to place the catalyst on the external surface of the support where the reaction concentration is the highest, while for negative-order kinetics (as in some concentration regimes of bimolecular Langmuir-Hin-shelwood reactions), it is beneficial to locate the catalyst in the interior of the support where reactant concentration is lower (1). For exothermic reactions, even if they are of positive order, catalyst effectiveness can be increased by locating the catalytic agent inside the support, thus taking advantage of heat transport resistances (2)."

EXHIBIT K

Invalidity of U.S. Patent No. 11,224,864 in View of *Papageorgiou Article*

'864 patent	***"Preparation of Pt/γ-Al_2O_3 Pellets with Internal Step-Distribution of Catalyst: Experiments and Theory"* ("*Papageorgiou Article*")**
	[Page 1-2] "A common method of catalyst preparation is by impregnation. A high-surface-area porous support is contacted with a liquid solution of active component The active agent enters the porous support via capillary action and diffusion and adsorbs on the available surface sites. Supported platinum catalysts are usually prepared by contacting hexachloroplatinic acid with either a dry or solvent-filled support. When γ-Al_2O_3 pellets are impregnated with hexachloroplatinic acid, the resulting catalyst profile is of the eggshell type; i.e., the platinum is located in a thin shell at the support surface. Maatman (6) found that the platinum distribution could be made more uniform by the addition of various acids to the impregnating solution. Michalko (7) and others *(cf.* (8)) demonstrated that the presence of citric acid in the impregnation solution results in a subsurface band of platinum in alumina pellets. Shyr and Ernst (9) have shown that a variety of platinum profile shapes can be obtained by using different coimpregnants. In all of the above investigators, the preparation of the supported platinum catalyst was based on the coimpregnation of hexachloroplatinic acid with another acid (usually citric acid). Lee and Aris (10) have provided an extensive review of catalyst preparation and have described models for various phases of single- and multicomponent impregnation." [Page 2] "At the end of the coimpregnation or the sequential impregnation period, the pellets were dried at 125°C for 18 h to immobilize both the citrate and the chloroplatinate ions. For visual observation the pellets were sectioned radially and dipped in boiling $SnCl_2$ solution. The location and width of the resulting platinum profile were determined by photographing the pellet and measuring the distances from the photographs. The pellets were calcined under a stream of oxygen from 25 to 500°C over a period of half an hour, followed by treatment in hydrogen for 1h at 500°C. After this, the pellets were cooled in a nitrogen atmosphere.

EXHIBIT K

Invalidity of U.S. Patent No. 11,224,864 in View of *Papageorgiou Article*

'864 patent	***"Preparation of Pt/γ-Al_2O_3 Pellets with Internal Step-Distribution of Catalyst: Experiments and Theory"* ("*Papageorgiou Article*")**
	Measurement of the platinum loading was performed by first crushing the pellets and extracting the platinum by a mixture of double-distilled HCl, HNO_3, and HF, in a ratio 3: 3: 1. The concentration of platinum in solution was then determined by ICP mass spectroscopy. The catalyst dispersion was determined by measuring the platinum surface area by using hydrogen chemisorption according to the procedure described by Lemaitre *et al.* (17). Details of the pulse chemisorption setup used for the experiments are given elsewhere (18). 'The catalyst in powder form was first treated in hydrogen at 450°C for 1 h and after cooling to 25°C, was subjected to an oxidation-reduction-oxidation cycle, to obtain reproducible results." [Page 6] "The amount of citric acid adsorbed during the multicomponent experiments was less than the amount which would be expected based on a purely competitive model described by Eq. [6] for citric acid and Eq. [7] for hexachloroplatinic acid. Such behavior could be due to steric hindrances provided by the platinum adsorbed on the surface of γ-Al_2O_3." [Page 9-11] "The diffusion-adsorption model described previously predicted successfully the effects of the various impregnation parameters on the platinum distribution within the pellet. The cross sections of pellets prepared by sequential and coimpregnation techniques are shown in Fig, 5. The dark rings in Figs. 5a-5i are the platinum catalyst, while the lighter portion is the γ-Al_2O_3 support. For comparison, eggshell, egg-yolk, and uniform pellets are also shown in Figs. 5j-5l.

EXHIBIT K

Invalidity of U.S. Patent No. 11,224,864 in View of *Papageorgiou Article*

'864 patent	***"Preparation of Pt/γ-Al_2O_3 Pellets with Internal Step-Distribution of Catalyst: Experiments and Theory"* ("*Papageorgiou Article*")**
	For both types of impregnations, the platinum band was located deeper inside the pellet for higher initial critic acid concentration (compare Figs. 5b-5c, 5e-5f, 5h-5i). In addition, higher impregnation time forced the platinum to move closer to the pellet center, since the citric acid could diffuse further inside the pellet and displace the adsorbed platinum (compare Figs. 5a-5b, 5d-5e, 5g-5h). Similar effects during coimpregnation of alumina pellets with hexachloroplatinic and citric acid were also observed by Becker and Nuttall (8). The initial hexachloroplatinic acid concentration did not have any noticeable effect on the catalyst location; how- ever, it resulted in catalysts with higher loading and somewhat larger bandwidths (compare Figs. 5a-5d, 5b-5e, 5c- 5f). The bandwidth was also influenced by impregnation time, owing primarily to geometric effects. Since the diameter of the ring, where the platinum was located, decreased with time, its width increased to accommodate the deposited metal (compare Figs. 5a-5b, 5d-5e, 5g-5h). In general, for the same impregnation time and adsorbate concentrations, a similar penetration depth was achieved by both types of impregnations (compare Figs. 5a-5g, 5b-5h), but the bandwidth was smaller (as small as 50 μm) when using sequential impregnation. In coimpregnation experiments the platinum loading of the samples was affected not only by the initial hexachloro-platinic acid concentration but also to a smaller extent, by the citric acid concentration. The loading decreased with higher initial concentration of citric acid. However, during sequential impregnation, the platinum loading of the pellets depended primarily on the conditions of the first impregnation step. It could be kept approximately constant by using the same hexachloroplatinic acid concentration for the first step. The fact that citric acid concentration did not affect the platinum loading of pellets prepared by sequential impregnation indicates a different mode of action: in coimpregnation it competes with hexachloroplatinic acid for adsorption on the support, while in sequential impregnation it enhances the desorption of adsorbed

EXHIBIT K

Invalidity of U.S. Patent No. 11,224,864 in View of *Papageorgiou Article*

'864 patent	***"Preparation of Pt/γ-Al_2O_3 Pellets with Internal Step-Distribution of Catalyst: Experiments and Theory"* ("*Papageorgiou Article*")**
	platinum. The platinum loading of the pellets as measured by ICP mass spectroscopy was in the range 0.1-0.4 wt%, depending on the initial hexachloroplatinic acid concentration."

EXHIBIT K

Invalidity of U.S. Patent No. 11,224,864 in View of *Papageorgiou Article*

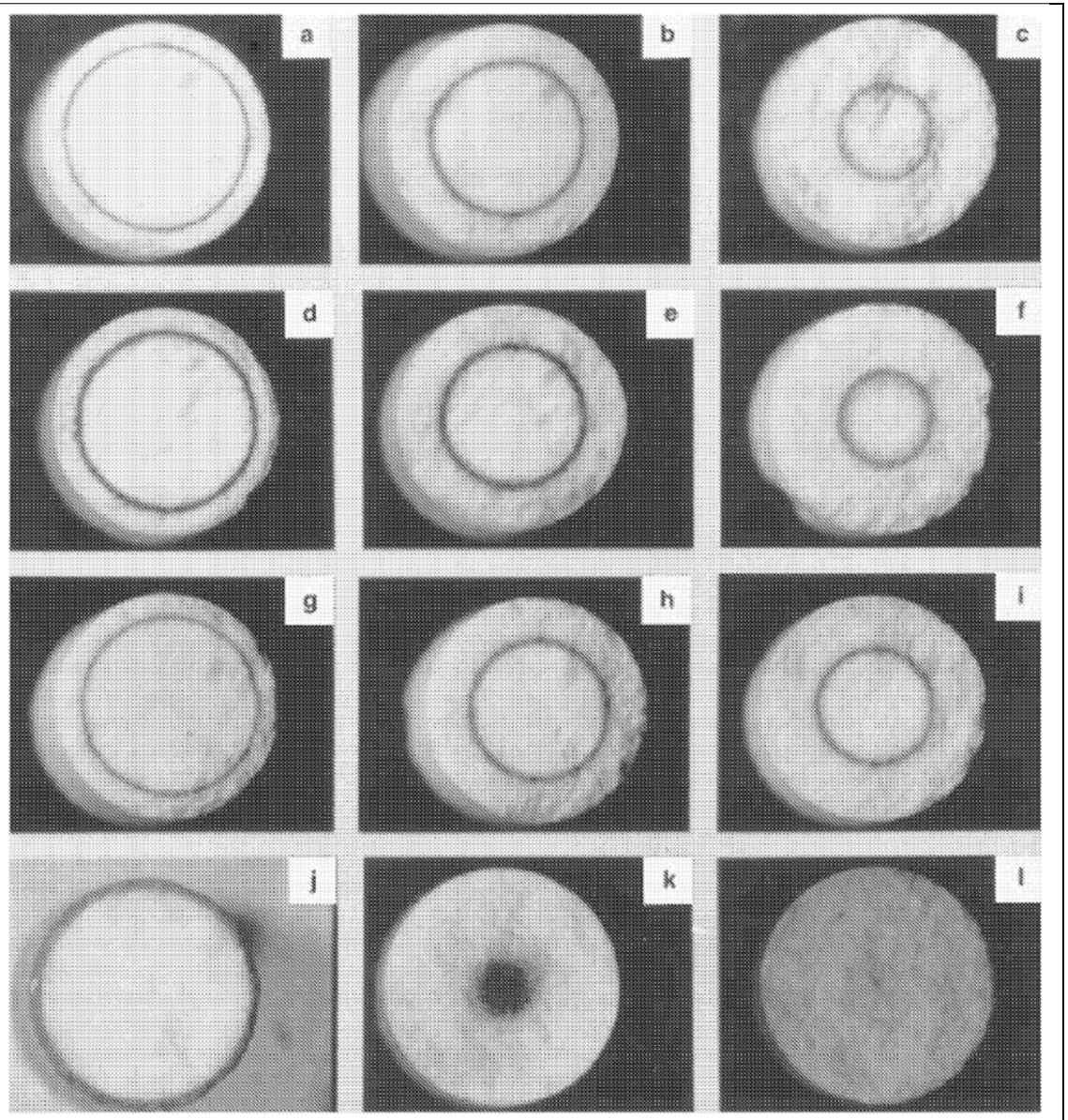
	

EXHIBIT K

Invalidity of U.S. Patent No. 11,224,864 in View of *Papageorgiou Article*

'864 patent	***"Preparation of Pt/γ-Al_2O_3 Pellets with Internal Step-Distribution of Catalyst: Experiments and Theory"* ("*Papageorgiou Article*")**
	[Page 11] "The eggshell distribution (Fig. 5j) was prepared by impregnating the pellet in a solution of hexachloroplatinic acid alone. The egg-yolk distribution (Fig. 5k) could be achieved by both sequential and coimpregnation, provided that the impregnation periods were sufficiently long, while the uniform distribution (Fig. 51) was obtained using the coimpregnation method. Note that the difference in the preparation procedure of pellets depicted in Fig. 5k and Fig. 5l is only the impregnation duration. Thus, for short coimpregnation time an egg-white deposition is obtained, while for longer impregnation periods egg-yolk and ultimately uniform distributions are attained. Catalyst dispersion of the pellets, including both step and uniform distributions, as measured by hydrogen chemisorption, ranged between 60 and 70%. Hence, for pellets with relatively low platinum loadings as considered in this study, the dispersion is virtually unaltered when concentrating the metal in a thin step-type distribution. These dispersion values are within the range reported for typical Pt/ γ-Al_2O_3 catalysts (cf. (30, 31)). Figures 6 and 7 show the predicted dimensionless platinum surface concentration profiles, along with the experimentally measured platinum band positions denoted by the shadowed regions for the sequential and coimpregnation studies, respectively. The numbers on the bands give the impregnation time (for sequential impregnation it is the time in the citric add solution). The penetration depth of platinum predicted by the model for both impregnation techniques is greater with increasing impregnation time, as also observed experimentally. In addition, the model is able to predict that the bandwidths are broader for the coimpregnation than for the sequential impregnation technique (compare Figs. 6b and 7b), as well as the fact that bandwidth increases with impregnation time. Note that the model predicts that some adsorbed platinum extends from the peak to the pellet surface, with coimpregnation exhibiting more platinum in that region than sequential impregnation. Our pellets showed evidence (i.e., light-gray

EXHIBIT K

Invalidity of U.S. Patent No. 11,224,864 in View of *Papageorgiou Article*

'864 patent	***"Preparation of Pt/γ-Al_2O_3 Pellets with Internal Step-Distribution of Catalyst: Experiments and Theory"* ("*Papageorgiou Article*")**
	color) of small amounts of platinum deposited between the step distribution and the pellet external surface. This was also observed previously by Kunimori *et al.* (32) in the preparation of similar catalysts using hexachloroplatinic acid and citric acid."

EXHIBIT K

Invalidity of U.S. Patent No. 11,224,864 in View of *Papageorgiou Article*

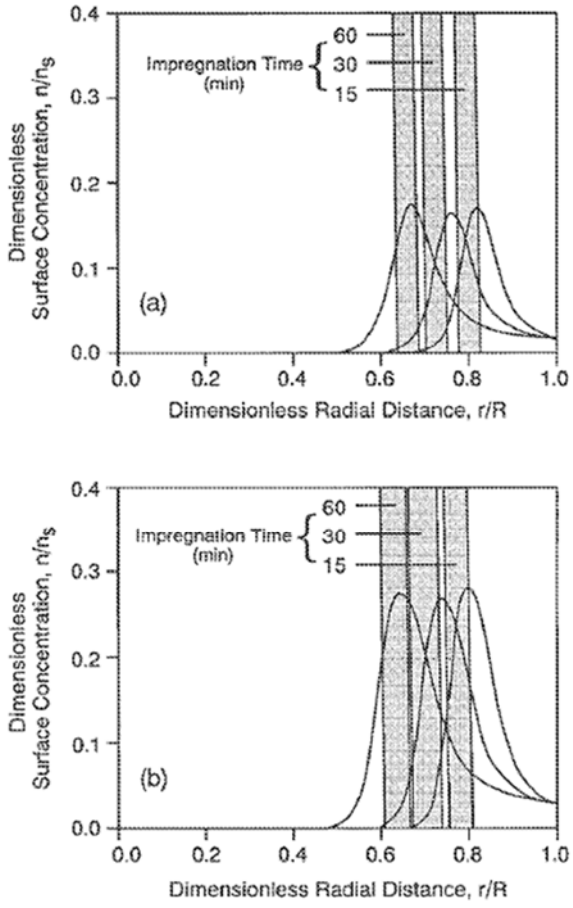
'864 patent	***"Preparation of Pt/γ-Al_2O_3 Pellets with Internal Step-Distribution of Catalyst: Experiments and Theory"* ("*Papageorgiou Article*")**
	 FIG. 6. Comparison of model predictions and experimental measurements of the surface concentration profiles of platinum for sequential impregnation with 0.1111 *M* citric acid, (a) 0.00589 *M* hexachloroplatinic acid, (b) 0.0117 *M* hexachloroplatinic acid. The shaded bands indicate the experimentally measured platinum step positions, and the numbers on the bands give the impregnation time in the citric acid solution.

EXHIBIT K

Invalidity of U.S. Patent No. 11,224,864 in View of *Papageorgiou Article*

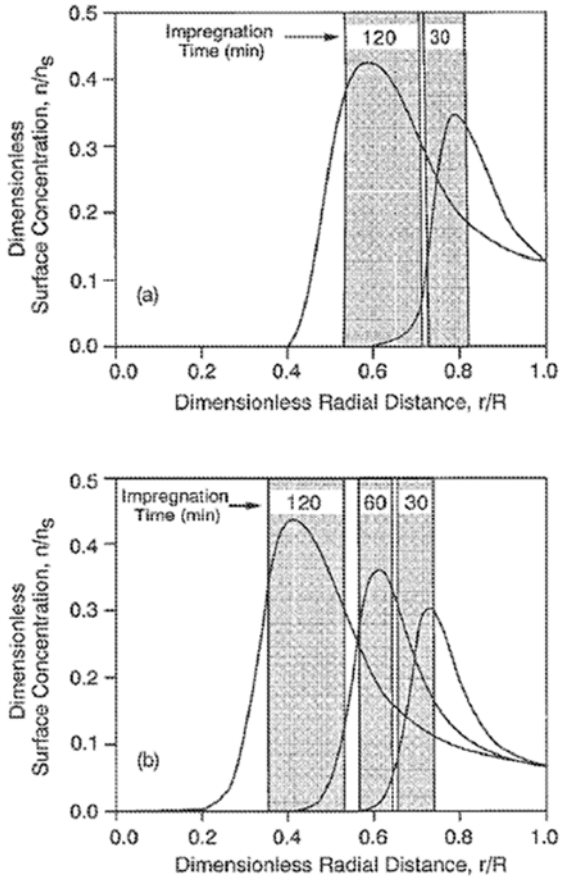
'864 patent	***"Preparation of Pt/γ-Al_2O_3 Pellets with Internal Step-Distribution of Catalyst: Experiments and Theory"* ("*Papageorgiou Article*")**
	 FIG. 7. Comparison of model predictions and experimental measurements of the surface concentration profiles of platinum for coimpregnation with 0.0117 *M* hexachloroplatinic acid, (a) 0.0556 *M* citric acid, (b) 0.1111 *M* citric acid. The shaded bands indicate the experimentally measured platinum step positions, and the numbers on the bands give the impregnation time.

EXHIBIT K

Invalidity of U.S. Patent No. 11,224,864 in View of *Papageorgiou Article*

'864 patent	***"Preparation of Pt/γ-Al_2O_3 Pellets with Internal Step-Distribution of Catalyst: Experiments and Theory"* ("*Papageorgiou Article*")**
	To the extent that this limitation is not disclosed, either explicitly or inherently, by *Papageorgiou Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[3] The combustion promoter according to claim 1, comprising a plurality of microsphere sized porous particles comprising alumina.	*Papageorgiou Article* discloses a combustion promoter comprising a plurality of microsphere sized porous particles comprising alumina. *See, e.g.*, The support provided above for Claim 1[b]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Papageorgiou Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[6] The combustion promoter according to claim 1, wherein the microsphere sized porous particles collectively have an average (D50) size of between 60 and 90 microns as measured by laser diffraction.	*Papageorgiou Article* discloses a combustion promoter comprising microsphere sized porous particles, wherein the microsphere sized porous particles collectively have an average (D50) size of between 60 and 90 microns as measured by laser diffraction. *See, e.g.*, The support provided above for Claim 1[a].

EXHIBIT K

Invalidity of U.S. Patent No. 11,224,864 in View of *Papageorgiou Article*

'864 patent	***"Preparation of Pt/γ-Al_2O_3 Pellets with Internal Step-Distribution of Catalyst: Experiments and Theory"* ("*Papageorgiou Article*")**
	To the extent that this limitation is not disclosed, either explicitly or inherently, by *Papageorgiou Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[12] The combustion promoter according to claim 1, wherein the one or more Group VIII noble metals comprises one or more of platinum, palladium, iridium, ruthenium, and rhodium.	*Papageorgiou Article* discloses a combustion promoter comprising one or more Group VIII noble metals, wherein the one or more Group VIII noble metals comprises one or more of platinum, palladium, iridium, ruthenium, and rhodium. *See, e.g.*, The support provided above for Claim 1[c]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Papageorgiou Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[15] The combustion promoter according to claim 12, wherein the one or more Group VIII noble metals comprises palladium.	*Papageorgiou Article* discloses a combustion promoter comprising one or more Group VIII noble metals, wherein the one or more Group VIII noble metals comprises palladium. *See, e.g.*, The support provided above for Claim 1[c].

EXHIBIT K

Invalidity of U.S. Patent No. 11,224,864 in View of *Papageorgiou Article*

'864 patent	***"Preparation of Pt/γ-Al_2O_3 Pellets with Internal Step-Distribution of Catalyst: Experiments and Theory"* ("*Papageorgiou Article*")**
	To the extent that this limitation is not disclosed, either explicitly or inherently, by *Papageorgiou Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.

EXHIBIT L

EXHIBIT L

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2009/0050529 ("*Rainer*")

U.S. Patent Application No. 2009/0050529 ("*Rainer*"), filed on June 8, 2007 and published on February 26, 2009, qualifies as prior art under at least 35 U.S.C. §§ 102 and/or 103 for the asserted claims of U.S. Patent No. 11,224,864 ("the '864 patent"), as discussed in detail in the claim chart below.

The Court has not yet construed the claims and therefore the meaning of the terms in the claims has yet to be resolved by the Court. The support identified here for these limitations is responsive to Plaintiffs' infringement contentions, which Defendant disagrees with. As such, nothing in Defendant's claim charts should be construed as an admission regarding infringement, either literally or under the doctrine of equivalents, or as an admission regarding Defendant's understanding of the proper scope of the asserted claims. Defendant reserves the right to rely on additional citations or sources of evidence that also may be applicable, or that may become applicable in light of claim construction, changes in Plaintiffs' infringement contentions, and/or information obtained during discovery as the case progresses.

To the extent Plaintiffs allege that *Rainer* does not disclose any particular limitation of the asserted claims of the '864 patent, either expressly or inherently, it would have been obvious to a person of ordinary skill in the art as of the priority date of the '864 patent to modify *Rainer* and/or to combine the teachings of *Rainer* with other prior art references, including but not limited to the present prior art references cited in the Cover Pleading and the relevant section(s) of claim charts for other prior art references for the '864 patent in a manner that would have rendered the asserted claims invalid as obvious.

Defendant reserves the right to amend or supplement this claim chart at a later date as more fully set forth in the Cover Pleading.

EXHIBIT L

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2009/0050529 ("*Rainer*")

'864 patent	**U.S. Patent Application No. 2009/0050529 ("*Rainer*")**
1[a] A CO to CO_2 combustion promoter comprising microsphere sized porous particles, each microsphere sized porous particle having a diameter of less than 1mm, and independently comprising	*Rainer* discloses a CO to CO_2 combustion promoter comprising microsphere sized porous particles, each microsphere sized porous particle having a diameter of less than 1mm, and independently comprising the limitations discussed in the following sections. *See, e.g.*, [Abstract] "Compositions suitable for use in FCC processes are provided that are capable of providing improved CO oxidation promotion activity along with NOx emission control. Generally, compositions are provided for promoting CO oxidation in FCC processes, wherein the compositions contain (i) a basic material support, (ii) at least one oxide of a lanthanide series element, (iii), optionally, at least one oxide of a transition metal selected from Groups Ib and IIb of the Periodic Table and (iv) at least one precious metal. The basic material support is preferably a hydrotalcite-like compound." [0008]-[0010] "Specific examples of treatments applied to regeneration operated in the complete combustion mode include the addition of a CO combustion promoter metal to the catalyst or to the regenerator. For example, U.S. Pat. No. 2,647,860 proposed adding 0.1 to 1 weight percent chromic oxide to a cracking catalyst to promote combustion of CO. U.S. Pat. No. 3,808,121 taught using relatively large-sized particles containing CO combustion-promoting metal into a regenerator. The small-sized catalyst is cycled between the cracking reactor and the catalyst regenerator while the combustion- promoting particles remain in the regenerator. Also, U.S. Pat. Nos. 4,072,600 and 4,093,535 teach the use of Pt, Pd, Ir, Rh, Os, Ru, and Re in cracking catalysts in concentrations of 0.01 to 50 ppm, based on total catalyst inventory to promote CO combustion in a complete bum unit. The use of precious metals to catalyze oxidation of carbon moNOxide in the regenerators of FCC units has gained broad commercial acceptance. Some of the

EXHIBIT L

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2009/0050529 ("*Rainer*")

'864 patent	U.S. Patent Application No. 2009/0050529 ("*Rainer*")
	history of this development is set forth in U.S. Pat. No. 4,171,286 and U.S. Pat. No. 4,222,856. In the earlier stages of the development, the precious metal was deposited on the particles of cracking catalyst. Present practice is generally to supply a promoter in the form of solid fluidizable particles containing a precious metal, such particles being physically separate from the particles of cracking catalyst. The precious metal or compound thereof, is supported on particles of suitable carrier material and the promoter particles are usually introduced into the regenerator separately from the particles of cracking catalyst. The particles of promoter are not removed from the system as fines and are cocirculated with cracking catalyst particles during the cracking/stripping/regeneration cycles. Judgment of the CO combustion efficiency of a promoter is done by the ability to control the difference in temperature, delta T, between the (hotter) dilute phase, cyclones or flue gas line, and the dense phase. Most FCC units now use a Pt CO combustion promoter. While the use of combustion promoters such as platinum reduce CO emissions, such reduction in CO emissions is usually accompanied by an increase in nitro- gen oxides (NOx) in the regenerator flue gas. Promoter products used on a commercial basis in FCC units include calcined spray dried porous microspheres of kaolin clay impregnated with a small amount (e.g., 100 to 1500 ppm) of platinum. Reference is made to U.S. Pat. No. 4,171,286 (supra). Most commercially used promoters are obtained by impregnating a source of platinum on micro- spheres of high purity porous alumina, typically gamma alumina. The selection of platinum as the precious metal in various commercial products appears to reflect a preference for this metal that is consistent with prior art disclosures that platinum is the most effective group VIII metal for carbon moNOxide oxidation promotion in FCC regenerators. See, for example, FIG. 3 in U.S. Pat. No. 4,107,032 and the same figure in U.S. Pat. No. 4,350,614. The figure illustrates the effect of increasing the concentration of various species of precious metal promoters from 0.5 to 10 ppm on CO_2/CO ratio."

EXHIBIT L

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2009/0050529 ("*Rainer*")

'864 patent	U.S. Patent Application No. 2009/0050529 ("*Rainer*")
	[0014] "Unfortunately, the more active combustion promoters such as platinum and palladium also serve to promote the formation of nitrogen oxides in the regeneration zone. It has been reported that the use of prior art CO promoters can cause a dramatic increase (e.g. >300%) in NOx." [0025]-[0028] "The present invention provides novel compositions for use in FCC processes that are capable of providing suitable improved CO oxidation promotion activity along with NOx emission control. In one aspect, the invention provides compositions for promoting CO oxidation in FCC processes, the compositions containing (i) basic material support, (ii) at least one oxide of a lanthanide series element, (iii), optionally, at least one oxide of a transition metal selected from Groups Ib and IIb of the Periodic Table and (iv) at least one precious metal. The basic material support is preferably a hydrotalcite-like compound. In another aspect, the invention encompasses FCC processes using the CO oxidation promotion compositions of this invention either as an integral part of the FCC catalyst particles or as separate particles admixed with the FCC catalyst. The composition provides lower NOx emissions than prior art CO oxidation promoters. In still another aspect of this invention, an additive for CO oxidation is provided by incorporating into a single particle, a platinum group metal for CO oxidation promotion and metal components for NOx decomposition, and in which the platinum group metal and other metals are distributed in separate areas of the particle to achieve improvements in NOx reduction."

EXHIBIT L

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2009/0050529 ("*Rainer*")

'864 patent	U.S. Patent Application No. 2009/0050529 ("*Rainer*")
	[0050] "The amount of precious metal can be varied considerably depending on the level of CO combustion promotion desired. Typical levels of precious metal will be in the range of 50 to 1500 ppm of total additive and/or catalyst. As used herein the precious metals include those selected from a group consisting of Ag, Cu, Pt, Pd, Ir, Os, Ru, Rh, Re and their precursors such as salts and amine complexes and mixtures thereof." [0052] "Where the additive composition is used as an additive particulate (as opposed to being integrated into the FCC catalyst particles themselves), the amount of additive component in the additive particles is preferably at least 50 wt %, more preferably at least 75 wt.%. Most preferably, the additive particles consist entirely of the additive component. The additive particles are preferably of a size suitable for circulation with the catalyst inventory in an FCC process. The additive particles preferably have an average particle size of about 20-200 μm. The additive particles preferably have attrition characteristics such that they can withstand the severe environment of an FCCU." [0059] "In another aspect of the present invention, a particulate additive for incorporation into an FCC process is described as having a specified distribution of precious metal so as to be effective in promoting the oxidation of carbon moNOxide while minimizing NOx production. The additive of this aspect of the present invention is based on the concept of distributing the precious metal CO oxidation promoter within the center of the particulate additive and distributing throughout the particle any metal or metal oxides which have been found useful in the decomposition *of* NOx. A graphic depiction of the additive of this invention is shown in FIG. 1. As depicted in FIG. 1, the porous, particulate additive 10 will include a precious metal designated 'Pt' such as previously described, which is primarily distributed in the

EXHIBIT L

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2009/0050529 ("*Rainer*")

'864 patent	U.S. Patent Application No. 2009/0050529 ("*Rainer*")
	center 12 of additive 10. What is theorized in accordance with this invention is that the precious metal such as Ag, Cu, Pt, Pd, Ir, Os, Ru, Rh, and/or Re distributed primarily in the central area 12 of the porous additive 10 will be able to promote the oxidation of CO to CO_2." [0060] "In general, however, the primary distribution of precious metal will be in the far interior of the porous particulate substrate whereas the other metals or metal oxides will be primarily distributed near the outer surface of the particle." [0061] "To achieve the desired distribution of metal catalytic components as in the additive of this invention as depicted in FIG. 1, any known method can be used by this invention. In one such method, the porous substrate can first be impregnated with the precious metal." [0062]-[0063] "It is also known to form an 'egg yolk' catalyst distribution in which a catalyst component, in this case, precious metal, is concentrated at the particle center. Such process involves impregnating the particle with a precious metal solution or complex, alone or with a co-impregnant. It is known that convective flow of the liquids through the particle can be affected by specific drying models that can effectively distribute a metal into the desired interior of a porous particle. Subsequent impregnation can distribute one or more additional metals upon the outer surfaces of the porous support. The effect of different drying processes on catalyst distribution is described in 'Predicting the Effect of Drying on Supported Coimpregnation Catalysts,' Lekhal et al., Ind. Eng. Chem. Res. 2001, 40, pgs. 3989-3999. This article is also incorporated in its entirety by reference herein.

EXHIBIT L

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2009/0050529 ("*Rainer*")

'864 patent	U.S. Patent Application No. 2009/0050529 ("*Rainer*")
	Internal distribution of a platinum group metal is also known by impregnating a porous support with a platinum group salt and an acid that competes with the platinum salt for adsorption within the porous support. For example, a high surface area porous support can be contacted with a liquid solution of the active component. The active component enters the porous support via capillary action and diffusion and absorbs on the available surface sites. When alumina supports are impregnated solely with hexachloroplatinic acid, it has been found that the resulting catalyst profile is typically of the 'eggshell' type, that being the platinum is located in a thin shell at the support surface. It has been known that the platinum distribution could be made more uniform or distributed in the center of a catalyst by co-impregnating the hexachloroplatinic acid with an acid such as HCl or citric acid wherein the platinum is now in a sub-surface band or within the core of the porous support. Other co-impregnants have been studied in order to change the distribution of the platinum component. Depending on the relative diffusion and adsorption strengths of the co-impregnants, the active component can be 'pushed' into the interior of the particle. A review of this type of process is described in 'Preparation of Pt/Gamma-Al2 O3 Pellets with Internal Step Distribution of Catalyst: Experiments and Theory,' Papageorgiou, et at., Journal of Catalysis 158, 1996, pgs 439-451, and 'Catalysis: An Integrated Approach, Second, revised and enlarged edition,' Netherlands Institute for Catalysis Research, 1999, Chapter 10, the entire contents of which are herein incorporated by reference." To the extent that this limitation is not disclosed, either explicitly or inherently, by *Rainer*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.

EXHIBIT L

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2009/0050529 ("*Rainer*")

'864 patent	U.S. Patent Application No. 2009/0050529 ("*Rainer*")
1[b] silica, alumina, or mixtures thereof; and	*Rainer* discloses a combustion promoter comprising silica, alumina, or mixtures thereof. *See, e.g.*, [Abstract] "Compositions suitable for use in FCC processes are provided that are capable of providing improved CO oxidation promotion activity along with NOx emission control. Generally, compositions are provided for promoting CO oxidation in FCC processes, wherein the compositions contain (i) a basic material support, (ii) at least one oxide of a lanthanide series element, (iii), optionally, at least one oxide of a transition metal selected from Groups Ib and IIb of the Periodic Table and (iv) at least one precious metal. The basic material support is preferably a hydrotalcite-like compound." [0010] "Promoter products used on a commercial basis in FCC units include calcined spray dried porous microspheres of kaolin clay impregnated with a small amount (e.g., 100 to 1500 ppm) of platinum. Reference is made to U.S. Pat. No. 4,171,286 (supra). Most commercially used promoters are obtained by impregnating a source of platinum on micro- spheres of high purity porous alumina, typically gamma alumina. The selection of platinum as the precious metal in various commercial products appears to reflect a preference for this metal that is consistent with prior art disclosures that platinum is the most effective group VIII metal for carbon moNOxide oxidation promotion in FCC regenerators. See, for example, FIG. 3 in U.S. Pat. No. 4,107,032 and the same figure in U.S. Pat. No. 4,350,614. The figure illustrates the effect of increasing the concentration of various species of precious metal promoters from 0.5 to 10 ppm on CO_2/CO ratio." [0034] "The carrier material may be acidic in nature. In many cases the basic material will cover the acidic sites of the carrier, resulting in a catalyst having the required basic

EXHIBIT L

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2009/0050529 ("*Rainer*")

'864 patent	U.S. Patent Application No. 2009/0050529 ("*Rainer*")
	character. Suitable carrier materials include the refractory oxides, in particular alumina, silica, silica-alumina, titania, zirconia, and mixtures thereof." [0045]-[0047] "Another preferred class of basic materials is the aluminum phosphates. Silica-magnesia is an example of a material having both basic and acidic sites. Suitable predominately basic materials having acidic sites include silica sol, metal doped silica sol, and nano-scale composites of silica with other refractory oxides." [0051] "The additive composition may contain minor amounts of other materials, which preferably do not adversely affect the CO oxidation or NOx control function in a significant way. Reference herein to NOx control typically refers to the reduction of NOx emissions from the FCC process, although other types of control are contemplated by this invention including, for example, the maintenance of the NOx emission levels in the FCC unit where increased emissions of NOx are expected. The additive composition may consist essentially of items (i)-(iv) mentioned above, item (iii) being optional. Where the composition of the invention is used as an additive particle for an FCC process (as opposed to being integrated into the FCC catalyst particles themselves), the additive composition may be combined with fillers (e.g. clay, silica-alumina, silica and/or alumina particles) and/or binders (e.g. silica sol, alumina sol, silica alumina sol, etc.) to form particles suitable for use in an FCC process, preferably by spray drying before the calcination of step." [0063] "When alumina supports are impregnated solely with hexachloroplatinic acid, it has been found that the resulting catalyst profile is typically of the 'eggshell' type, that being the platinum is located in a thin shell at the support surface."

EXHIBIT L

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2009/0050529 ("*Rainer*")

'864 patent	U.S. Patent Application No. 2009/0050529 ("*Rainer*")
	To the extent that this limitation is not disclosed, either explicitly or inherently, by *Rainer*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
1[c] one or more Group VIII noble metals distributed in the particle as an eggshell such that a higher concentration of the one or more Group VIII noble metals is present in the outer region of the microsphere sized porous particle as compared to the concentration of the one or more Group VIII noble metals in the centre of the microsphere sized porous particle.	*Rainer* discloses a combustion promoter comprising one or more Group VIII noble metals, the metals distributed in the particle as an eggshell such that a higher concentration of the one or more Group VIII noble metals is present in the outer region of the microsphere sized porous particle as compared to the concentration of the one or more Group VIII noble metals in the centre of the microsphere sized porous particle. *See, e.g.*, [Abstract] "Compositions suitable for use in FCC processes are provided that are capable of providing improved CO oxidation promotion activity along with NOx emission control. Generally, compositions are provided for promoting CO oxidation in FCC processes, wherein the compositions contain (i) a basic material support, (ii) at least one oxide of a lanthanide series element, (iii), optionally, at least one oxide of a transition metal selected from Groups Ib and IIb of the Periodic Table and (iv) at least one precious metal. The basic material support is preferably a hydrotalcite-like compound." [0008]-[0010] "Specific examples of treatments applied to regeneration operated in the complete combustion mode include the addition of a CO combustion promoter metal to the catalyst or to the regenerator. For example, U.S. Pat. No. 2,647, 860 proposed

EXHIBIT L

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2009/0050529 ("*Rainer*")

'864 patent	U.S. Patent Application No. 2009/0050529 ("*Rainer*")
	adding 0.1 to 1 weight percent chromic oxide to a cracking catalyst to promote combustion of CO. U.S. Pat. No. 3,808,121 taught using relatively large-sized particles containing CO combustion-promoting metal into a regenerator. The small-sized catalyst is cycled between the cracking reactor and the catalyst regenerator while the combustion- promoting particles remain in the regenerator. Also, U.S. Pat. Nos. 4,072,600 and 4,093,535 teach the use of Pt, Pd, Ir, Rh, Os, Ru, and Re in cracking catalysts in concentrations of 0.01 to 50 ppm, based on total catalyst inventory to promote CO combustion in a complete bum unit. The use of precious metals to catalyze oxidation of carbon moNOxide in the regenerators of FCC units has gained broad commercial acceptance. Some of the history of this development is set forth in U.S. Pat. No. 4,171,286 and U.S. Pat. No. 4,222,856. In the earlier stages of the development, the precious metal was deposited on the particles of cracking catalyst. Present practice is generally to supply a promoter in the form of solid fluidizable particles containing a precious metal, such particles being physically separate from the particles of cracking catalyst. The precious metal or compound thereof, is supported on particles of suitable carrier material and the promoter particles are usually introduced into the regenerator separately from the particles of cracking catalyst. The particles of promoter are not removed from the system as fines and are cocirculated with cracking catalyst particles during the cracking/stripping/regeneration cycles. Judgment of the CO combustion efficiency of a promoter is done by the ability to control the difference in temperature, delta T, between the (hotter) dilute phase, cyclones or flue gas line, and the dense phase. Most FCC units now use a Pt CO combustion promoter. While the use of combustion promoters such as platinum reduce CO emissions, such reduction in CO emissions is usually accompanied by an increase in nitro- gen oxides (NOx) in the regenerator flue gas. Promoter products used on a commercial basis in FCC units include calcined spray dried porous microspheres of kaolin clay impregnated with a small amount (e.g.,

EXHIBIT L

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2009/0050529 ("*Rainer*")

'864 patent	U.S. Patent Application No. 2009/0050529 ("*Rainer*")
	100 to 1500 ppm) of platinum. Reference is made to U.S. Pat. No. 4,171,286 (supra). Most commercially used promoters are obtained by impregnating a source of platinum on micro- spheres of high purity porous alumina, typically gamma alumina. The selection of platinum as the precious metal in various commercial products appears to reflect a preference for this metal that is consistent with prior art disclosures that platinum is the most effective group VIII metal for carbon moNOxide oxidation promotion in FCC regenerators. See, for example, FIG. 3 in U.S. Pat. No. 4,107,032 and the same figure in U.S. Pat. No. 4,350,614. The figure illustrates the effect of increasing the concentration of various species of precious metal promoters from 0.5 to 10 ppm on CO_2/CO ratio." [0014] "Unfortunately, the more active combustion promoters such as platinum and palladium also serve to promote the formation of nitrogen oxides in the regeneration zone. It has been reported that the use of prior art CO promoters can cause a dramatic increase (e.g. >300%) in NOx." [0026] "In one aspect, the invention provides compositions for promoting CO oxidation in FCC processes, the compositions containing (i) basic material support, (ii) at least one oxide of a lanthanide series element, (iii), optionally, at least one oxide of a transition metal selected from Groups Ib and IIb of the Periodic Table and (iv) at least one precious metal. The basic material support is preferably a hydrotalcite-like compound." [0028] "In still another aspect of this invention, an additive for CO oxidation is provided by incorporating into a single particle, a platinum group metal for CO oxidation promotion and metal components for NOx decomposition, and in which the

EXHIBIT L

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2009/0050529 ("*Rainer*")

'864 patent	U.S. Patent Application No. 2009/0050529 ("*Rainer*")
	platinum group metal and other metals are distributed in separate areas of the particle to achieve improvements in NOx reduction." [0050] "The amount of precious metal can be varied considerably depending on the level of CO combustion promotion desired. Typical levels of precious metal will be in the range of 50 to 1500 ppm of total additive and/or catalyst. As used herein the precious metals include those selected from a group consisting of Ag, Cu, Pt, Pd, Ir, Os, Ru, Rh, Re and their precursors such as salts and amine complexes and mixtures thereof." [0059] "In another aspect of the present invention, a particulate additive for incorporation into an FCC process is described as having a specified distribution of precious metal so as to be effective in promoting the oxidation of carbon monoxide while minimizing NOx production. The additive of this aspect of the present invention is based on the concept of distributing the precious metal CO oxidation promoter within the center of the particulate additive and distributing throughout the particle any metal or metal oxides which have been found useful in the decomposition of NOx. A graphic depiction of the additive of this invention is shown in FIG. 1. As depicted in FIG. 1, the porous, particulate additive 10 will include a precious metal designated 'Pt' such as previously described, which is primarily distributed in the center 12 of additive 10. What is theorized in accordance with this invention is that the precious metal such as Ag, Cu, Pt, Pd, Ir, Os, Ru, Rh, and/or Re distributed primarily in the central area 12 of the porous additive 10 will be able to promote the oxidation of CO to CO_2."

EXHIBIT L

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2009/0050529 ("*Rainer*")

'864 patent	U.S. Patent Application No. 2009/0050529 ("*Rainer*")
	[0060] "In general, however, the primary distribution of precious metal will be in the far interior of the porous particulate substrate whereas the other metals or metal oxides will be primarily distributed near the outer surface of the particle." [0061] "To achieve the desired distribution of metal catalytic components as in the additive of this invention as depicted in FIG. 1, any known method can be used by this invention. In one such method, the porous substrate can first be impregnated with the precious metal." [0062]-[0063] "It is also known to form an 'egg yolk' catalyst distribution in which a catalyst component, in this case, precious metal, is concentrated at the particle center. Such process involves impregnating the particle with a precious metal solution or complex, alone or with a co-impregnant. It is known that convective flow of the liquids through the particle can be affected by specific drying models that can effectively distribute a metal into the desired interior of a porous particle. Subsequent impregnation can distribute one or more additional metals upon the outer surfaces of the porous support. The effect of different drying processes on catalyst distribution is described in 'Predicting the Effect of Drying on Supported Coimpregnation Catalysts,' Lekhal et al., Ind. Eng. Chem. Res. 2001, 40, pgs. 3989-3999. This article is also incorporated in its entirety by reference herein. Internal distribution of a platinum group metal is also known by impregnating a porous support with a platinum group salt and an acid that competes with the platinum salt for adsorption within the porous support. For example, a high surface area porous support can be contacted with a liquid solution of the active component. The active component enters the porous support via capillary action and diffusion and absorbs on the available surface sites. When alumina supports are impregnated

EXHIBIT L

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2009/0050529 ("*Rainer*")

'864 patent	U.S. Patent Application No. 2009/0050529 ("*Rainer*")
	solely with hexachloroplatinic acid, it has been found that the resulting catalyst profile is typically of the 'eggshell' type, that being the platinum is located in a thin shell at the support surface. It has been known that the platinum distribution could be made more uniform or distributed in the center of a catalyst by co-impregnating the hexachloroplatinic acid with an acid such as HCl or citric acid wherein the platinum is now in a sub-surface band or within the core of the porous support. Other co-impregnants have been studied in order to change the distribution of the platinum component. Depending on the relative diffusion and adsorption strengths of the co-impregnants, the active component can be 'pushed' into the interior of the particle. A review of this type of process is described in 'Preparation of Pt/Gamma-Al2 O3 Pellets with Internal Step Distribution of Catalyst: Experiments and Theory,' Papageorgiou, et at., Journal of Catalysis 158, 1996, pgs 439-451, and 'Catalysis: An Integrated Approach, Second, revised and enlarged edition,' Netherlands Institute for Catalysis Research, 1999, Chapter 10, the entire contents of which are herein incorporated by reference." To the extent that this limitation is not disclosed, either explicitly or inherently, by *Rainer*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[3] The combustion promoter according to claim 1, comprising a plurality of microsphere sized porous particles comprising alumina.	*Rainer* discloses a combustion promoter comprising a plurality of microsphere sized porous particles comprising alumina. *See, e.g.*, The support provided above for Claim 1[b].

EXHIBIT L

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2009/0050529 ("*Rainer*")

'864 patent	**U.S. Patent Application No. 2009/0050529 ("*Rainer*")**
	To the extent that this limitation is not disclosed, either explicitly or inherently, by *Rainer*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[6] The combustion promoter according to claim 1, wherein the microsphere sized porous particles collectively have an average (D50) size of between 60 and 90 microns as measured by laser diffraction.	*Rainer* discloses a combustion promoter comprising microsphere sized porous particles, wherein the microsphere sized porous particles collectively have an average (D50) size of between 60 and 90 microns as measured by laser diffraction. *See, e.g.*, The support provided above for Claim 1[a]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Rainer*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[12] The combustion promoter according to claim 1, wherein the one or more Group VIII noble metals comprises one or more of platinum, palladium, iridium, ruthenium, and rhodium.	*Rainer* discloses a combustion promoter comprising one or more Group VIII noble metals, wherein the one or more Group VIII noble metals comprises one or more of platinum, palladium, iridium, ruthenium, and rhodium. *See, e.g.*, The support provided above for Claim 1[c].

EXHIBIT L

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent Application No. 2009/0050529 ("*Rainer*")

'864 patent	**U.S. Patent Application No. 2009/0050529 ("*Rainer*")**
	To the extent that this limitation is not disclosed, either explicitly or inherently, by *Rainer*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[15] The combustion promoter according to claim 12, wherein the one or more Group VIII noble metals comprises palladium.	*Rainer* discloses a combustion promoter comprising one or more Group VIII noble metals, wherein the one or more Group VIII noble metals comprises palladium. *See, e.g.*, The support provided above for Claim 1[c]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Rainer*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.

EXHIBIT M

EXHIBIT M

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 5,017,541 ("*Schmidt*")

U.S. Patent Publication No. 5,017,541 ("*Schmidt*"), filed on June 27, 1990 and published on May 21, 1991, qualifies as prior art under at least 35 U.S.C. §§ 102 and/or 103 for the asserted claims of U.S. Patent No. 11,224,864 ("the '864 patent"), as discussed in detail in the claim chart below.

The Court has not yet construed the claims and therefore the meaning of the terms in the claims has yet to be resolved by the Court. The support identified here for these limitations is responsive to Plaintiffs' infringement contentions, which Defendant disagrees with. As such, nothing in Defendant's claim charts should be construed as an admission regarding infringement, either literally or under the doctrine of equivalents, or as an admission regarding Defendant's understanding of the proper scope of the asserted claims. Defendant reserves the right to rely on additional citations or sources of evidence that also may be applicable, or that may become applicable in light of claim construction, changes in Plaintiffs' infringement contentions, and/or information obtained during discovery as the case progresses.

To the extent Plaintiffs allege that *Schmidt* does not disclose any particular limitation of the asserted claims of the '864 patent, either expressly or inherently, it would have been obvious to a person of ordinary skill in the art as of the priority date of the '864 patent to modify *Schmidt* and/or to combine the teachings of *Schmidt* with other prior art references, including but not limited to the present prior art references cited in the Cover Pleading and the relevant section(s) of claim charts for other prior art references for the '912 patent in a manner that would have rendered the asserted claims invalid as obvious.

Defendant reserves the right to amend or supplement this claim chart at a later date as more fully set forth in the Cover Pleading.

EXHIBIT M

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 5,017,541 ("*Schmidt*")

'864 patent	**U.S. Patent No. 5,017,541 ("*Schmidt*")**
1[a] A CO to CO_2 combustion promoter comprising microsphere sized porous particles, each microsphere sized porous particle having a diameter of less than 1mm, and independently comprising	*Schmidt* discloses a CO to CO_2 combustion promoter comprising microsphere sized porous particles, each microsphere sized porous particle having a diameter of less than 1mm, and independently comprising the limitations discussed in the following sections. *See, e.g.*, [Abstract] "An improved catalyst particle is disclosed for the conversion of hydrocarbons which comprises a refractory inorganic-oxide support, a Friedel-Crafts metal halide, and a surface-layer platinum-group metal component, wherein the concentration of platinum-group metal component on the surface layer of each catalyst particle is at least 1.5 times the concentration in the central core of the catalyst particle. An isomerization process also is disclosed which is particularly effective for the conversion of C4-C7 alkanes." [1:13-16] "This invention relates to an improved catalyst useful in the conversion of hydrocarbons, particularly for the isomerization of alkanes." [1:45-57] "Catalyst exhibiting a dual cracking and hydrogenation-dehydrogenation function are applied widely in the petroleum refining and petrochemical industries to the reforming and isomerization of hydrocarbons. Such catalysts generally have the cracking function imparted by an inorganic oxide, zeolite, or halogen, with a platinum-group component usually imparting the hydrogenation-dehydrogenation function. A catalyst useful in isomerization must be formulated to properly balance its cracking and hydrogenation-dehydrogenation functions to achieve the desired conversion over a prolonged period of time, while effectively utilizing the expensive platinum group metal component." [3:15-35] "It is an object of the present invention to provide a novel catalyst particle, useful particularly for the isomerization of isomerizable hydrocarbons. A corollary object of the invention is to

EXHIBIT M

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 5,017,541 ("*Schmidt*")

'864 patent	U.S. Patent No. 5,017,541 ("*Schmidt*")
	provide a process for isomerizing isomerizable hydrocarbons, particularly alkanes having from four to seven carbon atoms per molecule. This invention is based on the discovery that a catalyst particle having a Friedel-Crafts metal halide and a surface-layer platinum-group metal on an inorganic-oxide support demonstrates surprising results in increasing the octane number of C_5/C_6 naphtha streams. A broad embodiment of the present invention is a catalyst particle comprising an inorganic-oxide binder, a Friedel-Crafts metal halide, and a surface-layer platinum-group metal component having a surface-layer concentration at least 1.5 times that in the central core of each particle." [4:4-47] "To reiterate briefly, one embodiment of the present invention is a catalyst particle comprising an inorganic- oxide binder, a Friedel-Crafts metal halide, and a surface-layer platinum-group metal component having a surface-layer concentration at least 1.5 times that in the central core of each catalyst particle. Considering first the refractory inorganic-oxide support utilized in the present invention, it is preferred that the material be a porous, adsorptive, high-surface-area support having a surface area of about 25 to about 500 m^2/g. The porous carrier material should also be uniform in composition and relatively refractory to the conditions utilized in the hydrocarbon conversion process. By the term 'uniform in composition,' it is meant that the support be unlayered, has no concentration gradients of the species inherent to its composition, and is completely homogeneous in composition. Thus, if the support is a mixture of two or more refractory materials, the relative amounts of these materials will be constant and uniform throughout the entire support. It is intended to include within the scope of the present invention carrier materials which have traditionally been utilized in dual-function hydrocarbon conversion catalysts such as: (1) refractory inorganic-oxides such as alumina, titanium dioxide, zirconium dioxide, chromium oxide, zinc oxide, magnesia, thoria, boria, silica-alumina, silica-magnesia,

EXHIBIT M

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 5,017,541 ("*Schmidt*")

'864 patent	U.S. Patent No. 5,017,541 ("*Schmidt*")
	chromia-alumina, alumina-boria, silica-zirconia, etc.; (2) ceramics, porcelain, bauxite; (3) silica or silica gel, silicon carbide, clays and silicates including those synthetically prepared and naturally occurring, which may or may not be acid treated, for example attapulgus clay, diatomaceous earth, fuller's earth, kaolin, kieselguhr, etc.; (4) crystalline zeolitic aluminosilicates, such as X-zeolite, Y-zeolite, mordenite, or L-zeolite, either in the hydrogen form or in nonacidic form with one or more alkali metals occupying the cationic exchangeable sites; (5) non-zeolitic molecular sieves, such as aluminophosphates or silico-aluminophosphates; (6) spinels such as $MgAl_2O_4$, $FeAl_2O_4$, $ZnAl_2O_4$, $CaAl_2O_4$, and other like compounds having the formula $MO\text{-}Al_2O_3$ where M is a metal having a valence of 2; and (7) combinations of materials from one or more of these groups." [5:3-45] "The alumina powder may be formed into a suitable catalyst material according to any of the techniques known to those skilled in the catalyst-carrier-forming art. Spherical carrier particles may be formed, for example, from this Ziegler alumina by: (1) converting the alumina powder into an alumina sol by reaction with a suitable peptizing acid and water and thereafter dropping a mixture of the resulting sol and a gelling agent into an oil bath to form spherical particles of an alumina gel which are easily converted to a gamma-alumina carrier material by known methods; (2) forming an extrudate from the powder by established methods and thereafter rolling the extrudate particles on a spinning disk until spherical particles are formed which can then be dried and calcined to form the desired particles of spherical carrier material; and (3) wetting the powder with a suitable peptizing agent and thereafter rolling the particles of the powder into spherical masses of the desired size. This alumina powder can also be formed in any other desired shape or type of carrier material known to those skilled in the art such as rods, pills, pellets, tablets, granules, extrudates, and like forms by methods well known to the practitioners of the catalyst material forming art. The preferred type of carrier material for the present invention is a cylindrical extrudate generally having a diameter of about 0.75 to 3.3 mm, with about 1.6 mm being especially preferred. The length-to-diameter ratio is about 1:1 to 5:1, with about 2:1 being especially

EXHIBIT M

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 5,017,541 ("*Schmidt*")

'864 patent	**U.S. Patent No. 5,017,541 ("*Schmidt*")**
	preferred. The preferred extrudate particle form of the carrier material is preferably prepared by mixing the alumina powder with water and suitable peptizing agents such as nitric acid, acetic acid, aluminum nitrate, and the like material until an extrudable dough is formed. The amount of water added to form the dough is typically sufficient to give a Loss on Ignition (LOI) at 500 DEG C. of about 45 to 65 mass %, with a value of 55 mass % being especially preferred. On the other hand, the acid addition rate is generally sufficient to provide 2 to 7 mass % of the volatile-free alumina powder used in the mix, with a value of 3 to 4 mass % being especially preferred. The resulting dough is then extruded through a suitably sized die to form extrudate particles." To the extent that this limitation is not disclosed, either explicitly or inherently, by *Schmidt*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
1[b] silica, alumina, or mixtures thereof; and	*Schmidt* discloses a combustion promoter comprising silica, alumina, or mixtures thereof. *See, e.g.*, [3:29-37] "A broad embodiment of the present invention is a catalyst particle comprising an inorganic-oxide binder, a Friedel-Crafts metal halide, and a surface-layer platinum-group metal component having a surface-layer concentration at least 1.5 times that in the central core of each particle. Alumina is the preferred inorganic-oxide binder, and aluminum chloride is the preferred Friedel-Crafts metal halide. Platinum is the preferred surface-layer platinum-group component."

EXHIBIT M

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 5,017,541 ("*Schmidt*")

'864 patent	U.S. Patent No. 5,017,541 ("*Schmidt*")
	[4:10-5:26] "Considering first the refractory inorganic-oxide support utilized in the present invention, it is preferred that the material be a porous, adsorptive, high-surface-area support having a surface area of about 25 to about 500 m^2/g. The porous carrier material should also be uniform in composition and relatively refractory to the conditions utilized in the hydrocarbon conversion process. By the term 'uniform in composition,' it is meant that the support be unlayered, has no concentration gradients of the species inherent to its composition, and is completely homogeneous in composition. Thus, if the support is a mixture of two or more refractory materials, the relative amounts of these materials will be constant and uniform throughout the entire support. It is intended to include within the scope of the present invention carrier materials which have traditionally been utilized in dual-function hydrocarbon conversion catalysts such as: (1) refractory inorganic-oxides such as alumina, titanium dioxide, zirconium dioxide, chromium oxide, zinc oxide, magnesia, thoria, boria, silica-alumina, silica-magnesia, chromia-alumina, alumina-boria, silica-zirconia, etc.; (2) ceramics, porcelain, bauxite; (3) silica or silica gel, silicon carbide, clays and silicates including those synthetically prepared and naturally occurring, which may or may not be acid treated, for example attapulgus clay, diatomaceous earth, fuller's earth, kaolin, kieselguhr, etc.; (4) crystalline zeolitic aluminosilicates, such as X-zeolite, Y-zeolite, mordenite, or L-zeolite, either in the hydrogen form or in nonacidic form with one or more alkali metals occupying the cationic exchangeable sites; (5) non-zeolitic molecular sieves, such as aluminophosphates or silico-aluminophosphates; (6) spinels such as $MgAl_2$ O4, $FeAl_2O_4$, $ZnAl_2O_4$, $CaAl_2O_4$, and other like compounds having the formula MO-Al_2O_3 where M is a metal having a valence of 2; and (7) combinations of materials from one or more of these groups. The preferred refractory inorganic-oxide for use in the present invention is alumina. Suitable alumina materials are the crystalline aluminas known as the gamma-, eta-, and theta-alumina, with gamma-or eta-alumina giving best results. The preferred refractory inorganic-oxide will have an apparent bulk density of about 0.3 to about 1.01 g/cc and surface area characteristics such that the average pore diameter is about 20 to 300 angstroms, the pore volume is about 0.1 to about 1 cc/g, and the surface area is about 100 to about 500 m^2 /g.

EXHIBIT M

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 5,017,541 ("*Schmidt*")

'864 patent	U.S. Patent No. 5,017,541 ("*Schmidt*")
	A particularly preferred alumina is that which has been characterized in U.S. Pat. Nos. 3,852,190 and 4,012,313 as a by-product from a Ziegler higher alcohol synthesis reaction as described in Ziegler's U.S. Pat. No. 2,892,858. For purposes of simplification, such an alumina will be hereinafter referred to as a 'Ziegler alumina.' Ziegler alumina is presently available from the Vista Chemical Company under the trademark 'Catapal' or from Condea Chemie GMBH under the trademark 'Pural.' This material is an extremely high purity pseudoboehmite powder which, after calcination at a high temperature, has been shown to yield a high-purity gamma-alumina. The alumina powder may be formed into a suitable catalyst material according to any of the techniques known to those skilled in the catalyst-carrier-forming art. Spherical carrier particles may be formed, for example, from this Ziegler alumina by: (1) converting the alumina powder into an alumina sol by reaction with a suitable peptizing acid and water and thereafter dropping a mixture of the resulting sol and a gelling agent into an oil bath to form spherical particles of an alumina gel which are easily converted to a gamma-alumina carrier material by known methods; (2) forming an extrudate from the powder by established methods and thereafter rolling the extrudate particles on a spinning disk until spherical particles are formed which can then be dried and calcined to form the desired particles of spherical carrier material; and (3) wetting the powder with a suitable peptizing agent and thereafter rolling the particles of the powder into spherical masses of the desired size. This alumina powder can also be formed in any other desired shape or type of carrier material known to those skilled in the art such as rods, pills, pellets, tablets, granules, extrudates, and like forms by methods well known to the practitioners of the catalyst material forming art." To the extent that this limitation is not disclosed, either explicitly or inherently, by *Schmidt*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from

EXHIBIT M

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 5,017,541 ("*Schmidt*")

'864 patent	U.S. Patent No. 5,017,541 ("*Schmidt*")
	known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
1[c] one or more Group VIII noble metals distributed in the particle as an eggshell such that a higher concentration of the one or more Group VIII noble metals is present in the outer region of the microsphere sized porous particle as compared to the concentration of the one or more Group VIII noble metals in the centre of the microsphere sized porous particle.	*Schmidt* discloses a combustion promoter comprising one or more Group VIII noble metals, the metals distributed in the particle as an eggshell such that a higher concentration of the one or more Group VIII noble metals is present in the outer region of the microsphere sized porous particle as compared to the concentration of the one or more Group VIII noble metals in the centre of the microsphere sized porous particle. *See, e.g.*, [Abstract] "An improved catalyst particle is disclosed for the conversion of hydrocarbons which comprises a refractory inorganic-oxide support, a Friedel-Crafts metal halide, and a surface-layer platinum-group metal component, wherein the concentration of platinum-group metal component on the surface layer of each catalyst particle is at least 1.5 times the concentration in the central core of the catalyst particle. An isomerization process also is disclosed which is particularly effective for the conversion of C_4-C_7 alkanes." [1:45-57] "Catalyst exhibiting a dual cracking and hydrogenation-dehydrogenation function are applied widely in the petroleum refining and petrochemical industries to the reforming and isomerization of hydrocarbons. Such catalysts generally have the cracking function imparted by an inorganic oxide, zeolite, or halogen, with a platinum-group component usually imparting the hydrogenation-dehydrogenation function. A catalyst useful in isomerization must be formulated to properly balance its cracking and hydrogenation-dehydrogenation functions to achieve the desired conversion over a prolonged period of time, while effectively utilizing the expensive platinum group metal component." [3:22-38] "This invention is based on the discovery that a catalyst particle having a Friedel-Crafts metal halide and a surface-layer platinum-group metal on an inorganic-oxide support demonstrates surprising results in increasing the octane number of C_5 /C_6 naphtha streams.

EXHIBIT M

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 5,017,541 ("*Schmidt*")

'864 patent	U.S. Patent No. 5,017,541 ("*Schmidt*")
	A broad embodiment of the present invention is a catalyst particle comprising an inorganic-oxide binder, a Friedel-Crafts metal halide, and a surface-layer platinum-group metal component having a surface-layer concentration at least 1.5 times that in the central core of each particle. Alumina is the preferred inorganic-oxide binder, and aluminum chloride is the preferred Friedel-Crafts metal halide. Platinum is the preferred surface-layer platinum-group component." [4:4-9] "To reiterate briefly, one embodiment of the present invention is a catalyst particle comprising an inorganic- oxide binder, a Friedel-Crafts metal halide, and a surface-layer platinum-group metal component having a surface-layer concentration at least 1.5 times that in the central core of each catalyst particle." [3:42-46] "In another aspect, the invention is a preferred method of preparing the present catalyst by surface-impregnating a platinum-group metal component and vaporizing and subliming the Friedel-Crafts metal component onto the inorganic-oxide binder."

EXHIBIT M

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 5,017,541 ("*Schmidt*")

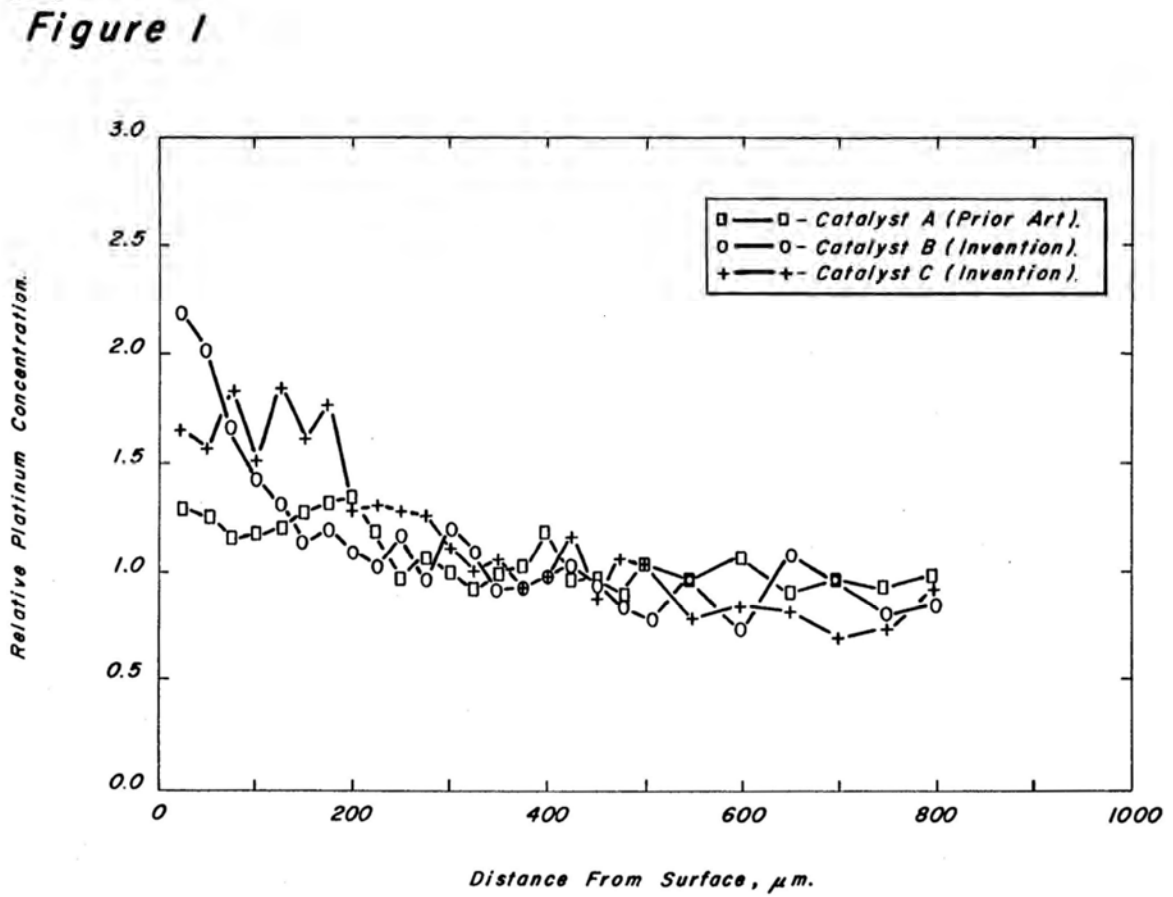
'864 patent	U.S. Patent No. 5,017,541 ("*Schmidt*")
	[Fig.1] 

EXHIBIT M

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 5,017,541 ("*Schmidt*")

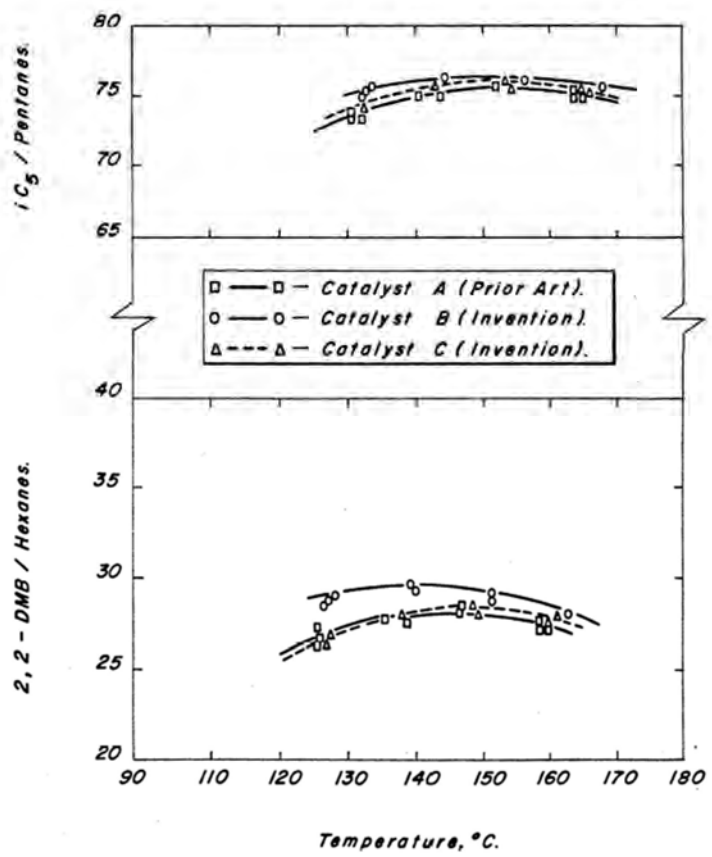
'864 patent	U.S. Patent No. 5,017,541 ("*Schmidt*")
	[Fig. 2]  [3:55-68] "FIG. 1 shows the platinum gradient across the radius of catalyst particles for the Examples described hereinafter. The platinum gradient of a control catalyst particle of the prior art is contrasted with the gradient of catalysts particles of the invention.

EXHIBIT M

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 5,017,541 ("*Schmidt*")

'864 patent	U.S. Patent No. 5,017,541 ("*Schmidt*")
	FIG. 2 shows comparative product isomer distribution from the isomerization of a light naphtha stream using catalyst particles containing surface-layer platinum contrasted with results using catalyst particles containing uniformly distributed platinum. Alternative levels of platinum on catalyst also are shown in order to show the impact on platinum requirements of concentrating the platinum in the surface-layer." [5:57-6:19] "An essential ingredient of the catalyst is the surface-layer platinum-group metal component. Of the platinum group, i.e., platinum, palladium, rhodium, ruthenium, osmium and iridium, palladium is a preferred component and platinum is especially preferred. Mixtures of platinum-group metals also are within the scope of this invention. This component may exist within the final catalytic composite as a compound such as an oxide, sulfide, halide, or oxyhalide, in chemical combination with one or more of the other ingredients of the composite, or as an elemental metal. Best results are obtained when substantially all of this component is present in the elemental state. This component may be present in the final catalyst composite in any amount which is catalytically effective, but relatively small amounts are preferred. In fact, the surface-layer platinum-group metal component generally will comprise about 0.01 to 2 mass % of the final catalyst, calculated on an elemental basis. Excellent results are obtained when the catalyst contains about 0.05 to l mass % of platinum. An essential feature of the catalyst of the present invention is that the platinum-group metal component is concentrated in the surface layer of each catalyst particle. In defining the present invention, a 'surface-layer' component has a concentration in the micron surface layer of the catalyst particle that is at least 1.5 times the concentration in the central core of the catalyst particle. Preferably, the surface-layer concentration of platinum- group metal is at least about twice the concentration in the central core."

EXHIBIT M

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 5,017,541 ("*Schmidt*")

'864 patent	U.S. Patent No. 5,017,541 ("*Schmidt*")
	[6:42-59] "The characterization of the platinum-group metal component as a 'surface-layer' component is intended to encompass a platinum-group metal component gradient upon and within the catalyst support. The concentration of platinum-group metal component tapers off in progressing from the surface to the center of the catalyst particle. The actual gradient of the platinum-group metal component within the catalyst particle varies depending upon the manufacturing method used to fabricate the catalyst. However, a substantially greater portion of the platinum-group metal component is located outside than within the central core of the catalyst particle. As previously stated, the concentration of the platinum-group metal component in the surface layer of the catalyst is at least 1.5 times and preferably twice the concentration in the central core of the catalyst particle. The gradient of the platinum-group metal component is determined by Scanning Electron Microscopy." [7:19-64] "The platinum-group metal component may be incorporated into the catalyst of the present invention by any means suitable to result in a surface-layer component having a concentration in the surface layer of each catalyst particle at least 1.5 times that of the central core of the particle. Best results are obtained when the platinum-group metal component is surface-impregnated. A preferred method of surface impregnating the platinum- group component is by means of a low-acid impregnation utilizing a solution of a soluble, decomposable complex compound of the platinum group component. In general the solvent used in this impregnation step is selected on the basis of its capability to dissolve the desired decomposable complex compound and is a low- acid, preferably aqueous solution. By low-acid it is meant that the impregnation solution generally has a normality of 2 or less. An HCI solution is preferred, but nitric acid and the like also can be used. Typical platinum-group compounds which may be employed in preparing the catalyst of the invention are chloroplantinic acid, ammonium chloroplatinate, bromoplatinic acid, platinum dichloride, platinum tetrachloride hydrate, platinum dichlorocarbonyl dichloride, dinitrodiaminoplatinum, palladium chloride, palladium chloride dihydrate, palladium nitrate,

EXHIBIT M

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 5,017,541 ("*Schmidt*")

'864 patent	U.S. Patent No. 5,017,541 ("*Schmidt*")
	etc. Chloroplatinic acid is preferred as a source of the especially preferred platinum component. A surface-layer platinum component may be impregnated onto the catalyst from a solution of chloroplatinic acid in the absence of strong mineral acids such as hydrochloric and nitric acid. The platinum group component may be surface im-pregnated via the formulation of a chemical complex of the platinum-group metal component which is strongly attracted to the refractory oxide support, resulting in the platinum group metal being retained primarily upon the outer surface of the catalyst. Any compound that is known to complex with the desired platinum-group component and with the metal component of the refractory inorganic-oxide support may be used in the preparation of the catalyst of the present invention. It has been found that a multi-dentated ligand is very useful in complexing with a platinum group metal and the refractory inorganic oxide support, resulting in the surface-impregnation of the platinum-group metal component." [8:3-26] "It is also an aspect of this invention that the multi-dentated ligand contains a functional group such as -SH or PR_2 (where R is hydrocarbon) that has a high affinity towards the platinum group metal component and a second functional group comprising a carboxylic acid or the like component that can be strongly adsorbed onto the metal oxide support. This preferred property of the multi-dentated ligand effectively insures that the platinum group metal component does not penetrate the catalyst particle, by binding strongly with the platinum group metal while also binding to the support quickly and strongly. Examples of some useful multi-dentated ligands include thiomalic acid, thiolactic acid, mercap-topropionic acid, thiodiacetic acid, thioglycollic acid, and thioproponic acid among others. It is within the scope of the present invention that the catalyst may contain other metal components known to modify the effect of the platinum-group metal component. Such metal modifiers may include rhenium, tin, germanium, lead, cobalt, nickel, indium, gallium, zinc,

EXHIBIT M

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 5,017,541 ("*Schmidt*")

'864 patent	**U.S. Patent No. 5,017,541 ("*Schmidt*")**
	uranium, dysprosium, thallium, and mixtures thereof. Catalytically effective amounts of such metal modifiers may be incorporated into the catalyst by any means known in the art." [14:58-15:2] "FIG. 1 shows the relative distribution of platinum level across the 800-micron radius of the catalyst particles from the surface to the center, relative to the aver- age concentration in the central core representing 50% of the volume of each particle. Catalyst A displayed a relatively even distribution of platinum, with the variation within a region of the catalyst comparable to the difference between the surface layer and central core. Catalysts B and C clearly displayed a relatively high concentration of platinum in the surface layer, with up to about 2.5 times the platinum concentration near the surface relative to the concentration near the center of the catalyst particle." [15:60-16:6] "The comparison of the relative performance of Catalysts A and B show the advantage of surface-layer platinum. Catalyst B shows an advantage of 1-2% for the highest-octane isomers with peak concentrations at lower temperatures where more favorable equilibrium is obtained. Even the surface-layer low-platinum Catalyst C shows an advantage over the high-platinum control Catalyst A, thus demonstrating that the present invention reduces requirements for costly platinum in an isomerization process." To the extent that this limitation is not disclosed, either explicitly or inherently, by *Schmidt*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[3] The combustion promoter according to claim 1, comprising a	*Schmidt* discloses a combustion promoter comprising a plurality of microsphere sized porous particles comprising alumina. *See, e.g.*,

EXHIBIT M

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 5,017,541 ("*Schmidt*")

'864 patent	**U.S. Patent No. 5,017,541 ("*Schmidt*")**
plurality of microsphere sized porous particles comprising alumina.	The support provided above for Claim 1[b]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Schmidt*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[6] The combustion promoter according to claim 1, wherein the microsphere sized porous particles collectively have an average (D50) size of between 60 and 90 microns as measured by laser diffraction.	*Schmidt* discloses a combustion promoter comprising microsphere sized porous particles, wherein the microsphere sized porous particles collectively have an average (D50) size of between 60 and 90 microns as measured by laser diffraction. *See, e.g.*, The support provided above for Claim 1[a]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Schmidt*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[12] The combustion promoter according to claim 1, wherein the one or more Group VIII noble metals comprises one or more of platinum, palladium, iridium, ruthenium, and rhodium.	*Schmidt* discloses a combustion promoter comprising one or more Group VIII noble metals, wherein the one or more Group VIII noble metals comprises one or more of platinum, palladium, iridium, ruthenium, and rhodium. *See, e.g.*, The support provided above for Claim 1[c].

EXHIBIT M

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 5,017,541 ("*Schmidt*")

'864 patent	**U.S. Patent No. 5,017,541 ("*Schmidt*")**
	To the extent that this limitation is not disclosed, either explicitly or inherently, by *Schmidt*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[15] The combustion promoter according to claim 12, wherein the one or more Group VIII noble metals comprises palladium.	*Schmidt* discloses a combustion promoter comprising one or more Group VIII noble metals, wherein the one or more Group VIII noble metals comprises palladium. *See, e.g.*, The support provided above for Claim 1[c]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Schmidt*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.

EXHIBIT N

EXHIBIT N

Invalidity of U.S. Patent No. 11,224,864 in View of *Shyr Article*

"*Preparation of Nonuniformly Active Catalysts*" ("*Shyr Article*"), authored by Yen-Shin Shyr et al. and published in 1980, qualifies as prior art of U.S. Patent No. 11,224,864 ("the '864 patent") under at least 35 U.S.C. §§ 102 and/or 103 for the asserted claims of the '864 patent, as discussed in detail in the claim chart below.

The Court has not yet construed the claims and therefore the meaning of the terms in the claims has yet to be resolved by the Court. The support identified here for these limitations is responsive to Plaintiffs' infringement contentions, which Defendant disagrees with. As such, nothing in Defendant's claim charts should be construed as an admission regarding infringement, either literally or under the doctrine of equivalents, or as an admission regarding Defendant's understanding of the proper scope of the asserted claims. Defendant reserves the right to rely on additional citations or sources of evidence that also may be applicable, or that may become applicable in light of claim construction, changes in Plaintiffs' infringement contentions, and/or information obtained during discovery as the case progresses.

To the extent Plaintiffs allege that *Shyr Article* does not disclose any particular limitation of the asserted claims of the '864 patent, either expressly or inherently, it would have been obvious to a person of ordinary skill in the art as of the priority date of the '864 patent to modify *Shyr Article* and/or to combine the teachings of *Shyr Article* with other prior art references, including but not limited to the present prior art references cited in the Cover Pleading and the relevant section(s) of claim charts for other prior art references for the '864 patent in a manner that would have rendered the asserted claims invalid as obvious.

Defendant reserves the right to amend or supplement this claim chart at a later date as more fully set forth in the Cover Pleading.

EXHIBIT N

Invalidity of U.S. Patent No. 11,224,864 in View of *Shyr Article*

'864 patent	**"*Preparation of Nonuniformly Active Catalysts*" ("*Shyr Article*")**
1[a] A CO to CO_2 combustion promoter comprising microsphere sized porous particles, each microsphere sized porous particle having a diameter of less than 1mm, and independently comprising	*Shyr Article* discloses a CO to CO_2 combustion promoter comprising microsphere sized porous particles, each microsphere sized porous particle having a diameter of less than 1mm, and independently comprising the limitations discussed in the following sections. *See, e.g.*, [Page 425] "The effect of adding chemical coingredients to H_2PtCl_6 impregnating solutions in order to modify Pt profiles in spherical γ-alumina beads was studied. Coingredients included the following acids and salts: HCI, HF, HNO_3, acetic acid, citric acid, tartaric, $AlCl_3$, NaCl, NaF, NaBr, $NaNO_3$, Na_3PO_4, Na benzoate, and Na citrate. Radial Pt profiles were examined by electron probe microanalysis of bead cross sections. Nine distinctly different types of profiles were produced. NaBr was the only coingredient which produced a monotonically increasing Pt profile from bead center to surface. A simple model which describes the role of NaBr during the impregnation process was discussed. A method of producing monotonically decreasing Pt profiles from bead center to surface which involves incorporating both NaBr and citric acid in H_2PtCl_6 impregnating solution was studied." [Page 425] "A recent paper presented the theoretical basis for a kinetic study to determine the effect of activity distribution in spherical catalyst particles on selectivity of the deuterium-hydride exchange reaction (7). The catalyst considered was alumina-supported platinum. (This reaction was shown previously to be a suitable probe for diffusional influences in uniformly active catalysts (8, 9)). This paper presents results of the catalyst preparation phase of this study. A later article will report results of characterization and kinetic studies." [Page 426] "A 0.0564 *M* stock solution of H_2PtCl_6 was prepared by dissolving Fisher Certified $H_2PtCl_6$was prepared by dissolving Fisher Certified H_2PtCl_6. $6H_2O$ in deionized and carbon-treated water. Solution concentrations were determined by a spectrophotometric method (29) on a Bausch and Lomb spectrophotometer. Concentrations were expressed as moles of Pt per liter. A 0.1 *M* stock solution of citric acid (CA) was prepared from Fisher

EXHIBIT N

Invalidity of U.S. Patent No. 11,224,864 in View of *Shyr Article*

'864 patent	***"Preparation of Nonuniformly Active Catalysts"* ("*Shyr Article*")**
	Certified CA. Solution concentrations were determined by titration. Stock solutions of various acids or salts (Table 1) were prepared to give concentrations of about 0.1 *M.* γ-Alumina beads of spherical geometry (Rhone Poulenc Inc., GOD-30) had the following properties: diameter, 0.36 cm; bulk density, 0.3 g/ cm^3 ; surface area, 150 m^2/ g; total pore volume, 1.70 cm^3/g; and average pore diameter, 450 A. Adsorption isotherm experiments involved contacting eight alumina beads (ca. 0.1 g) with 0.5 cm^3 of solution containing CA or H_2PtCl_6 at 25°C. All solutions were prepared by diluting the stock solutions described above. After 2 hr of contacting, beads were separated from solution by filtration and washed with 2 cm^3 of water to quantitatively remove and recover external solution. Washing time and volume were kept to a minimum to avoid removal of active ingredient from pore solution. The recovered solution was diluted with water to a volume of 5 cm^3 and analyzed for active ingredient. The amount of adsorption was determined by the difference between initial and final concentrations of active ingredient in solution after correcting for pore volume." [Page 427] "Figure 1 shows the total uptake of Pt after beads were contacted for 2 hr with impregnating solution containing H_2PtCl_6, then washed, dried, and reduced. The ordinate of this graph, 'total Pt uptake,' takes into account both adsorbed Pt and Pt deposited from pore solution during drying. The solid line shows the trend of the data. The dashed 45° line is a theoretical line for irreversible adsorption. The fact that data points at initial Pt values of about 0.006 mole and below fall near the theoretical line shows the strong interaction between Pt and γ-alumina. The other dashed line shows the trend of data at large values of initial Pt. The increase in the total Pt uptake in this linear region is due to Pt deposited from pore solution during drying. This increase can be used to determine the effective pore volume for the Pt in solution *(21, 31, 32).* The intersection of the two dashed lines represents the Pt uptake at saturation coverage of adsorption sites. The saturation coverage, 0.012 mmole Pt, divided by alumina sample weight and surface area corresponds to 110 μmole/g and 750 μmole/m^2, respectively."

EXHIBIT N

Invalidity of U.S. Patent No. 11,224,864 in View of *Shyr Article*

'864 patent	***"Preparation of Nonuniformly Active Catalysts"* ("*Shyr Article*")**
	[Page 428] "Adsorption isotherms (Fig. 3) were determined for both Pt and CA from data in Figs. 1 and 2 by subtracting the effective pore volume contribution from total uptake." [Page 428-429] "Table 1 presents a list of acids and salts which were studied in a set of coimpregnation experiments. The purpose was to screen a large number of chemicals as coingredients with H_2PtCl_6 to determine their effects upon the final Pt profile in the alumina beads. It was desirable to examine these effects after relatively short (1 hr) and long (22 hr) impregnation times, and to eliminate profile changes over this time period due to Pt diffusing into the pores from external impregnating solution. To avoid this undesirable effect, the total amount of Pt and total volume of solution were kept low-total Pt in solution was about 23% of the amount required to saturate the adsorption sites in the alumina (Fig. 1); solution volume was about 2.7 times the pore volume. Under these conditions, at short impregnation time, the solution would become essentially depleted of Pt as the Pt adsorbed on the support thus substantially reducing any driving force for diffusion. A control experiment in which no coingredient (Table 1, first entry) was added to the solution confirmed these expectations: after 1 hr of contact, the resulting catalyst contained a sharply defined shell of adsorbed Pt. After 22 hr of contact, the Pt shell showed only slight dispersion at the inner boundary." To the extent that this limitation is not disclosed, either explicitly or inherently, by *Shyr Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
1[b] silica, alumina, or mixtures thereof; and	*Shyr Article* discloses a combustion promoter comprising silica, alumina, or mixtures thereof. *See, e.g.*,

EXHIBIT N

Invalidity of U.S. Patent No. 11,224,864 in View of *Shyr Article*

'864 patent	**"*Preparation of Nonuniformly Active Catalysts*" ("*Shyr Article*")**
	[Page 425] "The effect of adding chemical coingredients to H_2PtCl_6 impregnating solutions in order to modify Pt profiles in spherical γ-alumina beads was studied. Coingredients included the following acids and salts: HCI, HF, HNO_3, acetic acid, citric acid, tartaric, $AlCl_3$,NaCl, NaF, NaBr, $NaNO_3$, Na_3PO_4, Na benzoate, and Na citrate. Radial Pt profiles were examined by electron probe microanalysis of bead cross sections. Nine distinctly different types of profiles were produced. NaBr was the only coingredient which produced a monotonically increasing Pt profile from bead center to surface. A simple model which describes the role of NaBr during the impregnation process was discussed. A method of producing monotonically decreasing Pt profiles from bead center to surface which involves incorporating both NaBr and citric acid in H_2PtCl6 impregnating solution was studied." [Page 425] "A recent paper presented the theoretical basis for a kinetic study to determine the effect of activity distribution in spherical catalyst particles on selectivity of the deuterium-hydride exchange reaction (7). The catalyst considered was alumina-supported platinum. (This reaction was shown previously to be a suitable probe for diffusional influences in uniformly active catalysts (8, 9)). This paper presents results of the catalyst preparation phase of this study. A later article will report results of characterization and kinetic studies." [Page 426] "An objective of this work was to investigate the possibility of using chemical additives (coingredients) in impregnating solutions as a means of controlling the distribution of active ingredient within the catalyst particle. Platinum on spherical γ- alumina beads was the system studied. Of particular interest for our future kinetic studies would be the ability to approximately reproduce some of the profiles that are considered in theoretical studies previously mentioned (3-6)—e.g., profiles which monotonically increase or decrease from particle center to surface. Linearly increasing or decreasing profiles were frequently considered in these theoretical studies."

EXHIBIT N

Invalidity of U.S. Patent No. 11,224,864 in View of *Shyr Article*

'864 patent	***"Preparation of Nonuniformly Active Catalysts"* ("*Shyr Article*")**
	[Page 426] "Hexachloroplatinic acid was the active ingredient selected for this study. The strong interaction between H_2PtCl_6 and γ- alumina is known to produce shell catalysts which have a well-defined boundary between Pt and bare support. Citric acid was employed as an additive to the H_2PtCl_6 solution because this combination is known to produce core catalysts. The approach that was used to find a means of controlling Pt profile was: first, to explore the interactions of H_2PtCl_6 and citric acid with the alumina supports; second, to experimentally survey a large number of chemical additives which, when included in the impregnating solutions, modify the shell or core profile; and third, to select and further investigate the chemical additive which yields the Pt profiles desired." [Page 426-427] "A 0.0564 *M* stock solution of H_2PtCl_6 was prepared by dissolving Fisher Certified H_2PtCl_6 was prepared by dissolving Fisher Certified H_2PtCl_6. $6H_2O$ in deionized and carbon-treated water. Solution concentrations were determined by a spectrophotometric method (29) on a Bausch and Lomb spectrophotometer. Concentrations were expressed as moles of Pt per liter. A 0.1 *M* stock solution of citric acid (CA) was prepared from Fisher Certified CA. Solution concentrations were determined by titration. Stock solutions of various acids or salts (Table 1) were prepared to give concentrations of about 0.1 *M*. γ-Alumina beads of spherical geometry (Rhone Poulenc Inc., GOD-30) had the following properties: diameter, 0.36 cm; bulk density, 0.3 g/ cm^3 ; surface area, 150 m^2/ g; total pore volume, 1.70 cm^3/g; and average pore diameter, 450 A. Adsorption isotherm experiments involved contacting eight alumina beads (ca. 0.1 g) with 0.5 cm^3 of solution containing CA or H_2PtCl_6 at 25°C. All solutions were prepared by diluting the stock solutions described above. After 2 hr of contacting, beads were separated from solution by filtration and washed with 2 cm^3 of water to quantitatively remove and recover external solution. Washing time and volume were kept to a minimum to avoid removal of active ingredient from pore solution. The recovered solution was diluted with

EXHIBIT N

Invalidity of U.S. Patent No. 11,224,864 in View of *Shyr Article*

'864 patent	***"Preparation of Nonuniformly Active Catalysts"* ("*Shyr Article*")**
	water to a volume of 5 cm^3 and analyzed for active ingredient. The amount of adsorption was determined by the difference between initial and final concentrations of active ingredient in solution after correcting for pore volume. In experiments designed to test a large number of acids and salts as coimpregnating ingredients, solutions were prepared by adding stock solutions of H_2PtCl_6, acid or salt solution, and water in the volumetric ratio 1: 1: 8. The experimental procedure involved placing eight alumina beads in contact with 0.5 cm^3 of impregnating solution for 1 or 22 hr at 25°C. The beads were removed from solution, washed with 2 cm^3 of water, dried in air at 120°C for 8 hr, and reduced in flowing H_2 at 500°C for 1 hr. Residual impregnating solution combined with wash water was analyzed for Pt. Characterization as to the type of Pt profile was determined by visual observation or by electron probe microanalysis (EPM) of beads that had been cleaved in half (*30*). EPM was performed on an Acton MS-64 microprobe using a point count technique. Beads that had been cleaved in half were scanned across the flat side through the bead center. Thirty-second counts were taken at 100-μm intervals across the surface. Scans at one to four orientations were taken for each bead. Data smoothing consisted of averaging the scan data taken at all orientations and reporting the averaged and normalized Pt profile from bead center *(r/R* = 0) to surface *(r/R* = 1). Visual observation was used when the profile was obviously a shell or core type with a sharp boundary between Pt and bare support. EPM was used when the profile was diffuse." [Page 427-428] "Absorption plays a large role in the impregnation of γ-alumina with H_2PtCl_6 solution. The bonding between Pt complex (henceforth referred to as Pt) and the surface is not well understood although mechanisms have been suggested. For example, recent studies have suggested that the adsorption of Pt is associated with either (1) variation of pH of the solution due to acid attack of the alumina with dissolution of surface aluminum ions (25), or (2) ligand displacement reaction of anionic Pt complexes with the alumina surface (27).

EXHIBIT N

Invalidity of U.S. Patent No. 11,224,864 in View of *Shyr Article*

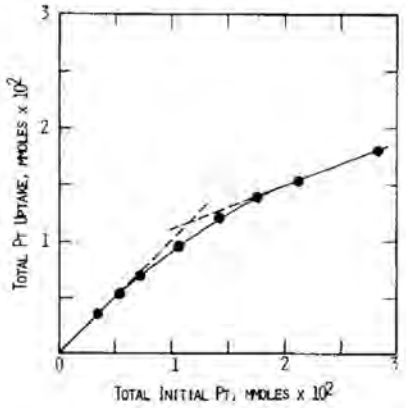
'864 patent	***"Preparation of Nonuniformly Active Catalysts"* ("*Shyr Article*")**
	Figure 1 shows the total uptake of Pt after beads were contacted for 2 hr with impregnating solution containing H_2PtCl_6, then washed, dried, and reduced. The ordinate of this graph, 'total Pt uptake,' takes into account both adsorbed Pt and Pt deposited from pore solution during drying. The solid line shows the trend of the data. The dashed 45° line is a theoretical line for irreversible adsorption. The fact that data points at initial Pt values of about 0.006 mole and below fall near the theoretical line shows the strong interaction between Pt and γ-alumina. The other dashed line shows the trend of data at large values of initial Pt. The increase in the total Pt uptake in this linear region is due to Pt deposited from pore solution during drying. This increase can be used to determine the effective pore volume for the Pt in solution *(21, 31, 32).* The intersection of the two dashed lines represents the Pt uptake at saturation coverage of adsorption sites. The saturation coverage, 0.012 mmole Pt, divided by alumina sample weight and surface area corresponds to 110 μmole/g and 750 μmole/m^2, respectively."  FIG. 1. Total uptake of Pt from impregnating solution by γ-alumina.

EXHIBIT N

Invalidity of U.S. Patent No. 11,224,864 in View of *Shyr Article*

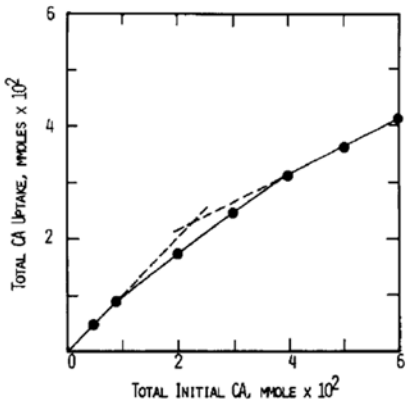
'864 patent	***"Preparation of Nonuniformly Active Catalysts"* ("*Shyr Article*")**
	[Page 428] "Figure 2 is a similar graph for the total uptake of CA. As in the case of Pt, data points at low initial CA fall near the dashed 45° line thus showing a strong interaction between CA and γ-alumina; at large initial CA the data trend is linear. Intersection of the two dashed lines, which corresponds to saturation coverage of adsorption sites, occurs at 0.023 mmole CA. Saturation coverage divided by sample weight and surface area equals 215 μmole/g and 1433 μmole/m2, respectively, or approximately twice the saturation coverage for Pt."  FIG. 2. Total uptake of CA from impregnating solution by γ-alumina.

EXHIBIT N

Invalidity of U.S. Patent No. 11,224,864 in View of *Shyr Article*

'864 patent	***"Preparation of Nonuniformly Active Catalysts"* (*"Shyr Article"*)**

[Table 1]

TABLE I

Properties of Coimpregnated Catalysts

Acid or salt	Impregnation time, 1 hr					Impregnation time, 22 hr		
	Conc (*M*)	Pt (wt%)	Pt band, *r*/*R*			Conc (*M*)	Pt (wt%)	Pt band
			Begins at	Ends at	Type			Type
None	—	0.51	1	0.77	1	—	0.51	2
$AlCl_3$	0.01	0.50	1	0.56	1	0.01	0.51	2
HCl	0.01	0.51	1	0.67	1	0.01	0.50	2
NaCl	0.01	0.49	1	0.81	1	0.01	0.51	2
HF	0.01	0.50	—	—	5	0.01	0.50	5
NaF	0.01	0.51	1	0.83	1	0.01	0.51	2
NaBr	0.01	0.50	1	0.77	1	0.01	0.50	9
HNO_3	0.01	0.50	1	0.50	1	0.01	0.51	3
$NaNO_3$	0.008	0.49	1	0.79	1	0.008	0.51	2
Na_3PO_4[a]	0.01	0.31	0.83	0	7	0.01	0.10	8
Na benzoate	0.01	0.48	1	0.78	1	0.01	0.50	2
Acetic acid	0.01	0.51	—	—	3	0.01	0.50	uniform
Citric acid	0.01	0.49	0.84	0.44	4	0.01	0.48	6
Na citrate[a]	0.02	0.19	0.33	0	7	0.01	0.14	8
Tartaric acid	0.01	0.49	—	—	6	0.01	0.51	7

[a] The 22-hr sample was prepared using a fresh solution of Na_3PO_4 (or Na citrate) and H_2PtCl_6 while the 1-hr sample was prepared using a portion of that same solution after it had been stored for 2 or 3 days.

[Page 428-429]

"Table 1 presents a list of acids and salts which were studied in a set of coimpregnation experiments. The purpose was to screen a large number of chemicals as coingredients with H_2PtCl_6 to determine their effects upon the final Pt profile in the alumina beads. It was desirable to examine these effects after relatively short (1 hr) and long (22 hr) impregnation times, and to eliminate profile changes over this time period due to Pt diffusing into the pores from external impregnating solution. To avoid this undesirable effect, the total amount of Pt

EXHIBIT N

Invalidity of U.S. Patent No. 11,224,864 in View of *Shyr Article*

'864 patent	***"Preparation of Nonuniformly Active Catalysts"* ("*Shyr Article*")**
	and total volume of solution were kept low-total Pt in solution was about 23% of the amount required to saturate the adsorption sites in the alumina (Fig. 1); solution volume was about 2.7 times the pore volume. Under these conditions, at short impregnation time, the solution would become essentially depleted of Pt as the Pt adsorbed on the support thus substantially reducing any driving force for diffusion. A control experiment in which no coingredient (Table 1, first entry) was added to the solution confirmed these expectations: after 1 hr of contact, the resulting catalyst contained a sharply defined shell of adsorbed Pt. After 22 hr of contact, the Pt shell showed only slight dispersion at the inner boundary." [Page 429] "For those catalysts which have sharply defined boundaries between regions with and without Pt, such as types 1, 4, and 7, the Pt bandwidth depended upon the coingredient, as shown in Table 1. With the exceptions of HF, acetic acid, and tartaric acid, all coingredients produced sharply defined Pt profiles after 1 hr of impregnation. Of these coingredients only CA and tartaric acid produced an inner Pt shell, and Na citrate, a Pt core. After 22 hr of impregnation, the Pt profiles produced with $AlCl_3$, HCl, NaCl, NaF, $NaNO_3$, and Na benzoate showed only slight dispersion at the inner boundaries. This finding illustrates the near irreversibility of the adsorption of Pt on alumina. With the exception of catalysts prepared with coingredients Na_3PO_4 and Na citrate, the weight percent of Pt in the FIG. 4. final catalyst represented greater than 95% total Pt uptake from solution." [Page 430] "Maatman (*11*) discussed the roles played by competitive adsorbates in distributing Pt throughout η-alumina supports."

EXHIBIT N

Invalidity of U.S. Patent No. 11,224,864 in View of *Shyr Article*

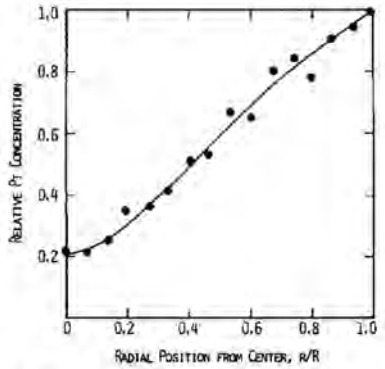
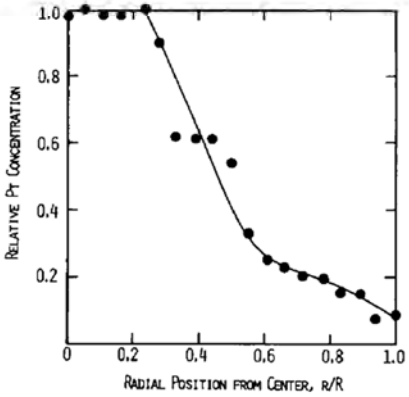
'864 patent	***"Preparation of Nonuniformly Active Catalysts"* ("*Shyr Article*")**
	[Fig. 5 & 6]  FIG. 5. Pt profile produced by coimpregnation of alumina beads with NaBr and H_2PtCl_6. Initial NaBr: Pt mole ratio, 1.77: 1; impregnation time, 24 hr; Pt content of beads, 0.50 wt%.  FIG. 6. Core Pt profile produced by coimpregnation of γ-alumina beads with CA and H_2PtCl_6 solution. Initial CA : Pt molar ratio, 5.31 : 1; impregnation time, 24 hr; Pt content of beads, 0.50 wt%.

EXHIBIT N

Invalidity of U.S. Patent No. 11,224,864 in View of *Shyr Article*

'864 patent	***"Preparation of Nonuniformly Active Catalysts"* ("*Shyr Article*")**
	[Page 430-431] "This simple model led to the idea of using three ingredients-NaBr, CA, and H_2PtCl_6 in impregnating solution as a means of producing monotonically decreasing Pt profiles. The reasoning used was as follows: It is known that CA interacts strongly with γ-alumina (Fig. 2) and successfully competes with Pt for adsorption sites to form core Pt profiles. If during the impregnation experiment, the core profile is first established and later modified by the relatively slow interaction between NaBr and adsorbed Pt, monotonically decreasing profiles might be achieved. Figure 7 shows that high CA concentrations can severely reduce the total uptake of Pt from aqueous solution by γ-alumina. It was therefore important in these catalyst preparation experiments to adjust the CA to Pt ratio sufficiently high to effect a core but low enough so that high Pt uptake was maintained."

EXHIBIT N

Invalidity of U.S. Patent No. 11,224,864 in View of *Shyr Article*

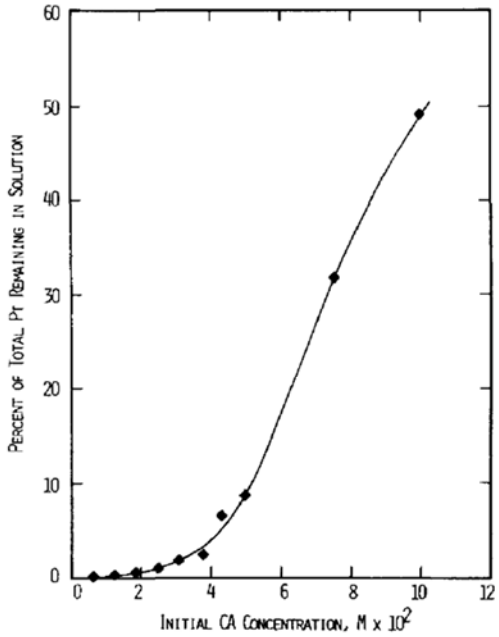
'864 patent	***"Preparation of Nonuniformly Active Catalysts"* (*"Shyr Article"*)**
	[Fig. 7 & 8 & 10]  FIG. 7. Effect of CA concentration on uptake of Pt from solution. Initial Pt concentration, 2.8×10^{-3} *M*; ratio of liquid volume to alumina weight, 7.5 ml/g.

EXHIBIT N

Invalidity of U.S. Patent No. 11,224,864 in View of *Shyr Article*

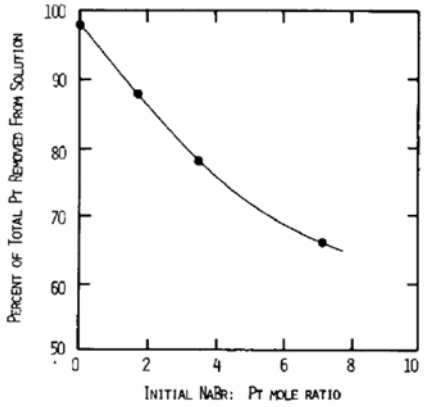
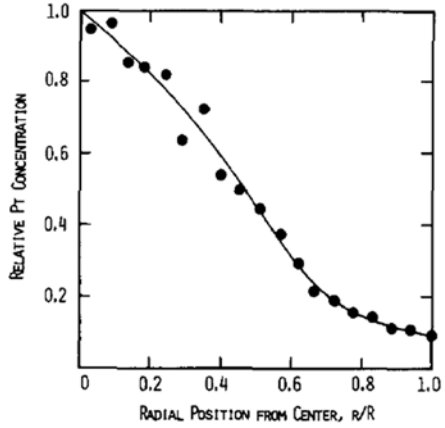
'864 patent	***"Preparation of Nonuniformly Active Catalysts"* (*"Shyr Article"*)**
	 FIG. 8. Effect of NaBr content on total Pt uptake by alumina beads. Initial CA : Pt mole ratio, 5.31 : 1, impregnation time, 24 hr.  FIG. 10. Pt profile produced by coimpregnation of γ-alumina beads with NaBr, CA, and H_2PtCl_6 solution. Initial NaBr : CA : Pt mole ratio, 1.77 : 5.31 : 1; impregnation time, 16 hr; Pt content of beads, 0.50%.

EXHIBIT N

Invalidity of U.S. Patent No. 11,224,864 in View of *Shyr Article*

'864 patent	**"*Preparation of Nonuniformly Active Catalysts*" ("*Shyr Article*")**
	[Page 432] "NaBr, when used as a coingredient in H_2PtCl_6 or CA-H_2PtCl_6 impregnating solutions, is effective in modifying the Pt distribution within γ-alumina supports. Pt profiles which result from the above solutions are respectively monotonically increasing and monotonically decreasing from bead center to surface. Without the NaBr coingredient the above solutions produce shell- and core-type Pt profiles respectively. The function of the NaBr is unclear although it appears to interact with the adsorbed Pt or surface and weaken the Pt-surface bond." To the extent that this limitation is not disclosed, either explicitly or inherently, by *Shyr Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
1[c] one or more Group VIII noble metals distributed in the particle as an eggshell such that a higher concentration of the one or more Group VIII noble metals is present in the outer region of the microsphere sized porous particle as compared to the concentration of the one or more Group VIII noble metals in the centre of the microsphere sized porous particle.	*Shyr Article* discloses a combustion promoter comprising one or more Group VIII noble metals, the metals distributed in the particle as an eggshell such that a higher concentration of the one or more Group VIII noble metals is present in the outer region of the microsphere sized porous particle as compared to the concentration of the one or more Group VIII noble metals in the centre of the microsphere sized porous particle. *See, e.g.*, [Page 425] "The effect of adding chemical coingredients to H_2PtCl_6 impregnating solutions in order to modify Pt profiles in spherical γ-alumina beads was studied. Coingredients included the following acids and salts: HCI, HF, HNO_3, acetic acid, citric acid, tartaric, $AlCl_3$, NaCl, NaF, NaBr, $NaNO_3$, Na_3PO_4, Na benzoate, and Na citrate. Radial Pt profiles were examined by electron probe microanalysis of bead cross sections. Nine distinctly different types of profiles were produced. NaBr was the only coingredient which produced a monotonically increasing

EXHIBIT N

Invalidity of U.S. Patent No. 11,224,864 in View of *Shyr Article*

'864 patent	**"*Preparation of Nonuniformly Active Catalysts*" ("*Shyr Article*")**
	Pt profile from bead center to surface. A simple model which describes the role of NaBr during the impregnation process was discussed. A method of producing monotonically decreasing Pt profiles from bead center to surface which involves incorporating both NaBr and citric acid in H_2PtCl6 impregnating solution was studied." [Page 425-426] "A recent paper presented the theoretical basis for a kinetic study to determine the effect of activity distribution in spherical catalyst particles on selectivity of the deuterium-hydride exchange reaction (7). The catalyst considered was alumina-supported platinum. (This reaction was shown previously to be a suitable probe for diffusional influences in uniformly active catalysts (8, 9)). This paper presents results of the catalyst preparation phase of this study. A later article will report results of characterization and kinetic studies. Impregnation was selected as the means of preparing the catalysts because of its simplicity and wide use. Previous workers have developed ways of producing uniform profiles of active ingredient (e.g., *(10, 11))* and sharply defined profiles such as outer shells, inner shells, and cores (e.g., *(12- 14)).* Workers have investigated many preparation variables and their influence on the distribution of active ingredient in a number of metal-support combinations *(11, 15-18).* There exists a broad body of knowledge of the physical and chemical processes of the impregnation process and surface chemistry of support materials (e.g., *(19-28)).* However, each metal-support combination is unique and hence introduces new challenges to the investigator. An objective of this work was to investigate the possibility of using chemical additives (coingredients) in impregnating solutions as a means of controlling the distribution of active ingredient within the catalyst particle. Platinum on spherical γ- alumina beads was the system studied. Of particular interest for our future kinetic studies would be the ability to approximately reproduce some of the profiles that are considered in theoretical studies previously mentioned (3-6)—e.g., profiles which monotonically increase or decrease from

'864 patent	***"Preparation of Nonuniformly Active Catalysts"* ("*Shyr Article*")**
	particle center to surface. Linearly increasing or decreasing profiles were frequently considered in these theoretical studies. Catalysts which have monotonically decreasing profiles are of some interest in kinetic studies because they serve as models of partially poisoned catalysts. There may also be commercial interest in such catalysts especially for reactions which are diffusion limited and in which the catalyst undergoes a shell progressive poisoning. They may exhibit activity or poisoning resistance that is superior to uniform or shell profiles. Hexachloroplatinic acid was the active ingredient selected for this study. The strong interaction between H_2PtCl_6 and γ- alumina is known to produce shell catalysts which have a well-defined boundary between Pt and bare support. Citric acid was employed as an additive to the H_2PtCl_6 solution because this combination is known to produce core catalysts. The approach that was used to find a means of controlling Pt profile was: first, to explore the interactions of H_2PtCl_6 and citric acid with the alumina supports; second, to experimentally survey a large number of chemical additives which, when included in the impregnating solutions, modify the shell or core profile; and third, to select and further investigate the chemical additive which yields the Pt profiles desired." [Page 426-427] "A 0.0564 *M* stock solution of H_2PtCl_6 was prepared by dissolving Fisher Certified H_2PtCl_6 was prepared by dissolving Fisher Certified $H_2PtCl_6 \cdot 6H_2O$ in deionized and carbon-treated water. Solution concentrations were determined by a spectrophotometric method (29) on a Bausch and Lomb spectrophotometer. Concentrations were expressed as moles of Pt per liter. A 0.1 *M* stock solution of citric acid (CA) was prepared from Fisher Certified CA. Solution concentrations were determined by titration. Stock solutions of various acids or salts (Table 1) were prepared to give concentrations of about 0.1 *M*. γ-Alumina beads of spherical geometry (Rhone Poulenc Inc., GOD-30) had the following properties: diameter, 0.36 cm; bulk density, 0.3 g/ cm^3 ; surface area, 150 m^2/ g; total pore volume, 1.70 cm^3/g; and average pore diameter, 450 A.

EXHIBIT N

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'864 patent	***"Preparation of Nonuniformly Active Catalysts"* ("*Shyr Article*")**
	Adsorption isotherm experiments involved contacting eight alumina beads (ca. 0.1 g) with 0.5 cm^3 of solution containing CA or H_2PtCl_6 at 25°C. All solutions were prepared by diluting the stock solutions described above. After 2 hr of contacting, beads were separated from solution by filtration and washed with 2 cm^3 of water to quantitatively remove and recover external solution. Washing time and volume were kept to a minimum to avoid removal of active ingredient from pore solution. The recovered solution was diluted with water to a volume of 5 cm^3 and analyzed for active ingredient. The amount of adsorption was determined by the difference between initial and final concentrations of active ingredient in solution after correcting for pore volume. In experiments designed to test a large number of acids and salts as coimpregnating ingredients, solutions were prepared by adding stock solutions of H_2PtCl_6, acid or salt solution, and water in the volumetric ratio 1: 1: 8. The experimental procedure involved placing eight alumina beads in contact with 0.5 cm^3 of impregnating solution for 1 or 22 hr at 25°C. The beads were removed from solution, washed with 2 cm^3 of water, dried in air at 120°C for 8 hr, and reduced in flowing H_2 at 500°C for 1 hr. Residual impregnating solution combined with wash water was analyzed for Pt. Characterization as to the type of Pt profile was determined by visual observation or by electron probe microanalysis (EPM) of beads that had been cleaved in half (*30*). EPM was performed on an Acton MS-64 microprobe using a point count technique. Beads that had been cleaved in half were scanned across the flat side through the bead center. Thirty-second counts were taken at 100-μm intervals across the surface. Scans at one to four orientations were taken for each bead. Data smoothing consisted of averaging the scan data taken at all orientations and reporting the averaged and normalized Pt profile from bead center *(r/R* = 0) to surface *(r/R* = 1). Visual observation was used when the profile was obviously a shell or core type with a sharp boundary between Pt and bare support. EPM was used when the profile was diffuse. More extensive coimpregnation studies using solutions prepared from various combinations of H_2PtCl_6, CA and NaBr stock solutions, and water of dilution were conducted in a manner

'864 patent	***"Preparation of Nonuniformly Active Catalysts"* ("*Shyr Article*")**
	similar to the above experiments; however, in the more extensive studies, the effects of impregnation time and NaBr and CA concentrations on final Pt profile were studied. EPM was used in determining Pt profiles for all samples." [Page 427-428] "Absorption plays a large role in the impregnation of γ-alumina with H_2PtCl_6 solution. The bonding between Pt complex (henceforth referred to as Pt) and the surface is not well understood although mechanisms have been suggested. For example, recent studies have suggested that the adsorption of Pt is associated with either (1) variation of pH of the solution due to acid attack of the alumina with dissolution of surface aluminum ions (25), or (2) ligand displacement reaction of anionic Pt complexes with the alumina surface (27). Figure 1 shows the total uptake of Pt after beads were contacted for 2 hr with impregnating solution containing H_2PtCl_6, then washed, dried, and reduced. The ordinate of this graph, 'total Pt uptake,' takes into account both adsorbed Pt and Pt deposited from pore solution during drying. The solid line shows the trend of the data. The dashed 45° line is a theoretical line for irreversible adsorption. The fact that data points at initial Pt values of about 0.006 mole and below fall near the theoretical line shows the strong interaction between Pt and γ-alumina. The other dashed line shows the trend of data at large values of initial Pt. The increase in the total Pt uptake in this linear region is due to Pt deposited from pore solution during drying. This increase can be used to determine the effective pore volume for the Pt in solution *(21, 31, 32).* The intersection of the two dashed lines represents the Pt uptake at saturation coverage of adsorption sites. The saturation coverage, 0.012 mmole Pt, divided by alumina sample weight and surface area corresponds to 110 μmole/g and 750 μmole/m^2, respectively."

EXHIBIT N

Invalidity of U.S. Patent No. 11,224,864 in View of *Shyr Article*

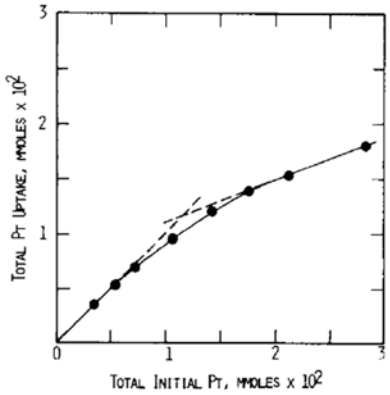
'864 patent	***"Preparation of Nonuniformly Active Catalysts"* ("*Shyr Article*")**
	 FIG. 1. Total uptake of Pt from impregnating solution by γ-alumina. [Page 428] "Figure 2 is a similar graph for the total uptake of CA. As in the case of Pt, data points at low initial CA fall near the dashed 45° line thus showing a strong interaction between CA and γ-alumina; at large initial CA the data trend is linear. Intersection of the two dashed lines, which corresponds to saturation coverage of adsorption sites, occurs at 0.023 mmole CA. Saturation coverage divided by sample weight and surface area equals 215 μmole/g and 1433 μmole/m^2, respectively, or approximately twice the saturation coverage for Pt. Adsorption isotherms (Fig. 3) were determined for both Pt and CA from data in Figs. 1 and 2 by subtracting the effective pore volume contribution from total uptake. The isotherms are expressed in terms of percent of adsorbed species. The line through the Pt data corresponds to the Langmuir equation: $$\text{wt\% Pt} = \frac{2880C}{1 + 1330C}$$ "

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Invalidity of U.S. Patent No. 11,224,864 in View of *Shyr Article*

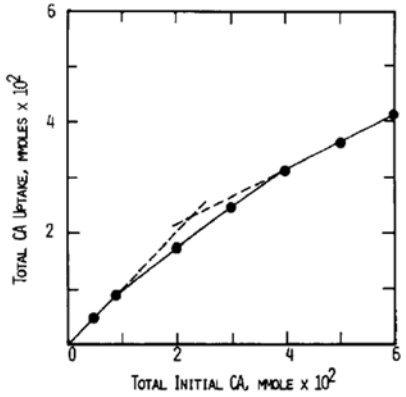
'864 patent	***"Preparation of Nonuniformly Active Catalysts"* ("*Shyr Article*")**
	 FIG. 2. Total uptake of CA from impregnating solution by γ-alumina.

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Invalidity of U.S. Patent No. 11,224,864 in View of *Shyr Article*

'864 patent	***"Preparation of Nonuniformly Active Catalysts"* (*"Shyr Article"*)**

[Table 1]

TABLE I

Properties of Coimpregnated Catalysts

Acid or salt	Impregnation time, 1 hr					Impregnation time, 22 hr		
	Conc (*M*)	Pt (wt%)	Pt band, *r*/*R*			Conc (*M*)	Pt (wt%)	Pt band
			Begins at	Ends at	Type			Type
None	—	0.51	1	0.77	1	—	0.51	2
$AlCl_3$	0.01	0.50	1	0.56	1	0.01	0.51	2
HCl	0.01	0.51	1	0.67	1	0.01	0.50	2
NaCl	0.01	0.49	1	0.81	1	0.01	0.51	2
HF	0.01	0.50	—	—	5	0.01	0.50	5
NaF	0.01	0.51	1	0.83	1	0.01	0.51	2
NaBr	0.01	0.50	1	0.77	1	0.01	0.50	9
HNO_3	0.01	0.50	1	0.50	1	0.01	0.51	3
$NaNO_3$	0.008	0.49	1	0.79	1	0.008	0.51	2
Na_3PO_4[a]	0.01	0.31	0.83	0	7	0.01	0.10	8
Na benzoate	0.01	0.48	1	0.78	1	0.01	0.50	2
Acetic acid	0.01	0.51	—	—	3	0.01	0.50	uniform
Citric acid	0.01	0.49	0.84	0.44	4	0.01	0.48	6
Na citrate[a]	0.02	0.19	0.33	0	7	0.01	0.14	8
Tartaric acid	0.01	0.49	—	—	6	0.01	0.51	7

[a] The 22-hr sample was prepared using a fresh solution of Na_3PO_4 (or Na citrate) and H_2PtCl_6 while the 1-hr sample was prepared using a portion of that same solution after it had been stored for 2 or 3 days.

[Page 428-430]

"Table 1 presents a list of acids and salts which were studied in a set of coimpregnation experiments. The purpose was to screen a large number of chemicals as coingredients with H_2PtCl_6 to determine their effects upon the final Pt profile in the alumina beads. It was desirable to examine these effects after relatively short (1 hr) and long (22 hr) impregnation times, and to eliminate profile changes over this time period due to Pt diffusing into the pores

'864 patent	***"Preparation of Nonuniformly Active Catalysts"* ("*Shyr Article*")**
	from external impregnating solution. To avoid this undesirable effect, the total amount of Pt and total volume of solution were kept low-total Pt in solution was about 23% of the amount required to saturate the adsorption sites in the alumina (Fig. 1); solution volume was about 2.7 times the pore volume. Under these conditions, at short impregnation time, the solution would become essentially depleted of Pt as the Pt adsorbed on the support thus substantially reducing any driving force for diffusion. A control experiment in which no coingredient (Table 1, first entry) was added to the solution confirmed these expectations: after 1 hr of contact, the resulting catalyst contained a sharply defined shell of adsorbed Pt. After 22 hr of contact, the Pt shell showed only slight dispersion at the inner boundary. In most experiments the coingredient concentration was 0.01 *M*, which corresponds to a coingredient to Pt mole ratio of 1.77. The columns in Table 1 labeled 'type' refer to types of profiles which were produced in these experiments after impregnated beads had been dried and reduced. Schematic representations which most closely describe these profiles are shown in Fig. 4. These representations can be described in qualitative terms as follows: Type 1-0uter shell, sharply defined Type 2-0uter shell, diffuse Type 3-0uter shell, diffuse to center Type 4-Inner shell, sharply defined Type 5-Inner shell, diffuse Type 6-lnner shell, diffuse to center Type 7-Core, sharply defined Type 8-Core, diffuse Type 9-Linearly increasing from center For those catalysts which have sharply defined boundaries between regions with and without Pt, such as types 1, 4, and 7, the Pt bandwidth depended upon the coingredient, as shown in Table 1. With the exceptions of HF, acetic acid, and tartaric acid, all coingredients produced sharply defined Pt profiles after 1 hr of impregnation. Of these coingredients only CA and

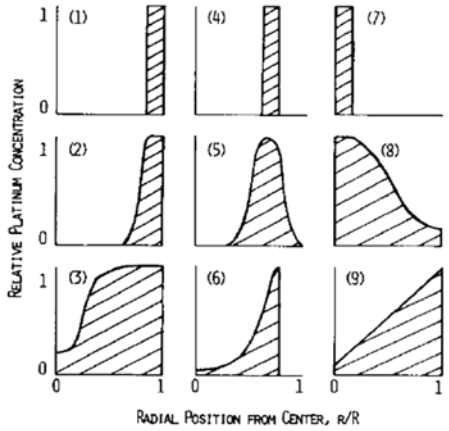
'864 patent	***"Preparation of Nonuniformly Active Catalysts"* ("*Shyr Article*")**
	tartaric acid produced an inner Pt shell, and Na citrate, a Pt core. After 22 hr of impregnation, the Pt profiles produced with $AlCl_3$, HCl, NaCl, NaF, $NaNO_3$, and Na benzoate showed only slight dispersion at the inner boundaries. This finding illustrates the near irreversibility of the adsorption of Pt on alumina. With the exception of catalysts prepared with coingredients Na_3PO_4 and Na citrate, the weight percent of Pt in the FIG. 4. final catalyst represented greater than 95% total Pt uptake from solution."  FIG. 4. Types of Pt profiles obtained in coimpregnation experiments. [Page 430] "Maatman (*11*) discussed the roles played by competitive adsorbates in distributing Pt throughout η-alumina supports. Michalko *(13, 14*) showed that dibasic acids, especially CA, could be used with H_2PtCl_6 to produce shell or core profiles. Becker and Nuttall (*12*) studied the influence of process variables on Pt profiles when CA was used with H_2PtCl_6. It is interesting to note that in Becker and Nuttall's work, impregnation time was an important variable-relatively short (e.g., *5* min) impregnation times produced sharp inner shell profiles, whereas 30-min impregnation time produced very diffuse almost uniform Pt profiles.

EXHIBIT N

Invalidity of U.S. Patent No. 11,224,864 in View of *Shyr Article*

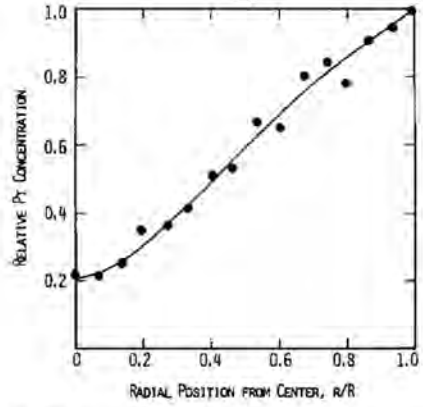
'864 patent	***"Preparation of Nonuniformly Active Catalysts" ("Shyr Article")***
	Maatman (*11*) also reported near uniform Pt profiles after several-hour impregnation times with various coingredients. In the present experiments, with most of the coingredients, impregnation time had less of an effect on Pt profiles than in previous works for reasons previously discussed. For catalysts produced with coingredients NaBr, HNO_3, Na_3PO_4, acetic acid, CA, Na citrate, and tartaric acid, increasing the impregnation time had a significant effect on Pt profile, although only acetic acid produced a uniform Pt profile at 22 hr. Figure 5 shows an example of a profile produced by coimpregnation with NaBr for 24 hr followed by drying and reduction. Figure 5 shows the typical data scatter that occurred in all EPM scans. An additional series of coimpregnation experiments were run with CA at a concentration of 0.03 *M* and a CA to Pt mole ratio of 5.31. This concentration was used in order to produce a core instead of an inner shell. The resulting core profiles showed slight dispersion at the boundary for low impregnation times. Pt dispersion increased substantially at higher impregnation times; however, even at times on the order of 24 hr, profiles maintained the broad flat center which typifies a core profile (see Fig. 6)."  FIG. 5. Pt profile produced by coimpregnation of alumina beads with NaBr and H_2PtCl_6. Initial NaBr: Pt mole ratio, 1.77:1; impregnation time, 24 hr; Pt content of beads, 0.50 wt%.

EXHIBIT N

Invalidity of U.S. Patent No. 11,224,864 in View of *Shyr Article*

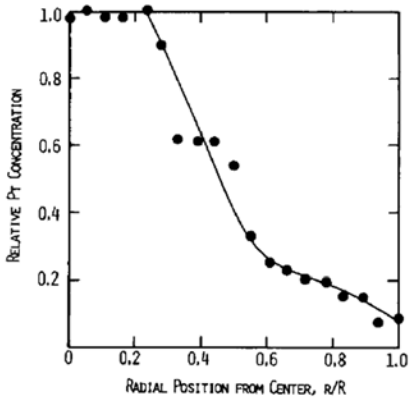
'864 patent	***"Preparation of Nonuniformly Active Catalysts"* ("*Shyr Article*")**
	 FIG. 6. Core Pt profile produced by coimpregnation of γ-alumina beads with CA and H_2PtCl_6 solution. Initial CA : Pt molar ratio, 5.31 : 1; impregnation time, 24 hr; Pt content of beads, 0.50 wt%. [Page 430-432] "NaBr was the only coingredient which produced a monotonically increasing Pt profile from bead center to surface (Fig. 5). It was therefore subjected to further investigation. Table 1 shows that NaBr had no effect on the 1-hr profile compared to the control, but had a great effect on the 22-hr profile. It appears that during the coimpregnation process, Pt adsorbed rapidly to form an outer shell catalyst; then in a comparatively slow process the NaBr interacted with the Pt or surface. This interaction increased the mobility of the Pt and allowed it to diffuse toward the center. This simple model led to the idea of using three ingredients-NaBr, CA, and H_2PtCl_6 in impregnating solution as a means of producing monotonically decreasing Pt profiles. The reasoning used was as follows: It is known that CA interacts strongly with γ-alumina (Fig. 2) and successfully competes with Pt for adsorption sites to form core Pt profiles. If during the impregnation experiment, the core profile is first established and later modified by the

EXHIBIT N

Invalidity of U.S. Patent No. 11,224,864 in View of *Shyr Article*

'864 patent	***"Preparation of Nonuniformly Active Catalysts"* ("*Shyr Article*")**
	relatively slow interaction between NaBr and adsorbed Pt, monotonically decreasing profiles might be achieved. Figure 7 shows that high CA concentrations can severely reduce the total uptake of Pt from aqueous solution by γ-alumina. It was therefore important in these catalyst preparation experiments to adjust the CA to Pt ratio sufficiently high to effect a core but low enough so that high Pt uptake was maintained. Figure 8 shows that high NaBr concentrations can also reduce total uptake of Pt from impregnating solutions. It was therefore important in these catalyst preparation experiments to adjust the NaBr to Pt ratio high enough so that it caused the Pt profile to move but low enough so that high Pt uptake was maintained. Figure 9 shows the relative Pt concentration profiles for coimpregnation experiments as a function of impregnation time. For these particular experiments, the solution contained NaBr, CA, and Pt in the molar ratio 1.77: 5.31: 1. (The lines represent smooth curves drawn through the EPM data to show the data trend.) Figure 10 shows the EPM data for the 16-hr sample. The interesting effect of the NaBr is that it produces a monotonically decreasing profile which has a sharp maximum at the bead center as opposed to a broad flat center as in Fig. 6."

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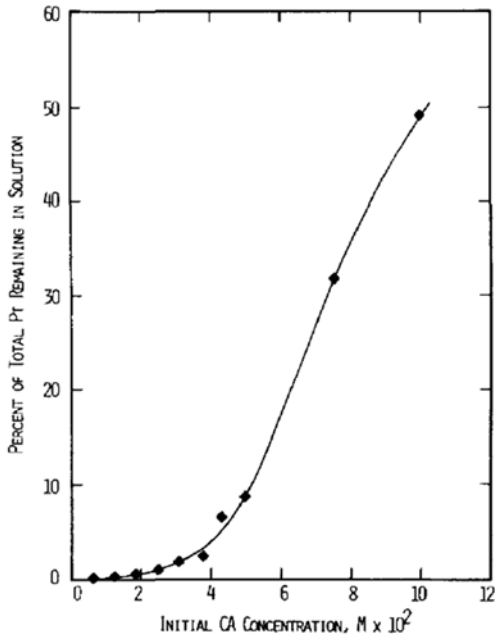
'864 patent	***"Preparation of Nonuniformly Active Catalysts"* ("*Shyr Article*")**
	 FIG. 7. Effect of CA concentration on uptake of Pt from solution. Initial Pt concentration, 2.8×10^{-3} *M*; ratio of liquid volume to alumina weight, 7.5 ml/g.

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Invalidity of U.S. Patent No. 11,224,864 in View of *Shyr Article*

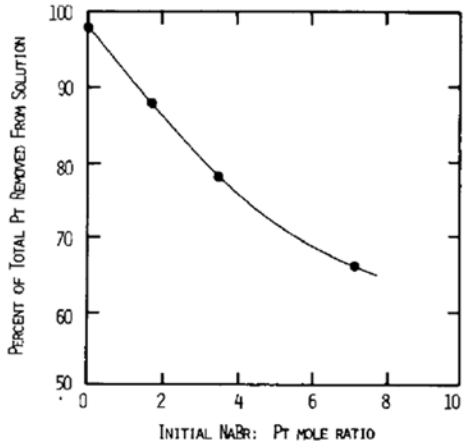
'864 patent	***"Preparation of Nonuniformly Active Catalysts"* ("*Shyr Article*")**
	 FIG. 8. Effect of NaBr content on total Pt uptake by alumina beads. Initial CA : Pt mole ratio, 5.31 : 1, impregnation time, 24 hr.

EXHIBIT N

Invalidity of U.S. Patent No. 11,224,864 in View of *Shyr Article*

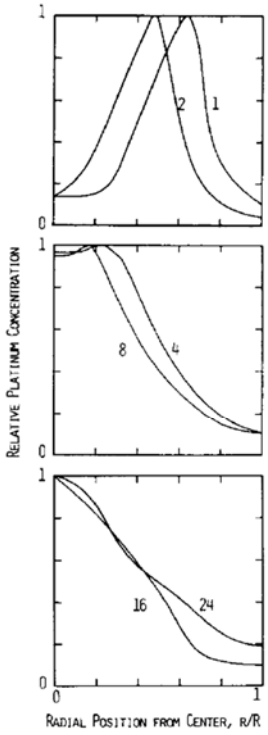
'864 patent	**"*Preparation of Nonuniformly Active Catalysts*" ("*Shyr Article*")**
	 FIG. 9. Effect of impregnation on Pt profile for NaBr : CA : Pt mole ratio 1.77 : 5.31 : 1. Impregnation times (hr) are shown on the figure.

EXHIBIT N

Invalidity of U.S. Patent No. 11,224,864 in View of *Shyr Article*

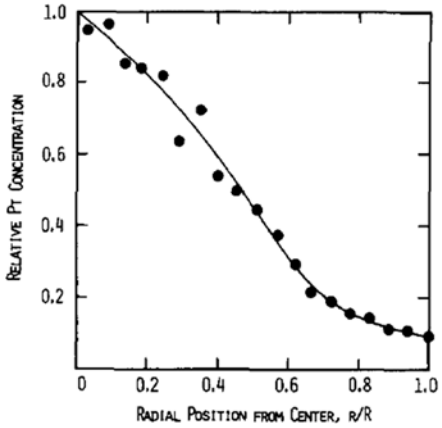
'864 patent	***"Preparation of Nonuniformly Active Catalysts"* ("*Shyr Article*")**
	 FIG. 10. Pt profile produced by coimpregnation of γ-alumina beads with NaBr, CA, and H_2PtCl_6 solution. Initial NaBr : CA : Pt mole ratio, 1.77 : 5.31 : 1; impregnation time, 16 hr; Pt content of beads, 0.50%. [Page 432] "NaBr, when used as a coingredient in H_2PtCl_6 or CA-H_2PtCl_6 impregnating solutions, is effective in modifying the Pt distribution within γ-alumina supports. Pt profiles which result from the above solutions are respectively monotonically increasing and monotonically decreasing from bead center to surface. Without the NaBr coingredient the above solutions produce shell- and core-type Pt profiles respectively. The function of the NaBr is unclear although it appears to interact with the adsorbed Pt or surface and weaken the Pt-surface bond." To the extent that this limitation is not disclosed, either explicitly or inherently, by *Shyr Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill

EXHIBIT N

Invalidity of U.S. Patent No. 11,224,864 in View of *Shyr Article*

'864 patent	***"Preparation of Nonuniformly Active Catalysts"* ("*Shyr Article*")**
	themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[3] The combustion promoter according to claim 1, comprising a plurality of microsphere sized porous particles comprising alumina.	*Shyr Article* discloses a combustion promoter comprising a plurality of microsphere sized porous particles comprising alumina. *See, e.g.*, The support provided above for Claim 1[b]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Shyr Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[6] The combustion promoter according to claim 1, wherein the microsphere sized porous particles collectively have an average (D50) size of between 60 and 90 microns as measured by laser diffraction.	*Shyr Article* discloses a combustion promoter comprising microsphere sized porous particles, wherein the microsphere sized porous particles collectively have an average (D50) size of between 60 and 90 microns as measured by laser diffraction. *See, e.g.*, The support provided above for Claim 1[a]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Shyr Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[12] The combustion promoter according to claim 1, wherein the one or more Group VIII noble	*Shyr Article* discloses a combustion promoter comprising one or more Group VIII noble metals, wherein the one or more Group VIII noble metals comprises one or more of platinum, palladium, iridium, ruthenium, and rhodium. *See, e.g.*,

EXHIBIT N

Invalidity of U.S. Patent No. 11,224,864 in View of *Shyr Article*

'864 patent	***"Preparation of Nonuniformly Active Catalysts"* ("*Shyr Article*")**
metals comprises one or more of platinum, palladium, iridium, ruthenium, and rhodium.	The support provided above for Claim 1[c]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Shyr Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[15] The combustion promoter according to claim 12, wherein the one or more Group VIII noble metals comprises palladium.	*Shyr Article* discloses a combustion promoter comprising one or more Group VIII noble metals, wherein the one or more Group VIII noble metals comprises palladium. *See, e.g.*, The support provided above for Claim 1[c]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Shyr Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.

EXHIBIT O

EXHIBIT O

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,358,533 ("*Tran*")

U.S. Patent No. 9,358,533 ("*Tran*"), filed on December 20, 2012 and published on June 7, 2016, qualifies as prior art under at least 35 U.S.C. §§ 102 and/or 103 for the asserted claims of U.S. Patent No. 11,224,864 ("the '864 patent"), as discussed in detail in the claim chart below.

The Court has not yet construed the claims and therefore the meaning of the terms in the claims has yet to be resolved by the Court. The support identified here for these limitations is responsive to Plaintiffs' infringement contentions, which Defendant disagrees with. As such, nothing in Defendant's claim charts should be construed as an admission regarding infringement, either literally or under the doctrine of equivalents, or as an admission regarding Defendant's understanding of the proper scope of the asserted claims. Defendant reserves the right to rely on additional citations or sources of evidence that also may be applicable, or that may become applicable in light of claim construction, changes in Plaintiffs' infringement contentions, and/or information obtained during discovery as the case progresses.

To the extent Plaintiffs allege that *Tran* does not disclose any particular limitation of the asserted claims of the '864 patent, either expressly or inherently, it would have been obvious to a person of ordinary skill in the art as of the priority date of the '864 patent to modify *Tran* and/or to combine the teachings of *Tran* with other prior art references, including but not limited to the present prior art references cited in the Cover Pleading and the relevant section(s) of claim charts for other prior art references for the '864 patent in a manner that would have rendered the asserted claims invalid as obvious.

Defendant reserves the right to amend or supplement this claim chart at a later date as more fully set forth in the Cover Pleading.

EXHIBIT O

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,358,533 ("*Tran*")

'864 patent	**U.S. Patent No. 9,358,533 ("*Tran*")**
1[a] A CO to CO_2 combustion promoter comprising microsphere sized porous particles, each microsphere sized porous particle having a diameter of less than 1mm, and independently comprising	*Tran* discloses a CO to CO_2 combustion promoter comprising microsphere sized porous particles, each microsphere sized porous particle having a diameter of less than 1mm, and independently comprising the limitations discussed in the following sections. *See, e.g.*, [Abstract] "Hollow porous metal oxide microspheres are provided. The microspheres may be used as a support for a catalyst, particularly an exhaust treatment catalyst for an internal combustion engine. Also provided are methods of making the microspheres, methods of using the microspheres as catalyst supports, and methods of exhaust treatment using catalyst articles comprising the microspheres." [1:13-29] "The exhaust gases of internal combustion engines contain pollutants such as hydrocarbons, carbon monoxide and nitrogen oxides (NO_x) that foul the air. Emission standards for unburned hydrocarbons, carbon monoxide and nitrogen oxide contaminants have been set by various governments and must be met by older as well as new vehicles. In order to meet such standards, catalytic converters containing a three way catalyst (TWC) may be located in the exhaust gas line of internal combustion engines. The use of exhaust gas catalysts have contributed to a significant improvement in air quality. The TWC is the most commonly used catalyst and it provides the three functions of oxidation of CO, oxidation of unburned hydrocarbons (HC's) and reduction of NOx to N_2. TWCs typically utilize one or more platinum group metals (PGM) to simultaneously oxidize CO and HC and reduce NOx compounds. The most common catalytic components of a TWC are platinum (Pt), rhodium (Rh) and palladium (Pd)." [1:42-52] "Refractory metal oxides such as alumina, rare-earth metal oxides, zirconia, titania, and their combinations, and other materials are commonly used as supports for the

EXHIBIT O

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,358,533 ("*Tran*")

'864 patent	U.S. Patent No. 9,358,533 ("*Tran*")
	catalytic components of a catalyst article and as oxygen storage materials (OSC). Currently, almost all of the alumina catalyst supports and OSC are in the form of solid powder particles with a particle size ranging from about 5-100 microns or are large extrudates above 100 microns in size. The alumina support materials typically exhibit a BET surface area in excess of 60 square meters per gram ('m^2/g'), often up to about 200 m^2/g or higher." [1:53-2:16] "In an internal combustion engine it is also desirable for these catalyst support materials to have high meso- and macro-porosity to enhance gas phase diffusion, which makes the catalysts more effective to achieve high nitrogen oxide (NOx) and hydrocarbon (HC) conversion at high space-velocity. In this regard, porous microspheres, including hollow microspheres, have been used as catalyst supports for the purpose of improving the porosity of the catalytic washcoat. Various preparation methods for such microspheres are reported in the literature. However, in general, hollow alumina microspheres that are formed at lower temperatures are thin-walled egg-shell structures that are too fragile to resist mechanical milling during catalyst preparation and hydrothermal aging in the engine. Thick-walled hollow alumina spheres are more robust against mechanical and thermal aging, and are available commercially (e.g., as insulation material), but these materials either have a large particle size or have been sintered at too high a temperature for catalyst applications. Hollow alumina microspheres made using ion-extraction of boehmite sols followed by firing at 1200° C. have been shown to have thick walls; however, these micro-spheres are in the dense alpha crystalline phase. There remains a need for hollow porous microspheres suitable for use as catalyst supports which can be made by simple manufacturing methods, and which have thick walls with small spherical diameters. The availability of such microspheres also leads to a significant reduction in raw material usage (e.g., precious metal,

EXHIBIT O

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,358,533 ("*Tran*")

'864 patent	U.S. Patent No. 9,358,533 ("*Tran*")
	alumina, and OSC) and therefore a substantial reduction in cost because of elimination of the dead space in the center of the conventional solid particle." [2:19-29] "In one aspect, the invention relates to a composition comprising hollow porous metal oxide microspheres. In a particular embodiment, the microspheres comprise walls having a thickness of about 1-5 μm, typically about 1-3 μm or about 2 μm, surrounding a hollow center and the diameter range of the hollow metal oxide microspheres is about 5-15 μm, typically 8-12 μm, or about 9-11 μm. Desirably, the average diameter (D_{90}) is about 10 μm. The microspheres may further comprise a catalyst, such as a catalyst for treatment of engine exhaust gases." [2:53-59] "In a further aspect, the invention relates to methods for treating engine exhaust comprising hydrocarbons, carbon monoxide and/or nitrogen oxides, the methods comprising contacting the exhaust with the hollow porous metal oxide microspheres or catalyst articles described above under conditions suitable for oxidation of carbon monoxide, oxidation of hydrocarbons and/or reduction of nitrogen oxides." [4:7-13] "As used herein, the terms 'nanoparticle' or 'nano-sized particle' refer to a particle having a diameter in the nanometer range (about 1-999 nm). The term 'microparticle' refers to a particle having a diameter in the micrometer or micron range (about 1-999 μm). Similarly, the term 'microsphere' refers to a generally spherical agglomeration of smaller particles which has a diameter in the micrometer or micron range." [4:38-50] "In certain embodiments, the average diameter of the metal oxide nanoparticles is about 50-500 nm. In specific embodiments, the average diameter of the metal oxide

EXHIBIT O

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,358,533 ("*Tran*")

'864 patent	U.S. Patent No. 9,358,533 ("*Tran*")
	nanoparticles is about 100 nm. If the particles of the starting material are larger, they may be milled to the desired size prior to forming the catalyst support. After spray-drying, the average diameter of the hollow porous microspheres, measured in D_{90}, is about 5-20 μm, typically about 8-12 μm, or about 9-11 μm. Desirably, the D_{90} diameter range is about 10 μm. The wall thickness of the microspheres is about 1-5 μm, typically about 1-3 μm, and more typically about 2 μm. If necessary to achieve the desired diameter range, the hollow porous microspheres may be milled to reduce the average diameter range." [8:46-63] "In general, the methods of making the hollow porous microspheres of the invention typically result in microspheres having an average diameter of approximately 10 μm. The D_{90} range of diameter is generally about 5-20 μm. In specific embodiments, the average diameter of the microspheres ranges from 8-12 or 9-11 μm measured in D_{90}. In a further aspect, the invention provides methods for treating exhaust gas from an internal combustion engine comprising carbon monoxide, hydrocarbons and/or nitrogen oxides, the method comprising contacting the gas in the exhaust stream of the engine with hollow porous microspheres supporting a catalyst, as described above, under conditions suitable for CO oxidation, HC oxidation and/or NOx reduction. The hollow porous microsphere catalyst may be formed as a layer on a support for use as a catalyst article as described above. In particular, CO and NOx in the exhaust stream are substantially reduced by contact with the catalysts and catalyst articles of the invention." [9:51-65] "Average particle size was measured on a Horiba LA-950 particle size analyzer. For a typical measurement, 0.1 g of sorbent was slurried in water. The particle size (diameter) is expressed as values less than 50% and 90% of total particles, D_{50} and

EXHIBIT O

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,358,533 ("*Tran*")

'864 patent	**U.S. Patent No. 9,358,533 ("*Tran*")**
	D_{90}, respectively. The D_{90} of the hollow microsphere alumina is about 10 μmas compared to about 100 nm (0.1 μm) for the starting pseudo boehmite, as shown in FIG. 3. After calcination at 550° C., the hollow sphere structure was fairly robust against milling or hydrothermal treatment. For example, the hollow structures remained relatively intact after a hydrothermal aging at 950° C./10% H_2O/air/4 hours or a physical ball-milling for about 30 minutes. The fine pseudo boehmite particles were changed to gamma alumina after the calcination and formed a strong bonded hollow microsphere structure." [11:4-15] "A coating slurry was made by mixing the 2% Pd/alumina powder with an alumina binder and DI-water. The slurry was ball-milled, which yielded a particle size of D_{90} about 10 μm. The slurry was coated on a ceramic monolith honeycomb core (1 inch diameter by 1 inch height). The coated core was dried and then calcined at 550° C. in air for 2 hours. The DG of powder on the core was 0.60 gram/core, which yielded a precious metal loading of 25 grams/ft^3. For comparison, the hollow microsphere alumina was replaced by a reference La-stabilized alumina particle or by a La-stabilized solid sphere alumina while other catalyst compositions remained the same." To the extent that this limitation is not disclosed, either explicitly or inherently, by *Tran*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
1[b] silica, alumina, or mixtures thereof; and	*Tran* discloses a combustion promoter comprising silica, alumina, or mixtures thereof. *See, e.g.*,

EXHIBIT O

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,358,533 ("*Tran*")

'864 patent	**U.S. Patent No. 9,358,533 ("*Tran*")**
	[1:42-52] "Refractory metal oxides such as alumina, rare-earth metal oxides, zirconia, titania, and their combinations, and other materials are commonly used as supports for the catalytic components of a catalyst article and as oxygen storage materials (OSC). Currently, almost all of the alumina catalyst supports and OSC are in the form of solid powder particles with a particle size ranging from about 5-100 microns or are large extrudates above 100 microns in size. The alumina support materials typically exhibit a BET surface area in excess of 60 square meters per gram ('m^2/g'), often up to about 200 m^2/g or higher." [1:53-2:16] "In an internal combustion engine it is also desirable for these catalyst support materials to have high meso- and macro-porosity to enhance gas phase diffusion, which makes the catalysts more effective to achieve high nitrogen oxide (NOx) and hydrocarbon (HC) conversion at high space-velocity. In this regard, porous microspheres, including hollow microspheres, have been used as catalyst supports for the purpose of improving the porosity of the catalytic washcoat. Various preparation methods for such microspheres are reported in the literature. However, in general, hollow alumina microspheres that are formed at lower temperatures are thin-walled egg-shell structures that are too fragile to resist mechanical milling during catalyst preparation and hydrothermal aging in the engine. Thick-walled hollow alumina spheres are more robust against mechanical and thermal aging, and are available commercially (e.g., as insulation material), but these materials either have a large particle size or have been sintered at too high a temperature for catalyst applications. Hollow alumina microspheres made using ion-extraction of boehmite sols followed by firing at 1200° C. have been shown to have thick walls; however, these micro-spheres are in the dense alpha crystalline phase.

EXHIBIT O

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,358,533 ("*Tran*")

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	There remains a need for hollow porous microspheres suitable for use as catalyst supports which can be made by simple manufacturing methods, and which have thick walls with small spherical diameters. The availability of such microspheres also leads to a significant reduction in raw material usage (e.g., precious metal, alumina, and OSC) and therefore a substantial reduction in cost because of elimination of the dead space in the center of the conventional solid particle." [4:22-37] "Certain aspects of the invention provide hollow porous microspheres suitable for use as catalyst supports, the micro-spheres comprising agglomerated nanoparticles of metal oxide, such as a high surface area metal oxide. The catalyst support is useful for supporting one or more catalysts or catalyst components on its surface or within the walls of the hollow microsphere. In one or more embodiments, the metal oxide comprises an activated compound selected from the group consisting of alumina, boehmite, pseudoboehmite, ceria, zirconia, ceria-zirconia, alumina-zirconia, alumina-ceria-zirconia, lanthana-alumina, lanthana-zirconia-alumina, baria-alumina, baria-lanthana-alumina, baria-lanthana-neodymia alumina, and alumina-ceria. In one or more specific embodiments the nanoparticles are alumina or boehmite nanoparticles. In further specific embodiments the nanoparticles are CeO_2 nanoparticles." [5:9-24] "In a specific embodiment, the microsphere comprises zirconium oxide on or near its exterior surface. It has been observed that zirconium oxide which is added to the slurry with alumina nanoparticles prior to spray-drying accumulates in higher concentration at or on the exterior surface of the microsphere during spray-drying, resulting in an exterior layer of zirconium oxide that is visible on SEM. This layered configuration is very useful as a barrier to separate a rhodium catalyst from alumina in the microspheres. The multi-layer structure is shown in FIG. 1B, where the exterior layer of zirconium oxide is seen as a lighter layer over the alumina walls of

EXHIBIT O

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,358,533 ("*Tran*")

'864 patent	U.S. Patent No. 9,358,533 ("*Tran*")
	the hollow microsphere on SEM. FIG. 1A shows that a similar zirconium oxide layer is also formed on the exterior surface of the solid alumina nanoparticles when they are spray dried without the soft polymer template and do not form hollow microspheres." [5:63-6:17] "It should be pointed out that the micro-porosity of the hollow porous microspheres is similar to the corresponding solid particle metal oxide, as illustrated in the following example of hollow porous alumina microspheres. This is because the formation of the hollow sphere structure increases the macro-porosity of the materials, but not their microsphere structures. FIGS. 2A, 2B and 2C are SEMs illustrating the difference in macroporosity of different Al_2O_3/ OSC washcoats. FIG. 2A shows the traditional washcoat, comprised of solid particles of alumina (dark color) and OSC (lighter color). It can be seen that the traditional wash- coat has low macroporosity, which results in limitation of gas diffusion through the washcoat. FIG. 2B shows a washcoat comprised of a mixture of hollow microsphere alumina according to the invention with solid OSC particles. This formulation produces a washcoat with increased macroporosity compared to the traditional washcoat, and it exhibits improved catalyst activity. FIG. 2C shows a washcoat comprised entirely of hollow microsphere alumina and OSC according to the invention. This washcoat has the highest macroporosity and therefore the highest rate of gas diffusion through the washcoat and most improved catalyst activity." [6:50-59] "The ceramic substrate may be made of any suitable refractory material, e.g., cordierite, cordierite-a alumina, silicon nitride, zircon mullite, spodumene, alumina-silica magnesia, zircon silicate, sillimanite, magnesium silicates, zircon, petalite, α-alumina, aluminosilicates and the like. The substrates useful for the catalyst supports of the present invention may also be composed of one or more metals or metal

EXHIBIT O

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,358,533 ("*Tran*")

'864 patent	U.S. Patent No. 9,358,533 ("*Tran*")
	alloys. The metallic substrates may be employed in various shapes such as corrugated sheet, metal plate, wire mesh or monolithic form." [7:41-47] "Nanoparticles of any suitable metal oxide may be used in the methods for manufacturing the hollow porous microspheres as discussed above; however, in specific embodiments the metal oxide is boehmite or pseudoboehmite, which have the advantage of a much smaller particle size than activated alumina such as γ-alumina for making the hollow microsphere alumina." [9:51-65] "Average particle size was measured on a Horiba LA-950 particle size analyzer. For a typical measurement, 0.1 g of sorbent was slurried in water. The particle size (diameter) is expressed as values less than 50% and 90% of total particles, D_{50} and D_{90}, respectively. The D_{90} of the hollow microsphere alumina is about 10 μmas compared to about 100 nm (0.1 μm) for the starting pseudo boehmite, as shown in FIG. 3. After calcination at 550° C., the hollow sphere structure was fairly robust against milling or hydrothermal treatment. For example, the hollow structures remained relatively intact after a hydrothermal aging at 950° C./10% H_2O/air/4 hours or a physical ball-milling for about 30 minutes. The fine pseudo boehmite particles were changed to gamma alumina after the calcination and formed a strong bonded hollow microsphere structure."

EXHIBIT O

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,358,533 ("*Tran*")

<table>
<tr><th>'864 patent</th><th>U.S. Patent No. 9,358,533 ("Tran")</th></tr>
<tr><td></td><td>[10:26-40]
TABLE 1
N_2 porosity of hollow microsphere alumina vs. solid alumina particles
<table><tr><th></th><th colspan="2">BET, m^2/g</th><th colspan="2">Pore volume, cc/g</th><th colspan="2">Pore Size, Å</th></tr><tr><th>Sample</th><th>Fresh</th><th>Aged</th><th>Fresh</th><th>Aged</th><th>Fresh</th><th>Aged</th></tr><tr><td>Solid Alumina Particles</td><td>191</td><td>131</td><td>0.43</td><td>0.43</td><td>89</td><td>133</td></tr><tr><td>Hollow Micro sphere Alumina</td><td>197</td><td>137</td><td>0.43</td><td>0.44</td><td>88</td><td>130</td></tr><tr><td>Hollow Micro sphere Alumina coated w/ZrO_2</td><td>181</td><td>129</td><td>0.34</td><td>0.35</td><td>76</td><td>108</td></tr></table>
To the extent that this limitation is not disclosed, either explicitly or inherently, by Tran, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.</td></tr>
<tr><td>1[c] one or more Group VIII noble metals distributed in the particle as an eggshell such that a higher concentration of the one or more Group VIII noble metals is present in the outer region of the microsphere sized porous particle as compared to the</td><td>Tran discloses a combustion promoter comprising one or more Group VIII noble metals, the metals distributed in the particle as an eggshell such that a higher concentration of the one or more Group VIII noble metals is present in the outer region of the microsphere sized porous particle as compared to the concentration of the one or more Group VIII noble metals in the centre of the microsphere sized porous particle. See, e.g.,</td></tr>
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EXHIBIT O

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,358,533 ("*Tran*")

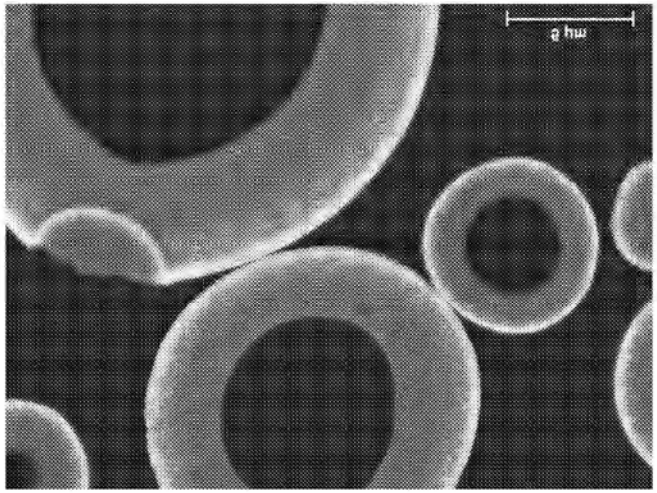
'864 patent	**U.S. Patent No. 9,358,533 ("*Tran*")**
concentration of the one or more Group VIII noble metals in the centre of the microsphere sized porous particle.	[Fig. 1B]  Fig. 1B

EXHIBIT O

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,358,533 ("*Tran*")

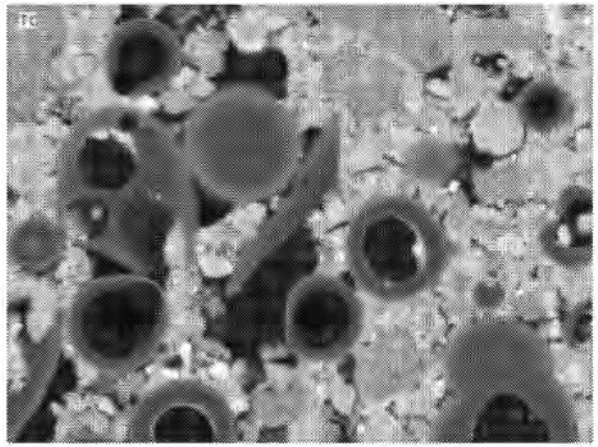
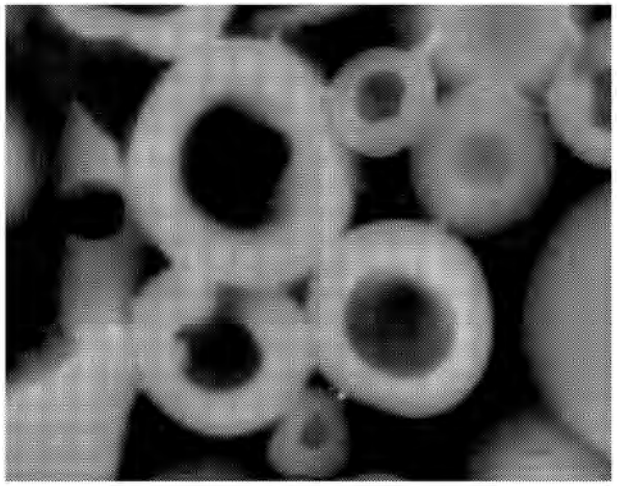
'864 patent	**U.S. Patent No. 9,358,533 ("*Tran*")**
	[Fig. 2B]  Fig. 2B [Fig. 2C]  Fig. 2C

EXHIBIT O

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,358,533 ("*Tran*")

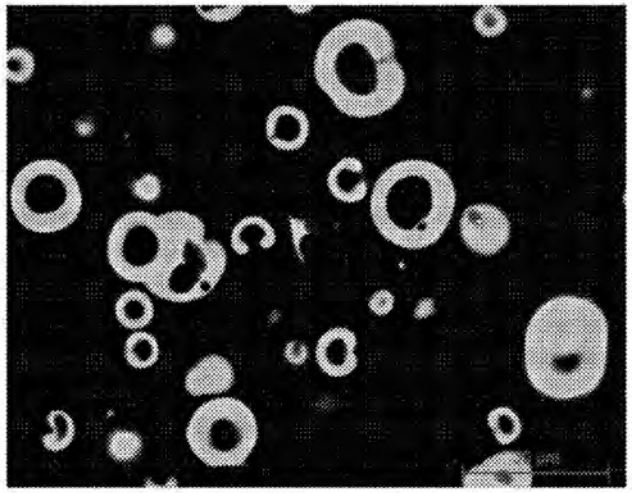
'864 patent	U.S. Patent No. 9,358,533 ("*Tran*")
	[Fig. 3B]  Fig. 3B [1:13-41] "The exhaust gases of internal combustion engines contain pollutants such as hydrocarbons, carbon monoxide and nitro- gen oxides (NO_x) that foul the air. Emission standards for unburned hydrocarbons, carbon monoxide and nitrogen oxide contaminants have been set by various governments and must be met by older as well as new vehicles. In order to meet such standards, catalytic converters containing a three way catalyst (TWC) may be located in the exhaust gas line of internal combustion engines. The use of exhaust gas catalysts have contributed to a significant improvement in air quality. The TWC is the most commonly used catalyst and it provides the three functions of oxidation of CO, oxidation of unburned hydrocarbons (HC's) and reduction of NOx to N_2. TWCs typically utilize one or more platinum group metals (PGM) to simultaneously oxidize CO and HC

EXHIBIT O

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,358,533 ("*Tran*")

'864 patent	U.S. Patent No. 9,358,533 ("*Tran*")
	and reduce NOx compounds. The most common catalytic components of a TWC are platinum (Pt), rhodium (Rh) and palladium (Pd). The platinum group metals (PGM) in the TWC catalysts (e.g., platinum, palladium, rhodium, ruthenium and iridium) are typically dispersed on a high surface area, refractory metal oxide support, e.g., a high surface area alumina coating, or on an oxygen storage component (OSC), or their mixtures. The support is carried on a suitable carrier or substrate such as a monolithic substrate comprising a refractory ceramic or metal honeycomb structure, or refractory particles such as spheres or short, extruded segments of a suitable refractory material. The TWC catalyst substrate may also be a wire mesh, typically a metal wire mesh, which is particularly useful in small engines." [1:53-2:16] "In an internal combustion engine it is also desirable for these catalyst support materials to have high mesa- and macro-porosity to enhance gas phase diffusion, which makes the catalysts more effective to achieve high nitrogen oxide (NOx) and hydrocarbon (HC) conversion at high space-velocity. In this regard, porous microspheres, including hollow microspheres, have been used as catalyst supports for the purpose of improving the porosity of the catalytic washcoat. Various preparation methods for such microspheres are reported in the literature. However, in general, hollow alumina microspheres that are formed at lower temperatures are thin-walled egg-shell structures that are too fragile to resist mechanical milling during catalyst preparation and hydrothermal aging in the engine. Thick-walled hollow alumina spheres are more robust against mechanical and thermal aging, and are available commercially (e.g., as insulation material), but these materials either have a large particle size or have been sintered at too high a temperature for catalyst applications. Hollow alumina microspheres made using ion- extraction of boehmite sols followed by firing at 1200° C. have been shown to have thick walls; however, these micro- spheres are in the dense alpha crystalline phase.

EXHIBIT O

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,358,533 ("*Tran*")

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	There remains a need for hollow porous microspheres suit- able for use as catalyst supports which can be made by simple manufacturing methods, and which have thick walls with small spherical diameters. The availability of such micro- spheres also leads to a significant reduction in raw material usage (e.g., precious metal, alumina, and OSC) and therefore a substantial reduction in cost because of elimination of the dead space in the center of the conventional solid particle." [2:30-34] "In one embodiment of the hollow porous metal oxide microspheres, a catalyst is incorporated within the walls of the hollow metal oxide microspheres. In an alternative embodiment, a catalyst is present in higher concentration on or near the external surface of the microspheres." [3:16-20] "In any embodiment of the hollow porous metal oxide microsphere compositions or catalytic articles comprising a catalyst, the catalyst may be a platinum group metal. Suitable platinum group metals include any of platinum, palladium, rhodium and combinations thereof." [3:44-51] "FIG. 6 illustrates the results of Example 5, showing the effect of catalyst support on the light-off temperature for single layer, Pd catalyst core samples. FIG. 7 illustrates the results of Example 6, comparing CO, hydrocarbon and nitric oxide conversion for a hollow alumina microsphere/platinum/palladium/rhodium catalyst and a solid alumina particle/platinum/palladium/rhodium catalyst in a motorcycle application."

EXHIBIT O

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,358,533 ("*Tran*")

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	[4:22-37] "Certain aspects of the invention provide hollow porous microspheres suitable for use as catalyst supports, the micro- spheres comprising agglomerated nanoparticles of metal oxide, such as a high surface area metal oxide. The catalyst support is useful for supporting one or more catalysts or catalyst components on its surface or within the walls of the hollow microsphere. In one or more embodiments, the metal oxide comprises an activated compound selected from the group consisting of alumina, boehmite, pseudoboehmite, ceria, zirconia, ceria-zirconia, alumina-zirconia, alumina-ceria-zirconia, lanthana-alumina, lanthana-zirconia-alumina, baria-alumina, baria-lanthana-alumina, baria-lanthana-neodymia alumina, and alumina-ceria. In one or more specific embodiments the nanoparticles are alumina or boehmite nanoparticles. In further specific embodiments the nanoparticles are CeO_2 nanoparticles." [6:18-27] "Another aspect of the invention provides a catalyst article comprising a catalyst layer on a substrate, the catalyst layer comprising the hollow porous microspheres according to any of the foregoing embodiments as a catalyst support. The hollow porous microspheres comprise agglomerated metal oxide nanospheres, a catalyst supported by the catalyst support, and, optionally, a stabilizer. The catalyst support may in the form of a washcoat on the substrate. In specific embodiments, the catalyst may be one or more PGM catalysts, such as palladium, platinum, rhodium or combinations thereof." [7:59-8:8] "In certain embodiments, a catalyst or catalyst component (for example one or more PGMs) is included in the slurry prior to spray-drying, in which case the catalyst is incorporated into the walls of the microsphere catalyst support in a single step. Alternatively, the catalyst may be impregnated into the walls or on the surface of the microsphere catalyst support after spray-drying using conventional impregnation

EXHIBIT O

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,358,533 ("*Tran*")

'864 patent	U.S. Patent No. 9,358,533 ("*Tran*")
	techniques. For example, a solution of a catalyst or catalyst component (e.g., platinum group metal) may be put into the pores of the catalyst support by incipient wetness, where a volume of diluted platinum group metal is approximately equal to the pore volume of the catalyst support. Incipient wetness impregnation generally leads to a substantially uniform distribution of the solution of the precursor throughout the pore system of the walls or an enriched layer of the catalyst on the surface of the hollow microsphere catalyst support." [11:21-52] "(1) Formation of the bottom catalytic layer. A palladium component in the form of a 20% aqueous solution and a platinum component in the form of 13% aqueous solution were mixed with the La-stabilized hollow microsphere alumina and water to form a wet powder achieved by incipient wetness. Separately, a palladium component in the form of 20% solution was mixed with OSC materials and water to form a wet powder achieved by incipient wetness. The two wet powders were mixed and milled to a particle size of D_{90} about 20 μm. Ceria-zirconia, octanol, acetic acid, alumina binder, and zirconium acetate were added and combined with the Pd/support mixture in a planetary mixer (P-mixer). The slurry was coated onto a metallic support carrier using deposition methods known in the art for depositing the catalyst on a metal substrate. After coating, the carrier with the bottom catalytic layers was dried, then calcined at a temperature of 550° C for about 2 hours. The final bottom coat catalyst composition contained the following components: La-stabilized hollow microsphere alumina 54.7% of dry gain (DG), OSC 41.0% of DG, barium oxide 2.7% of DG, palladium 1.6% of DG, and platinum 0.01% of DG. (2) Formation of the top coat catalytic layer. The second catalytic layer consisting of rhodium and platinum supported on OSC materials was coated on the first catalytic layer (bottom coat) using substantially similar procedures for the bottom coat. The final top coat catalyst layer composition contained the following components: OSC 69.6% of dry gain (DG), Zr-stabilized alumina 27.8% of DG, ZrO_2 2.3% of DG,

EXHIBIT O

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,358,533 ("*Tran*")

'864 patent	U.S. Patent No. 9,358,533 ("*Tran*")
	rhodium 0.19% of DG, and platinum 0.08% of DG. For comparison, the hollow microsphere alumina was replaced by a reference La-stabilized alumina particle in the bottom coat while other catalyst compositions remained the same." [10:58-11:15] "Example 3: Preparation of a Single Layer Core Catalyst Containing Palladium Supported on Hollow Microsphere Alumina A powder catalyst containing hollow sphere alumina was prepared by impregnating hollow microsphere alumina or La- or Ba-stabilized hollow microsphere alumina with palladium nitrate aqueous solution. The impregnated paste was dried at 110° C. overnight, ground, and calcined at 550° C. in air for 2 hours. The dry gain (DG) of palladium on the alumina was 2%. A coating slurry was made by mixing the 2% Pd/alumina powder with an alumina binder and DI-water. The slurry was ball-milled, which yielded a particle size of D_{90} about 10 μm. The slurry was coated on a ceramic monolith honeycomb core (1 inch diameter by 1 inch height). The coated core was dried and then calcined at 550° C. in air for 2 hours. The DG of powder on the core was 0.60 gram/core, which yielded a precious metal loading of 25 grams/ft^3. For comparison, the hollow microsphere alumina was replaced by a reference La-stabilized alumina particle or by a La-stabilized solid sphere alumina while other catalyst compositions remained the same." [11:21-52] "(1) Formation of the bottom catalytic layer. A palladium component in the form of a 20% aqueous solution and a platinum component in the form of 13% aqueous solution were mixed with the La-stabilized hollow microsphere alumina and water to form a wet powder achieved by incipient wetness. Separately, a palladium component in the form of 20% solution was mixed with OSC materials and water to

EXHIBIT O

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 9,358,533 ("*Tran*")

'864 patent	**U.S. Patent No. 9,358,533 ("*Tran*")**
	form a wet powder achieved by incipient wetness. The two wet powders were mixed and milled to a particle size of D_{90} about 20 μm. Ceria-zirconia, octanol, acetic acid, alumina binder, and zirconium acetate were added and combined with the Pd/support mixture in a planetary mixer (P-mixer). The slurry was coated onto a metallic support carrier using deposition methods known in the art for depositing the catalyst on a metal substrate. After coating, the carrier with the bottom catalytic layers was dried, then calcined at a temperature of 550° C. for about 2 hours. The final bottom coat catalyst composition contained the following components: La-stabilized hollow microsphere alumina 54.7% of dry gain (DG), OSC 41.0% of DG, barium oxide 2.7% of DG, palladium 1.6% of DG, and platinum 0.01% of DG. (2) Formation of the top coat catalytic layer. The second catalytic layer consisting of rhodium and platinum supported on OSC materials was coated on the first catalytic layer (bot- tom coat) using substantially similar procedures for the bot- tom coat. The final top coat catalyst layer composition contained the following components: OSC 69.6% of dry gain (DG), Zr-stabilized alumina 27.8% of DG, ZrO_2 2.3% of DG, rhodium 0.19% of DG, and platinum 0.08% of DG. For comparison, the hollow microsphere alumina was replaced by a reference La-stabilized alumina particle in the bottom coat while other catalyst compositions remained the same." To the extent that this limitation is not disclosed, either explicitly or inherently, by *Tran*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[3] The combustion promoter according to claim 1, comprising a plurality of	*Tran* discloses a combustion promoter comprising a plurality of microsphere sized porous particles comprising alumina. *See, e.g.*,

EXHIBIT O

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microsphere sized porous particles comprising alumina.	The support provided above for Claim 1[b]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Tran*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[6] The combustion promoter according to claim 1, wherein the microsphere sized porous particles collectively have an average (D50) size of between 60 and 90 microns as measured by laser diffraction.	*Tran* discloses a combustion promoter comprising microsphere sized porous particles, wherein the microsphere sized porous particles collectively have an average (D50) size of between 60 and 90 microns as measured by laser diffraction. *See, e.g.*, The support provided above for Claim 1[a]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Tran*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[12] The combustion promoter according to claim 1, wherein the one or more Group VIII noble metals comprises one or more of	*Tran* discloses a combustion promoter comprising one or more Group VIII noble metals, wherein the one or more Group VIII noble metals comprises one or more of platinum, palladium, iridium, ruthenium, and rhodium. *See, e.g.*,

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platinum, palladium, iridium, ruthenium, and rhodium.	The support provided above for Claim 1[c]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Tran*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[15] The combustion promoter according to claim 12, wherein the one or more Group VIII noble metals comprises palladium.	*Tran* discloses a combustion promoter comprising one or more Group VIII noble metals, wherein the one or more Group VIII noble metals comprises palladium. *See, e.g.*, The support provided above for Claim 1[c]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Tran*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.

EXHIBIT P

EXHIBIT P

Invalidity of U.S. Patent No. 11,224,864 in View of *Venuto Article*

"*Fluid Catalytic Cracking with Zeolite Catalysts*" ("*Venuto Article*"), authored by Paul B. Venuto et al. and published in 1979, qualifies as prior art of U.S. Patent No. 11,224,864 ("the '864 patent") under at least 35 U.S.C. §§ 102 and/or 103 for the asserted claims of the '864 patent, as discussed in detail in the claim chart below.

The Court has not yet construed the claims and therefore the meaning of the terms in the claims has yet to be resolved by the Court. The support identified here for these limitations is responsive to Plaintiffs' infringement contentions, which Defendant disagrees with. As such, nothing in Defendant's claim charts should be construed as an admission regarding infringement, either literally or under the doctrine of equivalents, or as an admission regarding Defendant's understanding of the proper scope of the asserted claims. Defendant reserves the right to rely on additional citations or sources of evidence that also may be applicable, or that may become applicable in light of claim construction, changes in Plaintiffs' infringement contentions, and/or information obtained during discovery as the case progresses.

To the extent Plaintiffs allege that *Venuto Article* does not disclose any particular limitation of the asserted claims of the '864 patent, either expressly or inherently, it would have been obvious to a person of ordinary skill in the art as of the priority date of the '864 patent to modify *Venuto Article* and/or to combine the teachings of *Venuto Article* with other prior art references, including but not limited to the present prior art references cited in the Cover Pleading and the relevant section(s) of claim charts for other prior art references for the '864 patent in a manner that would have rendered the asserted claims invalid as obvious.

Defendant reserves the right to amend or supplement this claim chart at a later date as more fully set forth in the Cover Pleading.

EXHIBIT P

Invalidity of U.S. Patent No. 11,224,864 in View of *Venuto Article*

'864 patent	***"Fluid Catalytic Cracking with Zeolite Catalysts"* ("*Venuto Article*")**
1[a] A CO to CO_2 combustion promoter comprising microsphere sized porous particles, each microsphere sized porous particle having a diameter of less than 1mm, and independently comprising	*Venuto Article* discloses a CO to CO_2 combustion promoter comprising microsphere sized porous particles, each microsphere sized porous particle having a diameter of less than 1mm, and independently comprising the limitations discussed in the following sections. *See, e.g.*, [Page 30-31] "As noted in Table 1, synthetic silica-alumina was introduced as a cracking catalyst about 1940 and, in one modification or another, dominated the cracking catalyst field until the early 1960s and the introduction of the zeolites [13, 17, 18]. However, over the decade of the 1950s, and particularly during the latter half, only incremental improvements in selectivity and regenerability of the amorphous catalysts (e.g., via silica-magnesia variations [128, 154, 155]) were achieved, and it remained for the zeolite catalysts to truly revolutionize the cracking catalyst field. Plank and Rosinski (see the series of patents issued to Mobil Oil [156-161]) discovered that incorporation of a minor proportion of zeolite (such as rare-earth cation exchanged X or Y synthetic faujasite) in a matrix such as silica-alumina, silica, or silica-clay, produced compositions of matter that, over more than a decade, have met, to a large extent, the most severe requirements [13, 17, 18] for successful cracking catalysts, including: 1. High activity (allowing economic throughput rates) 2. Reasonable activity stability (resistance to time-dependent deactivation by coke) 3. High selectivity to gasoline vs coke and dry gas 4. Thermal and hydrothermal stability (ruggedness to withstand the regenerator environment) 5. Accessibility (sufficient diffusivity and internal porosity to allow small, economic reactor volumes) 6. Attrition resistance 7. Resistance to poisons (metals, nitrogen, etc.)

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'864 patent	***"Fluid Catalytic Cracking with Zeolite Catalysts"* ("*Venuto Article*")**
	8. Acceptably low cost In this section we attempt to focus on highlights of the zeolite cracking catalysts, limiting our discussion of the amorphous silica- alumina type to selected comparisons only, as these have been well described by Sittig [10], Voge [11], and others [45, 122, 128, 162-180]."

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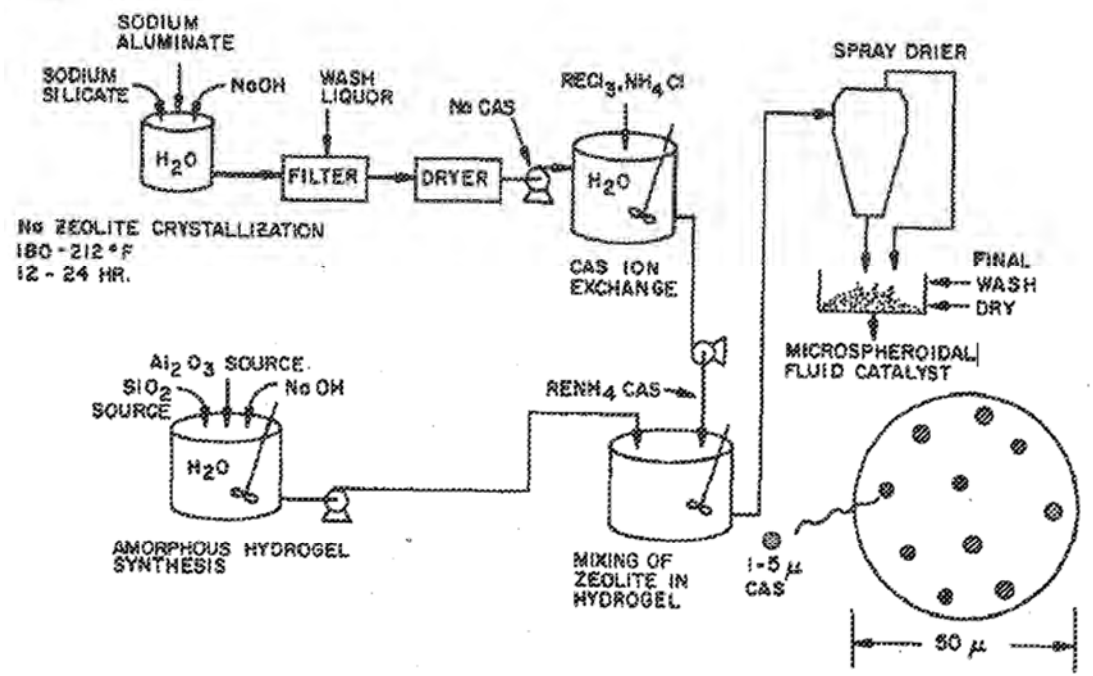
'864 patent	***"Fluid Catalytic Cracking with Zeolite Catalysts" ("Venuto Article")***
	[Page 31]  FIG. 13. Simplified conceptual scheme for manufacture of commercial fluid cracking catalyst (XZ-25). Modified after flow sheets in Koffler [181]. [Page 41-42] "As visualized in Fig. 13, the major operations in the manufacture of a commercial zeolite cracking catalyst [182] may include synthesis and ion exchange of the zeolite component, preparation of the amorphous gel, intimate admixture of the zeolite in the gel matrix, and spray drying.

EXHIBIT P

Invalidity of U.S. Patent No. 11,224,864 in View of *Venuto Article*

'864 patent	***"Fluid Catalytic Cracking with Zeolite Catalysts"* ("*Venuto Article*")**
	Manufacture of zeolites commercially involves combination of readily available, inexpensive, raw materials, including a silica source, an alumina source, and a strong base such as sodium hydroxide [13, 181, 183, 184]. These materials, in proper stoichiometric ratios, molarity, and state (i.e., solution or colloidal since not all deposited metals are equally effective in producing coke and hydrogen [12]. On a weight basis, nickel shows four times the coke- and hydrogen-forming tendency of vanadium [29, 224], and similar relative responses for both metals on amorphous silica-alumina have been reported [221]." [Page 42] "Freshly deposited metals are more active than "older" metals that have been subjected to numerous cycles in the FCC regenerator/reactor circuit [102]. Upon exposure to such repeated cycles of oxidation/reduction, the poisoning effects of metals contaminants are diminished, but those on zeolites lose their effectiveness more slowly than those on amorphous catalysts [28]. Grane, Conner, and Masologites [221], in a study of metals-poisoned silica-alumina, suggested that the repeated shift from oxidizing to reducing environment in going from regenerator to reactor may influence the oxidation state of the metals or cause sintering (and loss of surface area) with consequent deactivation, Morozov et al. [225] observe that, for a given feedstock, a greater metals buildup may occur on fluid catalysts than on bead types, because with the latter there is removal of significant amounts of surface metals by attrition. Montgomery [102] observes that sulfur in FCC feed may tend to sulfide-and there-by activate-the metals on a cracking catalyst. On the other hand, it has been observed that zeolite catalysts have excellent metals resistance, particularly in short contact time operation [92, 226]; this may occur because metal oxides on the circulating catalyst are not retained in the reaction zone long enough to become sufficiently sulfided to show strong dehydrogenation activity [92, 226]."

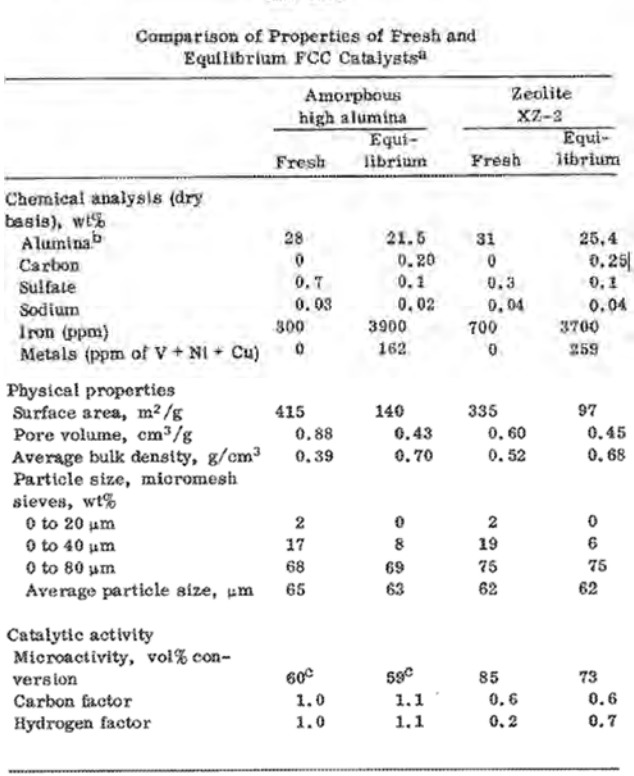
EXHIBIT P

Invalidity of U.S. Patent No. 11,224,864 in View of *Venuto Article*

'864 patent	***"Fluid Catalytic Cracking with Zeolite Catalysts"* ("*Venuto Article*")**
	[Page 43] "In a sense, the matrix protects the small (~1 to 5 μm), relatively fragile zeolite crystallites, building the composite up to a ~50 μm microsphere and providing shape and bulk for fluidization [76] and minimizing mechanical attrition (see Fig. 13). Eastwood, Plank, and Weisz [101] observe that the high surface area matrix surrounding and diluting the zeolite crystals acts as a heat carrier or 'sink,' facilitating heat transfer during regeneration and cracking. The stability of the composite to high severity steaming is also enhanced [13, 101]."

EXHIBIT P

Invalidity of U.S. Patent No. 11,224,864 in View of *Venuto Article*

'864 patent	***"Fluid Catalytic Cracking with Zeolite Catalysts" ("Venuto Article")***
	[Page 46] TABLE 8 Comparison of Properties of Fresh and Equilibrium FCC Catalysts[a] 

	Amorphous high alumina		Zeolite XZ-2	
	Fresh	Equilibrium	Fresh	Equilibrium
Chemical analysis (dry basis), wt%				
Alumina[b]	28	21.5	31	25.4
Carbon	0	0.20	0	0.25
Sulfate	0.7	0.1	0.3	0.1
Sodium	0.03	0.02	0.04	0.04
Iron (ppm)	300	3900	700	3700
Metals (ppm of V + Ni + Cu)	0	162	0	259
Physical properties				
Surface area, m^2/g	415	140	335	97
Pore volume, cm^3/g	0.88	0.43	0.60	0.45
Average bulk density, g/cm^3	0.39	0.70	0.52	0.68
Particle size, micromesh sieves, wt%				
0 to 20 μm	2	0	2	0
0 to 40 μm	17	8	19	6
0 to 80 μm	68	69	75	75
Average particle size, μm	65	63	62	62
Catalytic activity				
Microactivity, vol% conversion	60[c]	59[c]	85	73
Carbon factor	1.0	1.1	0.6	0.6
Hydrogen factor	1.0	1.1	0.2	0.7

[Page 47]
"Surface areas are sharply lower (by a factor of ~3 to 3.5) in equilibrium vs fresh catalyst; likewise, there is a drop in pore volume and an increase in bulk density. Letzsch [227] suggests that, in some cases, the initial loss in surface area with catalyst use may in large part be due to the collapse of small matrix pores and some zeolite regions which were not properly stabilized (i.e., had high residual sodium or inadequate rare-earth exchange levels)."

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Invalidity of U.S. Patent No. 11,224,864 in View of *Venuto Article*

'864 patent	***"Fluid Catalytic Cracking with Zeolite Catalysts"* ("*Venuto Article*")**
	To the extent that this limitation is not disclosed, either explicitly or inherently, by *Venuto Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
1[b] silica, alumina, or mixtures thereof; and	*Venuto Article* discloses a combustion promoter comprising silica, alumina, or mixtures thereof. *See, e.g.*, [Page 30-31] "As noted in Table 1, synthetic silica-alumina was introduced as a cracking catalyst about 1940 and, in one modification or another, dominated the cracking catalyst field until the early 1960s and the introduction of the zeolites [13, 17, 18]. However, over the decade of the 1950s, and particularly during the latter half, only incremental improvements in selectivity and regenerability of the amorphous catalysts (e.g., viasilica-magnesia variations [128, 154, 155]) were achieved, and it remained for the zeolite catalysts to truly revolutionize the cracking catalyst field. Plank and Rosinski (see the series of patents issued to Mobil Oil [156-161]) discovered that incorporation of a minor proportion of zeolite (such as rare-earth cation exchanged X or Y synthetic faujasite) in a matrix such as silica-alumina, silica, or silica-clay, produced compositions of matter that, over more than a decade, have met, to a large extent, the most severe requirements [13, 17, 18] for successful cracking catalysts, including: 1. High activity (allowing economic throughput rates) 2. Reasonable activity stability (resistance to time-dependent deactivation by coke) 3. High selectivity to gasoline vs coke and dry gas

EXHIBIT P

Invalidity of U.S. Patent No. 11,224,864 in View of *Venuto Article*

'864 patent	***"Fluid Catalytic Cracking with Zeolite Catalysts"* ("*Venuto Article*")**
	4. Thermal and hydrothermal stability (ruggedness to withstand the regenerator environment) 5. Accessibility (sufficient diffusivity and internal porosity to allow small, economic reactor volumes) 6. Attrition resistance 7. Resistance to poisons (metals, nitrogen, etc.) 8. Acceptably low cost In this section we attempt to focus on highlights of the zeolite cracking catalysts, limiting our discussion of the amorphous silica- alumina type to selected comparisons only, as these have been well described by Sittig [10], Voge [11], and others [45, 122, 128, 162-180]."

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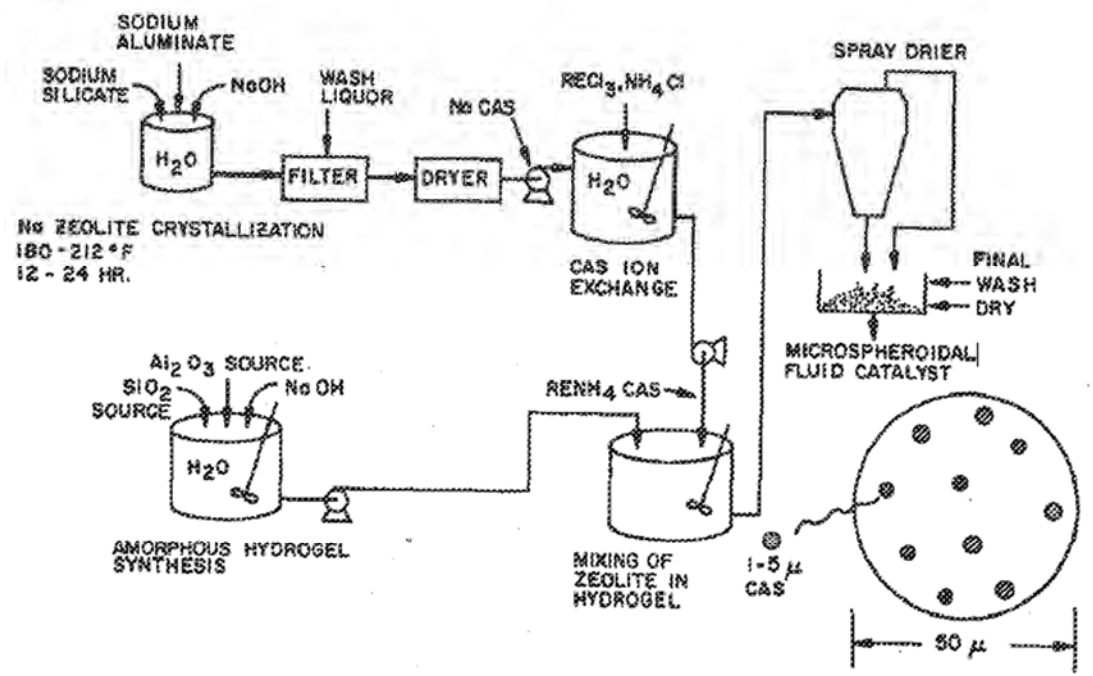
'864 patent	***"Fluid Catalytic Cracking with Zeolite Catalysts" ("Venuto Article")***
	[Page 31]  FIG. 13. Simplified conceptual scheme for manufacture of commercial fluid cracking catalyst (XZ-25). Modified after flow sheets in Koffler [181]. [Page 41-42] "As visualized in Fig. 13, the major operations in the manufacture of a commercial zeolite cracking catalyst [182] may include synthesis and ion exchange of the zeolite component, preparation of the amorphous gel, intimate admixture of the zeolite in the gel matrix, and spray drying.

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'864 patent	***"Fluid Catalytic Cracking with Zeolite Catalysts"* ("*Venuto Article*")**
	Manufacture of zeolites commercially involves combination of readily available, inexpensive, raw materials, including a silica source, an alumina source, and a strong base such as sodium hydroxide [13, 181, 183, 184]. These materials, in proper stoichiometric ratios, molarity, and state (i.e., solution or colloidal since not all deposited metals are equally effective in producing coke and hydrogen [12]. On a weight basis, nickel shows four times the coke- and hydrogen-forming tendency of vanadium [29, 224], and similar relative responses for both metals on amorphous silica-alumina have been reported [221]." [Page 42] "Freshly deposited metals are more active than "older" metals that have been subjected to numerous cycles in the FCC regenerator/reactor circuit [102]. Upon exposure to such repeated cycles of oxidation/reduction, the poisoning effects of metals contaminants are diminished, but those on zeolites lose their effectiveness more slowly than those on amorphous catalysts [28]. Grane, Conner, and Masologites [221], in a study of metals-poisoned silica-alumina, suggested that the repeated shift from oxidizing to reducing environment in going from regenerator to reactor may influence the oxidation state of the metals or cause sintering (and loss of surface area) with consequent deactivation, Morozov et al. [225] observe that, for a given feedstock, a greater metals buildup may occur on fluid catalysts than on bead types, because with the latter there is removal of significant amounts of surface metals by attrition. Montgomery [102] observes that sulfur in FCC feed may tend to sulfide-and there-by activate-the metals on a cracking catalyst. On the other hand, it has been observed that zeolite catalysts have excellent metals resistance, particularly in short contact time operation [92, 226]; this may occur because metal oxides on the circulating catalyst are not retained in the reaction zone long enough to become sufficiently sulfided to show strong dehydrogenation activity [92, 226]."

EXHIBIT P

Invalidity of U.S. Patent No. 11,224,864 in View of *Venuto Article*

'864 patent	***"Fluid Catalytic Cracking with Zeolite Catalysts"* ("*Venuto Article*")**
	[Page 42] "The role of the matrix in composite catalysts has been discussed in some detail by several authors [13, 101, 125]. Matrices encompass a broad range of SiO_2 – Al_2O_3 gels, clays, Al_2O_3, and other siliceous materials, and may be expected to vary in different catalysts [227]. Natural, chemically treated, and calcined clay have also been specified [158] as possible constituents. Although most fundamental studies have been conducted on zeolites alone, Oblad [13] points out that a fair amount of catalyst mechanistic patterning may also be true for matrix-zeolite blends."

EXHIBIT P

Invalidity of U.S. Patent No. 11,224,864 in View of *Venuto Article*

'864 patent	***"Fluid Catalytic Cracking with Zeolite Catalysts" ("Venuto Article")***

[Page 46]

TABLE 8

Comparison of Properties of Fresh and Equilibrium FCC Catalysts[a]

	Amorphous high alumina		Zeolite XZ-2	
	Fresh	Equi-librium	Fresh	Equi-librium
Chemical analysis (dry basis), wt%				
Alumina[b]	28	21.5	31	25.4
Carbon	0	0.20	0	0.25
Sulfate	0.7	0.1	0.3	0.1
Sodium	0.03	0.02	0.04	0.04
Iron (ppm)	300	3900	700	3700
Metals (ppm of V + Ni + Cu)	0	162	0	259
Physical properties				
Surface area, m^2/g	415	140	335	97
Pore volume, cm^3/g	0.88	0.43	0.60	0.45
Average bulk density, g/cm^3	0.39	0.70	0.52	0.68
Particle size, micromesh sieves, wt%				
0 to 20 μm	2	0	2	0
0 to 40 μm	17	8	19	6
0 to 80 μm	68	69	75	75
Average particle size, μm	65	63	62	62
Catalytic activity				
Microactivity, vol% conversion	60[c]	58[c]	85	73
Carbon factor	1.0	1.1	0.6	0.6
Hydrogen factor	1.0	1.1	0.2	0.7

[Page 47]

"Conversions are higher, and carbon producing (CPF) and hydrogen factors are lower for the zeolite, as expected. CPF is a useful guide to the degree of contamination of a catalyst, and is the ratio of coke yield for a given catalyst to that for a standard clean

EXHIBIT P

Invalidity of U.S. Patent No. 11,224,864 in View of *Venuto Article*

'864 patent	***"Fluid Catalytic Cracking with Zeolite Catalysts"* ("*Venuto Article*")**
	silica-alumina catalyst at the same conversion level [28]. The hydrogen factor-defined as the ratio of moles of H_2/moles of C_1^+ moles of C_2's – increases with metals level." To the extent that this limitation is not disclosed, either explicitly or inherently, by *Venuto Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
1[c] one or more Group VIII noble metals distributed in the particle as an eggshell such that a higher concentration of the one or more Group VIII noble metals is present in the outer region of the microsphere sized porous particle as compared to the concentration of the one or more Group VIII noble metals in the centre of the microsphere sized porous particle.	*Venuto Article* discloses a combustion promoter comprising one or more Group VIII noble metals, the metals distributed in the particle as an eggshell such that a higher concentration of the one or more Group VIII noble metals is present in the outer region of the microsphere sized porous particle as compared to the concentration of the one or more Group VIII noble metals in the centre of the microsphere sized porous particle. *See, e.g.*, [Page 41-42] "As visualized in Fig. 13, the major operations in the manufacture of a commercial zeolite cracking catalyst [182] may include synthesis and ion exchange of the zeolite component, preparation of the amorphous gel, intimate admixture of the zeolite in the gel matrix, and spray drying. Manufacture of zeolites commercially involves combination of readily available, inexpensive, raw materials, including a silica source, an alumina source, and a strong base such as sodium hydroxide [13, 181, 183, 184]. These materials, in proper stoichiometric ratios, molarity, and state (i.e., solution or colloidal since not all deposited metals are equally effective in producing coke and hydrogen [12]. On a weight basis, nickel shows four times the coke- and hydrogen-forming tendency of

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'864 patent	***"Fluid Catalytic Cracking with Zeolite Catalysts"* ("*Venuto Article*")**
	vanadium [29, 224], and similar relative responses for both metals on amorphous silica-alumina have been reported [221]." [Page 42] "Freshly deposited metals are more active than "older" metals that have been subjected to numerous cycles in the FCC regenerator/reactor circuit [102]. Upon exposure to such repeated cycles of oxidation/reduction, the poisoning effects of metals contaminants are diminished, but those on zeolites lose their effectiveness more slowly than those on amorphous catalysts [28]. Grane, Conner, and Masologites [221], in a study of metals-poisoned silica-alumina, suggested that the repeated shift from oxidizing to reducing environment in going from regenerator to reactor may influence the oxidation state of the metals or cause sintering (and loss of surface area) with consequent deactivation, Morozov et al. [225] observe that, for a given feedstock, a greater metals buildup may occur on fluid catalysts than on bead types, because with the latter there is removal of significant amounts of surface metals by attrition. Montgomery [102] observes that sulfur in FCC feed may tend to sulfide-and there-by activate-the metals on a cracking catalyst. On the other hand, it has been observed that zeolite catalysts have excellent metals resistance, particularly in short contact time operation [92, 226]; this may occur because metal oxides on the circulating catalyst are not retained in the reaction zone long enough to become sufficiently sulfided to show strong dehydrogenation activity [92, 226]." [Page 45-47] "Table 8 gives a representative comparison of the properties of fresh and equilibrium zeolite and amorphous FCC catalysts as reported by Baker and Blazek [124]. The 0.20 to 0.25% carbon on the equilibrium catalysts is the residual level remaining after regeneration. Note the relatively low sodium levels and the sharply higher levels of iron, and the appearance of contaminant metals such· as nickel and vanadium on the equilibrium catalysts. Bass [231] reports that clay-based catalysts surprisingly often have titanium inclusions in local concentrations far greater than levels indicated by bulk

EXHIBIT P

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'864 patent	***"Fluid Catalytic Cracking with Zeolite Catalysts"* ("*Venuto Article*")**
	chemical analysis. Typical rare-earth element distributions and levels were discussed earlier, With the use of a scanning electron microscope/energy dispersive x-ray analyzer, Bass has also been able to distinguish the presence of plate-like kaolin clay, granular gel, and crystalline zeolite in the interior of lightly ground cracking catalyst powder [231]." [Page 47] "Conversions are higher, and carbon producing (CPF) and hydrogen factors are lower for the zeolite, as expected. CPF is a useful guide to the degree of contamination of a catalyst, and is the ratio of coke yield for a given catalyst to that for a standard clean silica-alumina catalyst at the same conversion level [28]. The hydrogen factor-defined as the ratio of moles of H_2/moles of C_1^+ moles of C_2's – increases with metals level." To the extent that this limitation is not disclosed, either explicitly or inherently, by *Venuto Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[3] The combustion promoter according to claim 1, comprising a plurality of microsphere sized porous particles comprising alumina.	*Venuto Article* discloses a combustion promoter comprising a plurality of microsphere sized porous particles comprising alumina. *See, e.g.*, The support provided above for Claim 1[b]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Venuto Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for

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'864 patent	***"Fluid Catalytic Cracking with Zeolite Catalysts"* ("*Venuto Article*")**
	this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[6] The combustion promoter according to claim 1, wherein the microsphere sized porous particles collectively have an average (D50) size of between 60 and 90 microns as measured by laser diffraction.	*Venuto Article* discloses a combustion promoter comprising microsphere sized porous particles, wherein the microsphere sized porous particles collectively have an average (D50) size of between 60 and 90 microns as measured by laser diffraction. *See, e.g.*, The support provided above for Claim 1[a]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Venuto Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[12] The combustion promoter according to claim 1, wherein the one or more Group VIII noble metals comprises one or more of platinum, palladium, iridium, ruthenium, and rhodium.	*Venuto Article* discloses a combustion promoter comprising one or more Group VIII noble metals, wherein the one or more Group VIII noble metals comprises one or more of platinum, palladium, iridium, ruthenium, and rhodium. *See, e.g.*, The support provided above for Claim 1[c]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Venuto Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions

EXHIBIT P

Invalidity of U.S. Patent No. 11,224,864 in View of *Venuto Article*

'864 patent	***"Fluid Catalytic Cracking with Zeolite Catalysts"* ("*Venuto Article*")**
	as embodied in these references, examples of which are discussed in the Cover Pleading.
[15] The combustion promoter according to claim 12, wherein the one or more Group VIII noble metals comprises palladium.	*Venuto Article* discloses a combustion promoter comprising one or more Group VIII noble metals, wherein the one or more Group VIII noble metals comprises palladium. *See, e.g.*, The support provided above for Claim 1[c]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Venuto Article*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.

EXHIBIT Q

EXHIBIT Q

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 10,258,967 ("*Witte*")

U.S. Patent No. 10,258,967 ("*Witte*"), filed on March 20, 2015 and published on April 16, 2019, qualifies as prior art under at least 35 U.S.C. §§ 102 and/or 103 for the asserted claims of U.S. Patent No. 11,224,864 ("the '864 patent"), as discussed in detail in the claim chart below.

The Court has not yet construed the claims and therefore the meaning of the terms in the claims has yet to be resolved by the Court. The support identified here for these limitations is responsive to Plaintiffs' infringement contentions, which Defendant disagrees with. As such, nothing in Defendant's claim charts should be construed as an admission regarding infringement, either literally or under the doctrine of equivalents, or as an admission regarding Defendant's understanding of the proper scope of the asserted claims. Defendant reserves the right to rely on additional citations or sources of evidence that also may be applicable, or that may become applicable in light of claim construction, changes in Plaintiffs' infringement contentions, and/or information obtained during discovery as the case progresses.

To the extent Plaintiffs allege that *Witte* does not disclose any particular limitation of the asserted claims of the '864 patent, either expressly or inherently, it would have been obvious to a person of ordinary skill in the art as of the priority date of the '864 patent to modify *Witte* and/or to combine the teachings of *Witte* with other prior art references, including but not limited to the present prior art references cited in the Cover Pleading and the relevant section(s) of claim charts for other prior art references for the '864 patent in a manner that would have rendered the asserted claims invalid as obvious.

Defendant reserves the right to amend or supplement this claim chart at a later date as more fully set forth in the Cover Pleading.

EXHIBIT Q

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 10,258,967 ("*Witte*")

'864 patent	**U.S. Patent No. 10,258,967 ("*Witte*")**
1[a] A CO to CO_2 combustion promoter comprising microsphere sized porous particles, each microsphere sized porous particle having a diameter of less than 1mm, and independently comprising	*Witte* discloses a CO to CO_2 combustion promoter comprising microsphere sized porous particles, each microsphere sized porous particle having a diameter of less than 1mm, and independently comprising the limitations discussed in the following sections. *See, e.g.*, [Abstract] "The present invention is in the field of catalysis. More particularly, the present invention is directed to supported precious metal catalysts, preferably palladium and/or platinum metal catalysts, having a high metal loading, a high degree of dispersion and a high degree of edge-coating. The present invention is further directed to a process for producing these catalysts, as well as to the use of these catalysts in chemical reactions." [1:29-47] "Supported PM catalysts are typically prepared by a deposition-reduction method, in which the metal is first deposited on a support and then reduced. In order to prepare a catalyst with a high dispersion typically a deposition-precipitation preparation procedure is carried out in which the metal salt has time to diffuse into the pores of the support. In this way, a high dispersion is achieved by the metal being distributed over the complete surface area of the support. This provides a large active metal surface area since the formed metal crystallites are very small, typically in the order of a few nanometers. On the other hand, for the preparation of edge-coated catalysts, the metal salt should not diffuse far into the pores of the support (i.e. typically less than 100 nm into the pores for a support particle having an average size of about 20 to 100 micron), but should deposit immediately upon contact with the support. These two methods contradict each other, so the preparation of high-dispersed edge-coated metal catalysts is typically a compromise between the two factors."

EXHIBIT Q

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 10,258,967 ("*Witte*")

'864 patent	**U.S. Patent No. 10,258,967 ("*Witte*")**
	[2:7-14] "The present invention aims at producing catalysts having even smaller precious metal particle (crystallite) sizes, in particular nanoparticles that are about the same size and/or smaller than the average pore size of the support. Furthermore, the present invention aims at producing catalysts wherein the supports are edge coated with the crystallites and wherein the catalysts have a high metal loading (i.e. typically 1.0 wt. % or more PM)." [2:22-29] "Accordingly it is a first object of the present invention to provide a precious metal catalyst, in particular a palladium and/or platinum metal catalyst, which is not only both edge coated and has a high metal dispersion, but also has a high metal loading. Another object of the present invention is to provide a process for producing said catalyst. A further object of the present invention is to provide said catalyst, so that it may be used in chemical reactions." [5:45-52] "The micropore surface area of the powder support is typically between 10 and 1000 m^2/g, preferably between 20 and 750 m^2/g, and more preferably between 30 and 500 m^2/g. The micropore surface area, as used herein, is the value that can be determined from a t-plot analysis, as described in Applied Catalysis A: General 174(1998) 137-146 and determined using the suite of software programs on a Micromeritics ASAP 2040. Typically, the powder support of the catalyst of the present invention is microporous and/or mesoporous."

EXHIBIT Q

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 10,258,967 ("*Witte*")

'864 patent	U.S. Patent No. 10,258,967 ("*Witte*")
	[5:60-6:2] "Typically the Dv(90) particle size distribution of the powder support is between 1 and 500 micron, preferably between 5 and 300 micron, and more preferably between 10 and 200 micron. The Dv(90) particle size distribution is defined as meaning that 90% of the particles have a volume average particle size less than the value in the above-mentioned ranges indicated. The Dv(90) particle size distribution, as used herein, is the value that can be measured of a pre-sonicated sample mixture in water by light scattering using a Malvern Mastersizer 2000." [9:20-26] "In a further embodiment, the precious metal catalyst of the present invention, preferably a palladium and/or platinum metal catalyst, may be used generally for all chemical reactions for which precious metal catalysts are suitable. Such reactions may include isomerization, oxidation, hydrogenolysis (e.g. hydro-desulfurization), hydrogenation/dehydrogenation and hydro-dewaxing reactions." [9:64-10:2] "A 500 mL beaker was equipped with baffles, filled with 10g (dry weight) of activated carbon powder (Dv(90) of 82 micron; BET surface area of 1432 m^2/g; and micropore surface area of 171 m^2/g) and 100 mL water at room temperature (i.e. 20-25° C.) was slowly added to prevent dust formation." [10:37-42] "A 800 mL beaker was equipped with baffles, filled with 20 g (dry weight) of activated carbon powder (Dv(90) of 82 micron; BET surface area of 1432 m^2/g; and micropore surface area of 171 m^2/g) and 200 mL water at room temperature (i.e. 20-25° C.) was slowly added to prevent dust formation."

EXHIBIT Q

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 10,258,967 ("*Witte*")

'864 patent	**U.S. Patent No. 10,258,967 ("*Witte*")**
	To the extent that this limitation is not disclosed, either explicitly or inherently, by *Witte*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
1[b] silica, alumina, or mixtures thereof; and	*Witte* discloses a combustion promoter comprising silica, alumina, or mixtures thereof. *See, e.g.*, [5:22-28] "The powder support of the catalyst of the present invention may be selected from the group consisting of silica, alumina, zirconia, titanium oxide, ceria, magnesium oxide, zinc oxide, metal silicates (e.g. titanium silicates (TiS)), metal aluminates, zeolites, carbon nanotubes, carbon nanofibres, graphitic carbon and activated carbon (AC) and combinations thereof, and preferably activated carbon." To the extent that this limitation is not disclosed, either explicitly or inherently, by *Witte*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
1[c] one or more Group VIII noble metals distributed in the particle as an eggshell such that a higher concentration of the one or	*Witte* discloses a combustion promoter comprising one or more Group VIII noble metals, the metals distributed in the particle as an eggshell such that a higher concentration of the one or more Group VIII noble metals is present in the outer

EXHIBIT Q

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 10,258,967 ("*Witte*")

'864 patent	**U.S. Patent No. 10,258,967 ("*Witte*")**
more Group VIII noble metals is present in the outer region of the microsphere sized porous particle as compared to the concentration of the one or more Group VIII noble metals in the centre of the microsphere sized porous particle.	region of the microsphere sized porous particle as compared to the concentration of the one or more Group VIII noble metals in the centre of the microsphere sized porous particle. *See, e.g.*, [Abstract] "The present invention is in the field of catalysis. More particularly, the present invention is directed to supported precious metal catalysts, preferably palladium and/or platinum metal catalysts, having a high metal loading, a high degree of dispersion and a high degree of edge-coating. The present invention is further directed to a process for producing these catalysts, as well as to the use of these catalysts in chemical reactions." [1:12-58] "Many supported precious metal (PM) catalysts are used inter alia for selective conversion of fine chemicals, which are typically highly functionalized large molecules. In order for the PM crystallites to be best accessible for the large molecules, it is desirable to have them situated in the outer shell of the support as much as possible. This is referred to as an edge-coated metal distribution. Typically a high degree of edge-coating is achieved when the majority of the PM crystallites are situated in the outer shell of the support. Such an edge-coated distribution has the additional advantage that the substrate molecule rapidly leaves the vicinity of the PM crystallite after it is converted, thereby minimizing any side reactions from taking place. However, the preparation of edge-coated catalysts with a high dispersion is difficult. Since only a small portion of the total surface area of the support is used, edge-coated catalysts generally have a low dispersion. Supported PM catalysts are typically prepared by a deposition-reduction method, in which the metal is first deposited on a support and then reduced. In order to prepare a catalyst with a high dispersion typically a deposition-precipitation preparation

EXHIBIT Q

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 10,258,967 ("*Witte*")

'864 patent	U.S. Patent No. 10,258,967 ("*Witte*")
	procedure is carried out in which the metal salt has time to diffuse into the pores of the support. In this way, a high dispersion is achieved by the metal being distributed over the complete surface area of the support. This provides a large active metal surface area since the formed metal crystallites are very small, typically in the order of a few nanometers. On the other hand, for the preparation of edge-coated catalysts, the metal salt should not diffuse far into the pores of the support (i.e. typically less than 100 nm into the pores for a support particle having an average size of about 20 to 100 micron), but should deposit immediately upon contact with the support. These two methods contradict each other, so the preparation of high-dispersed edge-coated metal catalysts is typically a compromise between the two factors. Preparing a supported PM catalyst with a low metal loading (i.e. typically less than 1 wt. % PM) potentially overcomes this problem, since the small amount of PM used can be easily accommodated in the outer shell of the support while maintaining a high dispersion. However, this has the disadvantage that it will lead to a PM catalyst with a low activity per gram of material. Although increasing the precious metal loading would overcome the low activity, maintaining both the high dispersion and high edge coating with the increased precious metal loading has proven to be difficult." [2:7-14] "The present invention aims at producing catalysts having even smaller precious metal particle (crystallite) sizes, in particular nanoparticles that are about the same size and/or smaller than the average pore size of the support. Furthermore, the present invention aims at producing catalysts wherein the supports are edge coated with the crystallites and wherein the catalysts have a high metal loading (i.e. typically 1.0 wt. % or more PM)."

EXHIBIT Q

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 10,258,967 ("*Witte*")

'864 patent	U.S. Patent No. 10,258,967 ("*Witte*")
	[2:22-36] "Accordingly it is a first object of the present invention to provide a precious metal catalyst, in particular a palladium and/or platinum metal catalyst, which is not only both edge coated and has a high metal dispersion, but also has a high metal loading. Another object of the present invention is to provide a process for producing said catalyst. A further object of the present invention is to provide said catalyst, so that it may be used in chemical reactions. Surprisingly it has been found that the edge-coating and metal dispersion of the catalyst can be controlled separately. As a result, both parameters can be optimized separately to obtain highly dispersed edge coated precious metal catalysts, in particular palladium and/or platinum metal catalysts, which also have a high metal loading, according to the present invention. The present invention is accordingly directed to a precious metal catalyst, wherein said catalyst comprises nanocrystallites of a precious metal on a powder support, wherein the nanocrystallites have an average size of less than 5 nm, wherein the catalyst comprises at least 1.0 wt. % precious metal, based on the weight of the catalyst, and wherein the precious metal is palladium and/or platinum metal; wherein the palladium metal catalyst has a surface enrichment value of from at least 6.5 to at most 150; and, wherein the platinum metal catalyst has a surface enrichment value of from at least 1.5 to at most 150." [2:61-4:12] "The precious metal catalyst of the present invention typically comprises nanocrystallites of precious metal, in particular palladium and/or platinum metal,

EXHIBIT Q

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 10,258,967 ("*Witte*")

'864 patent	U.S. Patent No. 10,258,967 ("*Witte*")
	having an average size of from 1 to less than 5 nm, preferably between 1 and 4 nm, and more preferably between 1.5 and 3 nm. The average size of the precious metal nanocrystallites, as used herein, is based on the value that can be determined by Transmission Electron Microscopy (TEM), unless otherwise indicated. Typically, for every measurement at least 200 PM crystallites are counted. Typically, a palladium metal catalyst of the present invention has a surface enrichment value of at least 6.5, preferably at least 8 and more preferably at least 10. The surface enrichment value of the palladium metal catalyst is typically at most 150, preferably at most 120, and more preferably at most 90. A platinum metal catalyst of the present invention typically has a surface enrichment value of at least 1.5, preferably at least 2 and more preferably at least 4. Typically, the surface enrichment value of the platinum metal catalyst is at most 150, preferably at most 120, and more preferably at most 90. The surface enrichment value (SEY) can be determined by using X-ray photoelectron spectroscopy (XPS) and inductively coupled plasma (ICP) measurements, in particular inductively coupled plasma optical emission spectrometry (ICP-OES) measurements, and the following formula (I): SEV = (XPS wt. %–ICP wt. %)/ICP wt. % (I) wherein XPS wt. % is the XPS measurement and ICP wt. % is the ICP measurement (i.e. ICP-OES measurement) of the PM content of the catalyst according to the present invention, in weight percent (wt. %). XPS measurements are used to determine the content of the precious metal in the outer shell of the powder support

EXHIBIT Q

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 10,258,967 ("*Witte*")

'864 patent	U.S. Patent No. 10,258,967 ("*Witte*")
	(i.e. typically about 0 to 10 nm in depth from the surface of the powder support particle in weight percent (wt. %). ICP measurements (i.e. ICP-OES measurements) are used to determine the precious metal content in the catalyst powder support (i.e. a bulk measurement), in weight percent (wt. %). In the case of a perfectly uniform distribution of precious metal on the powder support the precious metal content as determined by XPS and ICP measurements (i.e. ICP-OES measurements) would be substantially the same and would result in a surface enrichment value of about zero. For an edge-coated metal distribution, the precious metal content in the outer shell of the catalyst powder support, as determined by XPS measurement, should be considerably higher than that in the powder support, as determined by ICP measurement (i.e. ICP-OES measurements). The reason that XPS measurements only give information on the outer few nanometers (i.e. typically on the outer shell of 5-10 nm of the surface of the measured material) of the measured material, is that although the X rays penetrate deep into a material, the released photoelectrons will only be measured if they have enough energy to escape from the material. The released photoelectrons can undergo inelastic collisions, recombination, excitation of the sample, recapture or trapping in various excited states within the material, all of which can reduce the number of escaping photoelectrons. These effects appear as an exponential attenuation function as the depth, increases, making the signals detected from elements at the surface much stronger than the signals detected from elements deeper below the material surface. Therefore, an XPS measurement typically only gives information on the outer few nanometers of the measured material. Unless explicitly indicated otherwise, all ICP measurements (i.e. ICP-OES measurements) of the catalysts, as used herein, are the values that can be measured on a Thermo Scientific™ iCAP™ 7400 ICP-OES Analyzer having an aerosol concentric nebulizer, using the autosampler CETAC ASX-260 and typically

'864 patent	U.S. Patent No. 10,258,967 ("*Witte*")
	calibration curves of 0-25 ppm. Two standard series typically used were of 0 ppm (blank); 5, 10 and 25 ppm of the elements Pd, Pt, Rh, and Ru, with each element having the same concentration, and with one of the series in an aqueous solution of 10 wt. % nitric acid (concentration of 65 wt. %) and the other series in an aqueous solution of 15 wt.% of H_3PO_4/H_2SO_4 (5:3) (H_3PO_4 having a concentration of 85 wt. %, and, H_2SO_4 having a concentration 98 wt. %. Typically all measurement series performed started with the standards, so as to determine the calibration curve and ensure that the correlation coefficient was greater than 0.999. The standard of 10 ppm was usually used as an internal standard during a series measurement." [4:57-5:18] "Surprisingly it has been found that the catalyst of the present invention has a high activity due to a combination of the high metal dispersion, which increases the number of active sites available to the reactants as well as the contribution of atoms at the comers or edges of the metal nanocrystallites, a high edge-coating, which leads to improved accessibility for reactants to the catalytic metal surface, and high metal loading, which leads to a high activity per gram of material. The catalyst of the present invention also surprisingly has the advantage of a high selectivity due to the edge-coating which decreases the likelihood of undesirable side reactions. A further advantage of the catalyst of the present invention is that internal heat transfer problems will not occur when it is used in an exothermic chemical reaction, because the precious metal is present mostly on the surface of the support. Therefore any heat that is generated during the chemical reaction is able to be easily transferred into the surrounding reaction mixture. A palladium metal catalyst of the present invention typically comprises palladium metal in an amount of between 1.0 and 20 wt. %, preferably between 1.5 and 15 wt. %, and more preferably between 2.0 and 10 wt. %, based on the weight of the catalyst.

EXHIBIT Q

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 10,258,967 ("*Witte*")

'864 patent	U.S. Patent No. 10,258,967 ("*Witte*")
	Typically a platinum metal catalyst of the present invention comprises platinum metal in an amount of between 1.0 and 20 wt. %, preferably between 1.5 and 15 wt. %, and more preferably between 2.0 and 10 wt. %, based on the weight of the catalyst." [6:3-39] "In an additional embodiment, the present invention is directed to a process for preparing a precious metal catalyst according to the present invention, wherein said process comprises the steps of: reducing a precious metal compound in an aqueous solution by contacting said solution with a reducing agent, a stabilizing agent and optionally a coordinating agent to form a colloidal precious metal suspension; contacting the suspension with a powder support at a pH value of between 9.5 and 11; and recovering the precious metal catalyst. The advantage of the process of the present invention is that the precious metal dispersion is set in the first step. This is done using a colloidal approach, in which the precious metal compound is reduced to precious metal crystallites in an aqueous solution in the presence of a reducing agent, a stabilizing agent and optionally a coordinating agent to form a colloidal suspension. Typically, the precious metal compound in the aqueous solution is contacted, in particular mixed, with the reducing agent, the stabilizing agent and optionally the coordinating agent. In the following step the edge coating and the precious metal loading on the powder support is set by adjusting the pH and depositing the resulting colloidal suspension onto a suitable powder sup- port by contacting the suspension, in particular by mixing, with the support to obtain the catalyst of the present invention.

EXHIBIT Q

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 10,258,967 ("*Witte*")

'864 patent	U.S. Patent No. 10,258,967 ("*Witte*")
	Preferably the precious metal is selected from platinum, palladium, iridium, rhodium, ruthenium, silver, gold and combinations thereof, and preferably palladium and/or platinum metal. Suitable precious metal compounds which may be used in the process of the present invention include water soluble salts, such as nitrates, acetates, sulfates, ammonium citrates and chlorides and combinations thereof. Preferably, the salt used is a palladium and/or platinum metal salt. More preferably, the salt used is Na_2PdCl_4 and/or H_2PtCl_6." [9:1-13] "Surprisingly it has been found that by adjusting the pH during the deposition step to a value of between 9.5 and 11, the amount of precious metal colloid deposited on the powder support can be increased, while maintaining the stability of the PM colloidal suspension. Advantageously, the deposition happens in such a way that the precious metal deposited is accessible to chemical reactants since it is deposited on the outer shell of the powder support particles, thereby ensuring an efficient access to the catalytically active surface area. Furthermore, the precious metal crystallites are typically distributed in a more uniform manner over the support surface with less agglomeration occurring when compared to a deposition carried out at a lower pH." [9:20-26] "In a further embodiment, the precious metal catalyst of the present invention, preferably a palladium and/or platinum metal catalyst, may be used generally for all chemical reactions for which precious metal catalysts are suitable. Such reactions may include isomerization, oxidation, hydrogenolysis (e.g. hydro-desulfurization), hydrogenation/dehydrogenation and hydro-dewaxing reactions."

EXHIBIT Q

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 10,258,967 ("*Witte*")

'864 patent	U.S. Patent No. 10,258,967 ("*Witte*")
	[9:44-10:15] "Example 1: Preparation of 2.43 wt. % Pt Supported on Activated Carbon Having Pt Metal Crystallites with an Average Size of 2 nm A 5 L beaker was equipped with baffles and filled with 3.6 L water at room temperature (i.e. 20-25° C.). Using a top stirrer, the water was stirred at 300 rpm. The water was heated to 95° C. The reducing/stabilizing agent (240 mL 30 wt. % Luviquat Mono CP (hydroxyethyl cetyldimonium phosphate) in water, obtained from Sigma-Aldrich) was added in one portion and the mixture was stirred for several minutes. A Pt solution (22.8 g 21.01 wt. % Pt as H_2PtCl_6 diluted to 300 mL by addition of water) was added to the mixture over a period of 10 min. After the addition of the Pt solution the pH was set to 5.0 by the addition of NaOH solution (10 wt.% NaOH in water). The mixture was stirred at 95° C. for two hours, while keeping the water level constant and while re-setting the pH to 5.0 every 30 min. After cooling, the pH of the colloidal Pt metal suspension was 4.9. A 500 mL beaker was equipped with baffles, filled with 10g (dry weight) of activated carbon powder (Dv(90) of 82 micron; BET surface area of 1432 m^2/g; and micropore surface area of 171 m^2/g) and 100 mL water at room temperature (i.e. 20-25° C.) was slowly added to prevent dust formation. Using a top stirrer, the slurry mixture was stirred at 300 rpm. By addition of NaOH solution (10 wt.% NaOH in water) the pH was set to 10.0 and the slurry mixture was stirred for one hour to obtain a homogeneous suspension. An amount of colloidal Pt suspension corresponding to 0.4 g Pt was added to this suspension over a period of 60 min. The pH was reset to 10.0 by the addition of NaOH solution (10 wt.% NaOH in water) and the slurry mixture was stirred for 1 hour. The resulting Pt/AC catalyst was filtered off and washed with water until no more Cl was found in the washing water (indicator test using $AgNO_3$ solution). TEM analysis of the catalyst showed the

'864 patent	U.S. Patent No. 10,258,967 ("*Witte*")
	formation of 2 nm Pt crystallites (see FIG. 1). ICP analysis determined that the catalyst had a Pt loading of 2.43 wt. %." [10:16-55] "Example 2: Preparation of 1.29 wt. % Pd Supported on Activated Carbon Catalyst Having Pd Metal Crystallites with an Average Size of 2 nm A 5 L beaker was equipped with baffles and filled with 3.6 L water at room temperature (i.e. 20-25° C.). Using a top stirrer, the water was stirred at 300 rpm. The reducing/stabilizing agent (200 mL 30 wt. % Luviquat Mono CP (hydroxyethyl cetyldimonium phosphate) in water, obtained from Sigma-Aldrich) and the coordinating agent (2.0 g urea) were added in one portion and the mixture was stirred for several minutes. A Pd solution (10.5g 18.98 wt. % Pd as Na_2PdCl_4 diluted to 500 mL by addition of water) was added to the mixture over a period of 30 min, during which the pH of the solution slowly dropped from 5.4 to 4.9. After the Pd addition the pH was set to 5.0 by addition of a few drops of NaOH solution (10 wt.% NaOH in water). The mixture was heated to 95° C. (pH 4.7) and kept at this temperature for two hours, while keeping the water level constant. After cooling the pH of the colloidal Pd suspension was 4.9. A 800 mL beaker was equipped with baffles, filled with 20g (dry weight) of activated carbon powder (Dv(90) of 82 micron; BET surface area of 1432 m^2/g; and micropore surface area of 171 m2/g) and 200 mL water at room temperature (i.e. 20-25° C.) was slowly added to prevent dust formation. Using a top stirrer, the slurry mixture was stirred at 500 rpm. By the addition of NaOH solution (10 wt. % in water) the pH was set to 10.0 and the mixture was stirred for one hour to obtain a homogeneous suspension. An amount of colloidal Pd suspension corresponding to 0.4 g Pd was added to this suspension over a period of 60 min. The pH was re-set to 10.0 by the addition of NaOH solution (10 wt. % NaOH in water) and the mixture

'864 patent	U.S. Patent No. 10,258,967 ("*Witte*")
	was stirred for one hour. The resulting Pd/AC catalyst was filtered off and washed with water until no more Cl was found in the washing water (indicator test using $AgNO_3$ solution). TEM analysis of the catalyst showed the formation of 2 nm Pd crystallites (see FIG. 2). ICP analysis determined that the catalyst had a Pd loading of 1.29 wt. %." [10:56-67] "Example 3 (Comparative): Preparation of 0.64 wt. % Pd Supported on Activated Carbon Having Pd Metal Crystallites with an Average Size of 2 nm, without Adjusting the pH in the Deposition Step An experiment is conducted with identical amounts as described in Experiment 2, only without the pH adjustment in the deposition step of Pd on AC. After the filtration step a darkly colored filtrate is observed. ICP analysis of the washed catalyst determined that the catalyst had a Pd loading of 0.64 wt. %."

EXHIBIT Q

Invalidity of U.S. Patent No. 11,224,864 in View of U.S. Patent No. 10,258,967 ("*Witte*")

'864 patent	**U.S. Patent No. 10,258,967 ("*Witte*")**
	[Table 1] TABLE 1 Characteristics of various catalysts Catalyst | Crystallite size (nm) | XPS (wt. %) | ICP (wt. %) | SEV Example 1 | 2* (Pt) | 53.7 | 2.43 | 21 Example 2 | 2* (Pd) | 27.4 | 1.29 | 20 Escat ™ 1421 | 4.3# (Pd) | 14.4 | 5 | 1.9 Escat ™ 1621 | 4.0# (Pd) | 17.8 | 5 | 2.6 Escat ™ 1911 | 3.0# (Pd) | 21.0 | 3 | 6.0 Escat ™ 1941 | 5.2# (Pd) | 24.1 | 5 | 3.8 Escat ™ 1971 | 4.3# (Pd) | 17.3 | 5 | 2.5 Escat ™ 2421 | 4.6# (Pt) | 9.3 | 5 | 0.9 Escat ™ 2431 | 4.5# (Pt) | 10.4 | 5 | 1.1 Escat ™ 2441 | 3.3# (Pt) | 11.1 | 5 | 1.2 F101 | 5.2# (Pt) | 8.7 | 5 | 0.7 Noblyst P2060 | 3.7# (Pt) | 8.2 | 5 | 0.6 Noblyst P2058 | 4.9# (Pt) | 11.8 | 5 | 1.4 *as determined by TEM #as determined by CO chemisorption [12:29-43] "Example 5: Semi-hydrogenation of 3-hexyn-1-ol Using a Supported Palladium Catalyst A 250 mL stainless steel autoclave was charged separately with 97 mg (dry weight) of the Pd/AC catalyst of Examples 2 and 3 and 100 g of a 4.5 wt.% solution of 3-hexyn-1-ol in 96 wt. % ethanol was added. The autoclave was closed and the mixture was heated to 30° C. with stirring. The stirring was stopped, and the air was replaced by flushing hydrogen over the mixture. After flushing the autoclave was pressurized with 3 bars of hydrogen. The stirring was resumed (1500 rpm) and the hydrogen consumption was recorded. After 90 min the stirring was stopped. The

EXHIBIT Q

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	reaction rates r1 (semi-hydrogenation) and r2 (over-hydrogenation) are calculated in mL H_2 per min from the H_2 uptake curve." To the extent that this limitation is not disclosed, either explicitly or inherently, by *Witte*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[3] The combustion promoter according to claim 1, comprising a plurality of microsphere sized porous particles comprising alumina.	*Witte* discloses a combustion promoter comprising a plurality of microsphere sized porous particles comprising alumina. *See, e.g.*, The support provided above for Claim 1[b]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Witte*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[6] The combustion promoter according to claim 1, wherein the microsphere sized porous particles collectively have an average (D50) size of between 60 and 90 microns as measured by laser diffraction.	*Witte* discloses a combustion promoter comprising microsphere sized porous particles, wherein the microsphere sized porous particles collectively have an average (D50) size of between 60 and 90 microns as measured by laser diffraction. *See, e.g.*,

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	The support provided above for Claim 1[a]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Witte*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[12] The combustion promoter according to claim 1, wherein the one or more Group VIII noble metals comprises one or more of platinum, palladium, iridium, ruthenium, and rhodium.	*Witte* discloses a combustion promoter comprising one or more Group VIII noble metals, wherein the one or more Group VIII noble metals comprises one or more of platinum, palladium, iridium, ruthenium, and rhodium. *See, e.g.*, The support provided above for Claim 1[c]. To the extent that this limitation is not disclosed, either explicitly or inherently, by *Witte*, this limitation is obvious in combination with the knowledge of one of skill in the art and/or it would have been obvious to one of skill in the art at the time of the '864 patent to combine this reference with any of the other references as charted for this claim limitation. The motivations to combine may come from the knowledge of the person of ordinary skill themselves, or from known problems and predictable solutions as embodied in these references, examples of which are discussed in the Cover Pleading.
[15] The combustion promoter according to claim 12, wherein the one or more Group VIII noble metals comprises palladium.	*Witte* discloses a combustion promoter comprising one or more Group VIII noble metals, wherein the one or more Group VIII noble metals comprises palladium. *See, e.g.*, The support provided above for Claim 1[c].

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