

UNITED STATES PATENT AND TRADEMARK OFFICE

BEFORE THE PATENT TRIAL AND APPEAL BOARD

TOPSOE, INC.,
Petitioner

v.

L'AIR LIQUIDE, SOCIÉTÉ ANONYME POUR L'ETUDE ET
L'EXPLOITATION DES PROCÉDÉS GEORGES CLAUDE,
Patent Owner

Case IPR2025-01173
&
Case IPR2025-01174

DECLARATION OF PROF. DR. HARALD KLEIN

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I, Dr. Harald Klein of Munich, Germany, declare as follows:

I. INTRODUCTION AND BACKGROUND

1. I have been retained as a technical expert by Petitioner to study and provide my independent opinions on the technology claimed in U.S. Patent No. 11,673,805 (the “’805 Patent”) and the issue of patentability of the ’805 Patent claims. I understand that this declaration supports arguments presented in two Petitions for Inter partes Review (IPR) which are being filed at the same time. A first IPR Petition (IPR2025-01173) is directed to claims 1-6, 11 and 12 with respect to a series configuration of an endothermic reforming step and an autothermal reforming step (Petition I, Grounds 1-4). A second IPR Petition (IPR2025-01174) is directed to claims 1-6, 11 and 12 with respect to a parallel configuration of an endothermic reforming step and an autothermal reforming step (Petition II, Grounds 1-3). I am competent to testify under oath, and will do so if called as a witness, as follows herein.

2. A copy of my curriculum vitae is attached as EX1004.

3. I hold the following educational degrees: an undergraduate degree from University of Stuttgart (Germany), a Master of Science in Chemical Engineering Process Engineering from University of Wisconsin-Madison (USA), Department of Chemical Engineering, a Master of Science (Dipl. Ing.) from University of Stuttgart (Germany) for Process Engineering, with

specialization in Thermal Separation Processes and Chemical Reaction Engineering, and a Ph.D. (Dr.-Ing.) from the University of Stuttgart (Germany).

4. I am currently a Professor at Technical University of Munich (TUM), where I have worked for the past 17 years. I have previously held positions as Vice Dean Department of Mechanical Engineering. I also currently hold the position of Professor for Chemical Engineering at TUM-Asia in cooperation with Singapore Institute of Technology (SIT) and Professor (Co-PI) at TUMCREATE Singapore.

5. I have spent more than 25 years working in the field of chemical engineering with extensive experience with processes and systems for steam reforming and synthesis gas (also labeled as syngas) production.

6. I spent five years working with Linde Group as Manager Process Engineering for Hydrogen and Synthesis Gas Plants with specific experience for steam reforming, physical and chemical absorption processes and cryogenic separation of synthesis gases.

7. I have engaged in technical consulting experience for engineering and petro-chemical companies in the field of chemical engineering. I have also engaged in consulting for intellectual property law firms in connection with patent matters.

8. Teaching all types of chemical engineering processes, industrial applications, and design modules, I have a detailed insight into the capabilities

and skills a B.Sc. and M.Sc. graduate shall have. Especially my process simulation courses prepare the students for their future role where different possible chemical process options need to be optimized and carried out to the most advantageous overall solution, taking into account environmental, economic and technical aspects.

9. During my work as process engineer at Linde Group in the field of hydrogen and synthesis gas processes and plants, I was trained in all the details of process design for steam reforming and partial oxidation plants. Besides process design, I also was involved in the detailed equipment design (e.g., heat exchangers, separations vessels, chemical reactors) for hydrogen and synthesis gas plants, including ammonia and methanol plants. After taking the role as department manager, responsible for 30 process engineers, I became involved in the creative design of plants implementing a range of chemical process options, including steam reforming, autothermal reforming, partial oxidation, water gas shift (WGS), absorptive CO₂ and H₂S removal, cryogenic synthesis gas separation, pressure swing adsorption (PSA), ammonia synthesis, and methanol synthesis. My knowledge of these process options was employed in the hydrogen production and synthesis gas field to implement economical and environmentally friendly plant designs.

10. My education, technical background and industry experience have given me specialized expertise in the fundamentals, technologies and industrial

applications associated with chemical processes for the synthesis, separation and production of industrial gases including hydrogen, synthesis gas and carbon dioxide.

11. I am being compensated at a rate of € 2000 Euros per day for my analysis and testimony in this matter. I am also being reimbursed for reasonable and customary expenses associated with my work and testimony in this investigation. My compensation is not contingent on the outcome of this matter or the specifics of my testimony.

II. SUMMARY OF OPINIONS

12. Based on my experience, my knowledge of the art prior to the earliest effective filing date (hereafter “critical date”) of the ’805 Patent, my analysis of prior art references, and the understanding a person of ordinary skill in the art would have had of the claim terms, in light of the specification as of that critical date, it is my opinion that certain claims of the ’805 Patent are anticipated or obvious in view of the prior art references discussed below. In forming my opinions, I understand from counsel that the critical date of the ’805 Patent is August 11, 2020. The opinions herein referencing the general knowledge of a person of ordinary skill in the art (“POSA”) refer to the general knowledge of a POSA at the time of the alleged invention claimed in the ’805 Patent, which I have been informed is the timeframe prior to the critical date.

III. INFORMATION CONSIDERED

13. In forming my opinions, I have considered the '805 Patent and its prosecution history, my own knowledge and experience, the exhibits itemized in the Exhibit List filed with the two petitions for Inter partes review (IPR) of the '805 Patent that are cited herein, as well as the other materials referenced herein. I have analyzed the following publications and materials:

Exhibit	Description
1001	U.S. Patent No. 11,673,805 to Schmidt et al.
1002	File History of U.S. Patent No. 11,673,805
1005	U.S. Patent Publication No. 2023/0119784 to Reinertsen et al. ("Reinertsen")
1006	Martin <i>et al.</i> , (2019) "Progress Update on the Allam Cycle: Commercialization of NET Power and the NET Power Demonstration Facility" by Scott Martin, et al as presented at the 14th Greenhouse Gas Control Technologies Conference (October 21st – 25th, 2018). Available at SSRN: https://ssrn.com/abstract=3366370 uploaded and downloadable April 11, 2019 ("Martin")
1007	PCT Publication WO 2014/091098 to Darde et al.
1008	Certified English Translation of Darde ("Darde")
1009	PCT Publication WO 2019/162236 to Rytter et al. ("Rytter")
1010	Cotton, B. (2019) "Clean Hydrogen. Part 1: Hydrogen from Natural Gas Through Cost Effective CO ₂ Capture", dated March 15, 2019, The Chemical Engineer website, available from https://www.thechemicalengineer.com/features/clean-hydrogen-

Exhibit	Description
	part-1-hydrogen-from-natural-gas-through-cost-effective-co2-capture/, for download, Published by Institution of Chemical Engineers (Rugby, UK) (“Cotton”)
1011	Patwardhan et al. (2013) “Optimised hydrogen production by steam reforming: part 2”. Digital Refining article 1000841, dated PTQ Q3 2013, available for download from http://www.digitalrefining.com/article/1000841 (“Patwardhan”)
1013	SSRN Cover for Martin et al. EX1006
1014	U.S. Patent Publication No. US 2009/0298957 to Gauthier et al. (“Gauthier”)
1015	U.S. Patent Publication No. 2019/0135626 to Rafati et al. (“Rafati”)
1016	Appl, M. (1997) Ammonia Methanol Hydrogen Carbon Monoxide, Modern Production Technologies, A Review; published by Nitrogen-The Journal of the World Nitrogen and Methanol Industries, an imprint of British Sulphur Publishing, a division of CRU Publishing Ltd. (London, England) (“Appl”)
1017	Aasberg-Petersen et al. (2011) “Natural gas to synthesis gas – Catalysts and catalytic processes”, J. Natural Gas Science and Engineering, 3:423-459 (“Aasberg-Petersen I”)
1018	Technology Handbook (Air Liquide, Engineering and Construction. Ed.) February 2018, version 2, available for download at https://www.scribd.com/document/386431469/Air-Liquide-Technology-Handbook-March-2018 , 108 pages (“Technology Handbook”)

Exhibit	Description
1019	Riquarts et al. (1985) “Gas separation using pressure swing absorption plants”, in Linde Reports on Science and Technology, No. 40, (LindeAktiengesellschaft), ISSN:0942-5268 (“Riquarts”)
1020	Xu et al. (2014) “An Improved CO ₂ Separation and Purification System Based on Cryogenic Separation and Distillation Theory”, Energies (ISSN 1996-1073), 7, 3484-3502; MDPI (Basel, CH) doi:10.3390/en7053484 (“Xu”)
1021	Keshavarz et al. (2019) “Cryogenic CO ₂ Capture”, in Sustainable Agriculture Reviews 38, (Inamuddin et al., Eds.), ISBN 978-3-030-29337-6 (eBook), Springer Nature Switzerland AG (“Keshavarz”)
1022	File History of UK application 2003317.1, filed March 6, 2020, priority document for U.S. Patent Publication No. 2023/0119784 to Reinertsen et al.
1023	File History of UK application 2010174.7, filed March 6, 2020, priority document for U.S. Patent Publication No. 2023/0119784 to Reinertsen et al.
1024	U.S. Patent Application No. 2012/0291484 to Terrien et al. (“Terrien”)
1026	K. Aasberg-Petersen et al. (2004) “Synthesis gas production for FT synthesis”, Chapter 4 in Studies in Surface Science and Catalysis, vol 152, pages 258-405 (“Aasberg-Petersen II”)
1027	J. R. Rostrup-Nielsen (1993) “Production of Synthesis Gas”, Catalysis Today, volume 18, pages 305-324 and volume 71, pages 243-247, 2002
1028	J. R. Rostrup-Nielsen (2002) “Syngas in perspective”, Catalysis Today, volume, pages 243-247

Exhibit	Description
1029	(Reserved)

IV. LEGAL STANDARDS

14. I have been informed that the following legal principles apply to analysis of patentability based on 35 U.S.C. §§102 and 103.

A. Level of Ordinary Skill in the Art

15. It is my understanding that the claims and specification of a patent are to be read and construed through the eyes of a POSA at the time the invention was made, which I have been informed is the timeframe prior to August 11, 2020 (critical date). To determine the appropriate level of one of ordinary skill in the art, I have been informed that the following factors may be considered: (a) the types of problems encountered by those working in the field and prior art solutions thereto; (b) the sophistication of the technology in question, and the rapidity with which innovations occur in the field; and (c) the educational level and experience of active workers in the field.

B. Claim Construction

16. I have also been informed that, in order to assess whether the prior art satisfies a certain claim element in an invalidity analysis, the claim element must first be construed, and then the prior art must be evaluated to determine whether it satisfies the properly-construed element of the claim. I have been

informed that a patent claim is construed from the perspective of a POSA at the time of the invention, taking into account the claim language, the teachings of the patent specification and the prosecution history.

17. Other than as specifically noted below, I have been informed that the claims in the '805 Patent should be understood based on the plain and ordinary meaning of the claim language in the context of the '805 Patent. Unless otherwise specifically noted, my opinions in this Declaration are consistent with the plain and ordinary meaning of the claim language to a POSA at the time of the invention.

C. Anticipation

18. I have been informed that a patent claim is invalid as being anticipated if each limitation of the claim is disclosed explicitly or inherently in a single prior art reference. I further understand that a limitation is disclosed inherently if it is necessarily present in the prior art reference or is the natural result of the combination of elements explicitly disclosed by the prior art.

D. Obviousness

19. I have been informed that a patent claim is invalid as being obvious if the differences between the claimed subject matter and the prior art are such that the claimed subject matter as a whole would have been obvious at the time the invention was made to a POSA, which I have been informed is the timeframe prior to August 11, 2020 (critical date). I also understand that in

order to assess whether a claim would have been obvious in light of the prior art, I must step back in time and take the viewpoint of the “hypothetical person of ordinary skill in the art” when the invention was unknown and just before it was made, which I understand is the timeframe prior to August 11, 2020 (critical date) in the present case. I must then make a determination whether the claimed invention “as a whole” would have been obvious at that time to that hypothetical person.

20. I have been informed that the following factors are considered in determining whether a claimed invention is invalid as obvious over the prior art: (1) the scope and content of the prior art; (2) the level of ordinary skill in the art; (3) the differences between the claimed invention and the prior art and (4) any “secondary considerations” that may serve as evidence of nonobviousness, such as unexpected benefits, commercial success, long felt but unsolved needs, and failure of others.

21. I have been informed that a POSA is presumed to know all relevant prior art. I have been informed that a reference may be modified or combined with other references or with the POSA’s own knowledge if the person would have found the modification or combination obvious. In considering whether a claim is obvious, I understand that I can take account of inferences and creative steps that a POSA would employ, and the common knowledge of the art that such a person would possess.

22. I have been told that there is no rigid rule that a reference or combination of references must contain a “teaching, suggestion, or motivation” to combine references. However, I also have been informed that the “teaching, suggestion, or motivation” test can be a useful guide in establishing a rationale for combining elements of the prior art. I have been informed that this test poses the question as to whether there is an express or implied teaching, suggestion or motivation to combine prior art elements in a way that realizes the claimed invention, and that it seeks to counter impermissible hindsight analysis.

23. I have also been informed that the combination of familiar elements disclosed in the prior art according to known methods is likely to be obvious when it does no more than yield predictable results. I understand that if a technique has been used to improve one product, and a POSA would have recognized that it would improve similar products in the same way; using the technique is obvious unless its actual application is beyond the skill of a person of ordinary skill in the art. I have been informed that the proper analysis of obviousness determines whether a POSA would have a “reasonable expectation of success” - not an “absolute predictability” of success - in achieving the claimed invention by combining prior art references.

24. I have been informed that, while there is no requirement that the prior art contain an express suggestion to combine known elements to achieve the claimed invention, a suggestion to combine known elements to achieve the

claimed invention may come from the prior art as a whole or individually, alone or in combination with the knowledge of a POSA. A POSA often will be able to fit the teachings of multiple references together using their own knowledge and experience. Any need or problem known in the field and addressed by the reference can provide a reason for combining the elements in the manner claimed.

25. I have also been informed that if a POSA can implement a predictable variation and would see the benefit of doing so, that variation is likely to be obvious. I have been told that in many fields, there may be little discussion of obvious combinations, and in these fields technical or market demands may drive design trends. I have been told that, when there is a design need or market pressure, and there are a finite number of predictable solutions, a person of ordinary skill in the art has good reason to pursue those known options.

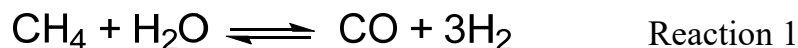
V. THE FIELD OF TECHNOLOGY AND STATE OF THE ART

26. I understand the '805 Patent is an issued U.S. patent which claims priority to European application 20020366 ("the '366 priority application"), filed on August 11, 2020, the critical date.

27. The '805 Patent relates to a process and plant for preparation of hydrogen from natural gas. The process described includes generation of synthesis gas and shifting of synthesis gas followed by separation of hydrogen

from the shifted synthesis gas and separation of CO₂ from hydrogen-depleted residual gas from the hydrogen separation. Gases produced in the component steps of the process can be further separated/purified and optionally recycled as feed or fuel to process units.

28. Synthesis gas is primarily a mixture of hydrogen (H₂) and carbon monoxide (CO), most often generated by reforming of hydrocarbons, particularly of natural gas/methane. Steam reforming of methane includes the following reaction:



This is an endothermic reaction requiring heat to proceed. The reaction is reversible and the extent to which CO and H₂ are formed depends upon reaction conditions. Steam reforming does not go to completion, as the reaction is in or close to equilibrium, such that at least some level of methane and water will remain in the synthesis gas produced. In a traditional steam reformer (SMR), the required heat for steam reforming is supplied from an externally fired furnace mainly by radiative heat transfer. The required heat for steam reforming in other types of units can be supplied for example by heat exchange with another process gas. It is known by a POSA that steam reforming is almost always accompanied by the water-gas shift reaction shown in Reaction 3. This reaction does not go to completion but will be at or close to equilibrium. The exit gas

from a steam reforming unit therefore typically contains CO, CO₂, H₂, CH₄, and steam.

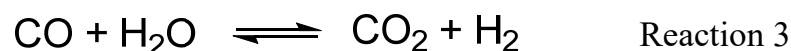
29. Synthesis gas can also be produced by technologies where oxygen is part of the feed. Such technologies are sometimes called “oxygen aided reforming” or “partial oxidation”. Technologies that could fall under this category include non-catalytic partial oxidation (often known as POX), catalytic partial oxidation (often known as CPO or CPOX), and autothermal reforming (mostly known as ATR). The partial oxidation is often expressed as:



This is an exothermic reaction which generates heat. It will be recognized by a POSA that Reaction 2 is a very simplified way of describing the partial oxidation technologies and in reality, a number of other reactions occur. For example, some full oxidation of methane to carbon dioxide and water (steam) also occurs. Furthermore, part of the hydrogen (e.g. generated from Reaction 2), is fully oxidized to water. Also, part of the CO (e.g. from Reaction 2) reacts with oxygen to form CO₂. Other side reactions, like the full and partial oxidation of higher hydrocarbons (if present in the feed) and catalytic or non-catalytic steam reforming (Reaction 1) and water gas shift (Reaction 3) take place (all these reactions are not shown here). The full oxidation reactions of methane to CO₂ and hydrogen to water are both highly exothermic and a high amount of energy is generated by these reactions. Thus, by the various oxidation

reactions (such as partial and full oxidation of methane and higher hydrocarbons, oxidation of hydrogen, oxidation of CO), all oxygen which is supplied is consumed.

30. Synthesis gas generated from steam reforming and/or partial oxidation can be further processed with the water-gas shift reaction to convert a major portion of the CO in the synthesis gas on reaction with H₂O to more hydrogen:



The water-gas shift (WGS) reaction does not go to completion, because the reaction is in equilibrium, such that at least some level of CO and water will remain in the shifted gas. Shifted synthesis gas thus primarily contains CO₂ and H₂ and unreacted CO. Again, shifted synthesis gas also contains unreacted CH₄ and residual H₂O (unreacted CH₄ originates from an upstream reforming step).

31. Generation of synthesis gas by steam reforming and partial oxidation of hydrocarbons, including methane, as well as the shifting (WGS) of synthesis gas to increase the H₂ and CO₂ of the product gases, was well-known to a POSA long before the critical date (*see*: Appl, EX1016). Furthermore, combination of steam reforming and autothermal reforming was known to a POSA before the critical date (*see*: Appl, EX1016).

32. Various reformer process configurations were well-known to a POSA prior to the critical date. Aasberg-Petersen I and II, EX1017 and

EX1026. Steam methane reforming (SMR) and autothermal reforming are specifically mentioned in the '805 Patent and were well-known to a POSA prior to the critical date.

33. An autothermal reformer (ATR) combines combustion and catalysis to generate synthesis gas. An exemplary ATR is illustrated below (EX1026, Figure 55):

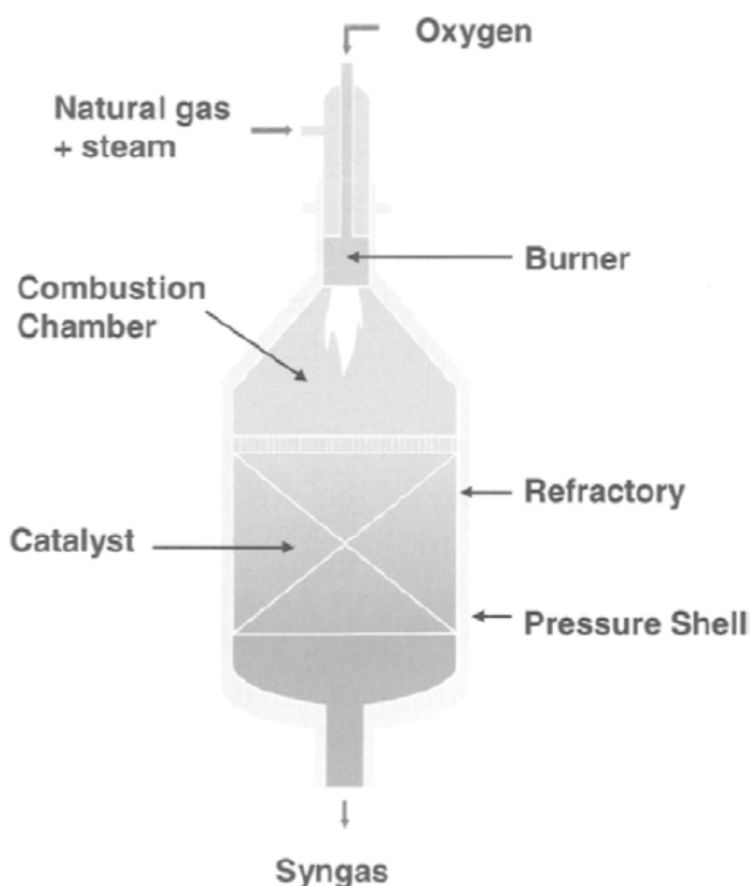


Figure 55

34. The ATR has a burner, a combustion chamber, and a catalyst bed within a refractory lined pressure shell. A feed gas containing natural gas (methane) and steam is introduced into the ATR along with oxygen. The feed

gas is partially converted in the combustion chamber where partial oxidation (Reaction 2) and other reactions occur, as described in ¶¶28–30, *supra*. Oxidation of hydrocarbons to CO₂ as well as oxidation of generated H₂ to H₂O also occur. Furthermore, some non-catalytic steam reforming (Reaction 1) as well as non-catalytic water-gas shift (Reaction 3) occur in the combustion chamber. All oxygen supplied to the ATR is converted in the combustion chamber. Additional hydrocarbon conversion occurs in the catalyst bed including steam methane reforming (Reaction 1) and water-gas shift (Reaction 3). These reactions will at the exit of the ATR normally be at or close to equilibrium and the exit gas therefore contains CO, H₂, CO₂, steam, and residual CH₄.

35. An autothermal reformer is often combined with a fired heater to preheat feed prior to entry into the ATR, as shown below (EX1026, Figure 12):

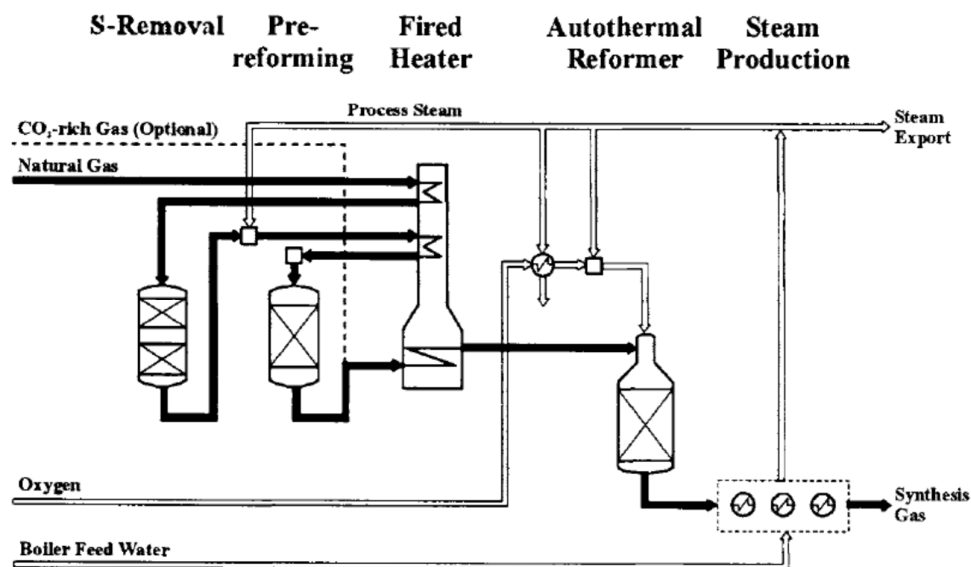
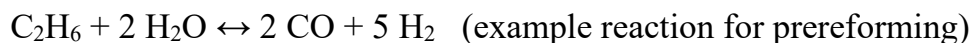


Figure 12

The use of the fired heater may increase process efficiency and can reduce the flow rate of oxygen when it is required to achieve a specified temperature at the outlet of the ATR. A POSA is aware of the fact that the oxygen flow rate determines the temperature of the gas mixture at the outlet of the combustion chamber. Figure 12 also illustrates the use of prereforming in this case upstream of an ATR. Prereforming converts higher hydrocarbons (e.g., ethane, propane, ...) present in, for example natural gas, according to the below reaction (shown for ethane):



In a prereformer (which in most cases is adiabatic in a temperature range of 400 to 500°C) all or essentially all of the higher hydrocarbons are converted. The prereformer is a catalytic reactor and Reactions 1 and 3 also take place, and are typically at or close to equilibrium at the outlet of the reactor. The use of (adiabatic) prereforming upstream of steam reformers and autothermal reformers was well known to a POSA prior to the critical date.

36. In addition, heat exchange reformers were well-known to a POSA prior to the critical date (*see*, EX1017). A heat exchange reformer is a steam reformer where the heat required for the endothermic reforming reaction is provided by convective heat transfer from a flue gas or a process gas. Three

exemplary heat exchange reformers (EX1017, p.14, Figure 23) are illustrated below:

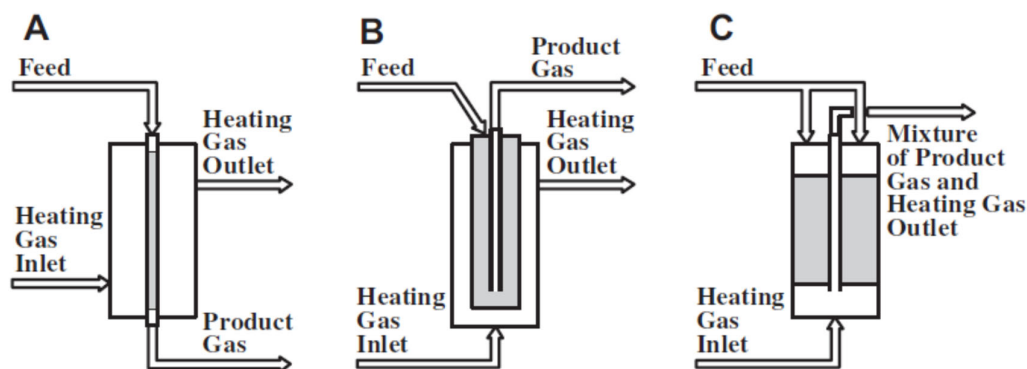


Fig. 23

Configuration A has a straight through tube, product gas is not mixed with heating gas and heating gas can be flue gas or process gas from another reformer. Configuration B has a bayonet tube, product gas is not mixed with heating gas and heating gas can be flue gas or process gas from another reformer. Additionally, in the bayonet tube the product gas is cooled and thereby also contributes to the heat supply to the reforming reaction.

Configuration C mixes heating gas and product gas. Heating gas is process gas from another reformer. Heat exchange reformers heated by process gas are in some cases called Gas Heated Reformers (GHR) (EX1017, p.14, 3.5.1.2) and are always combined with a second reformer to provide the heated process gas which transfers heat to the GHR. A GHR typically includes a catalyst bed to support the necessary reforming reactions by ensuring that the reactions occur at practical temperatures and pressures, thereby promoting the conversion of

hydrocarbons. Such combinations of a GHR and a second reformer can be called dual reformers and share the feature of heat exchange from the second reformer to provide heat to the endothermic process of Reaction 1 in the GHR. In particular, the second reformer from which heat is exchanged can be an ATR.

37. At least two dual heat exchange reformer arrangements were known in the art prior to the critical date: a series arrangement and a parallel arrangement. EX1017, p.15, 3.5.2.1 and 3.5.2.2; EX1026, p.43–47. Both can include a GHR combined with an ATR. In the series arrangement, the feed gas (FG) passes first through a GHR and then through an ATR. Product gas from the ATR supplies heat to the GHR. An exemplary series arrangement of an ATR and GHR is illustrated below (EX1026, p.44, Figure 31):

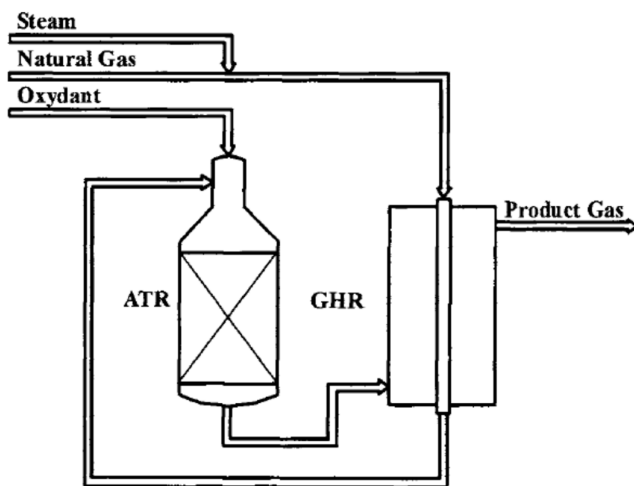


Figure 31

38. In parallel arrangements, feed gas (FG) is split into two streams, one going through a GHR and the other through an ATR. Heat is transferred from the ATR to the GHR, for example, from the hot outlet gas of the second

reformer, and the separate product gases, for example, can be mixed, if desired, downstream of the reformers described as a ‘two-in, two-out’ configuration (EX1026, p.46, Figure 34):

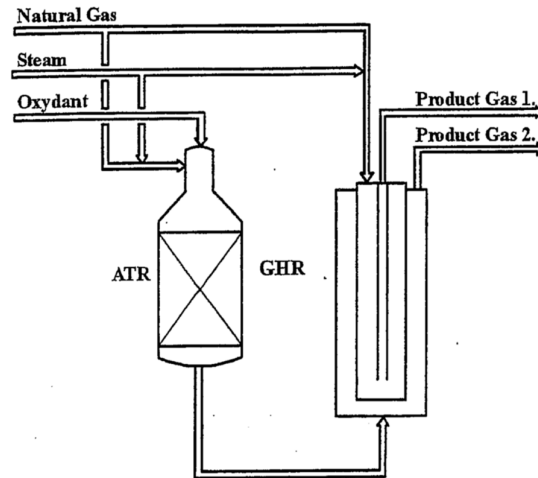


Figure 34

Alternatively, reformer product gases are mixed within the GHR and combined product gas exits the GHR described as ‘two-in, one-out’ (EX1026, p.47,

Figure 36):

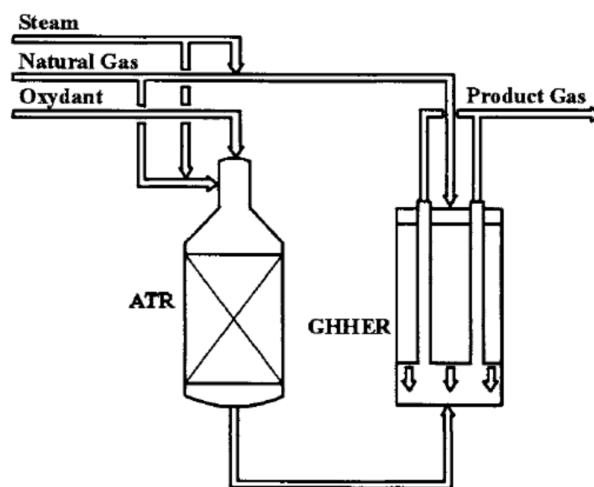


Figure 36

39. In the '805 Patent, H_2 in the shifted synthesis gas is separated by pressure swing absorption (PSA) resulting in a first hydrogen-rich gas stream (HG1) and a residual gas stream comprising CO_2 (RG1). It was well-known to a POSA prior to the critical date to employ pressure swing adsorption (PSA) for hydrogen purification from various hydrogen containing PSA gases. A POSA is well aware that these hydrogen containing PSA feed gases can originate from various upstream process units, including steam methane reformer (SMR) with downstream water-gas shift (WGS), autothermal reformer (ATR) with downstream WGS, partial oxidation with downstream WGS. EX1018, p.38; EX1019, entire document.

40. In the '805 Patent, CO_2 in RG1 (exiting the PSA) is separated by cryogenic CO_2 separation. It was well-known to a POSA prior to the critical date to employ a cryogenic process to separate CO_2 from gases. Xu, EX1020, p.2, third paragraph “(d)uring the cryogenic separation process, the components of gas mixtures are separated by a series of compression, refrigeration and separation steps.” Thus, cryogenic CO_2 separation was known in the art to include at least one compression step and at least one cooling step. Keshavarz et al., EX1021, Figure 10.3, provides an example of cryogenic CO_2 separation employing gas compression and refrigeration (cooling).

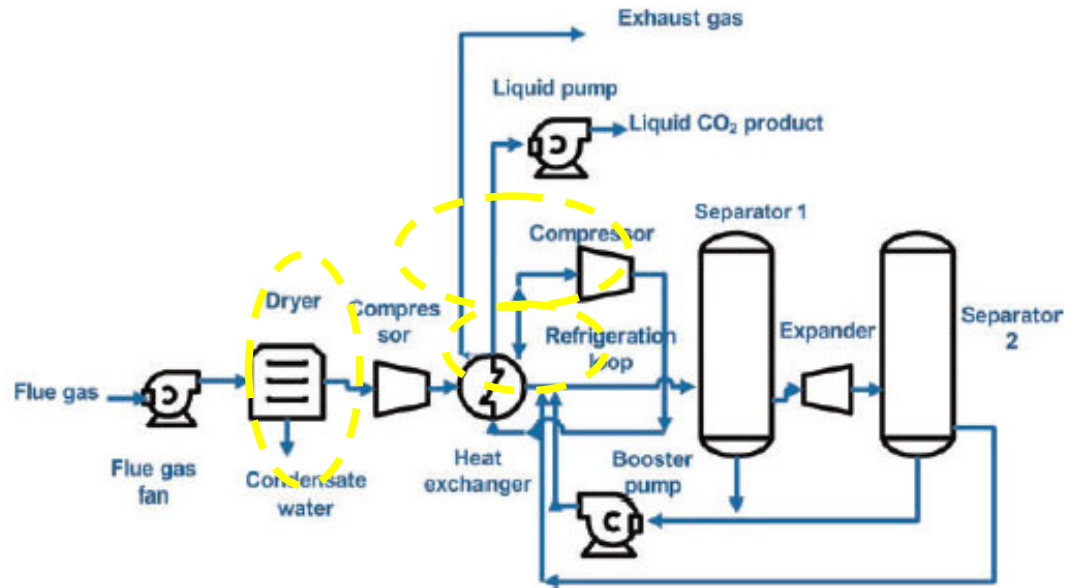


Fig. 10.3, annotated.

It was also well known to a POSA prior to the critical date to employ cryogenic CO₂ separation to separate CO₂ from gases generated during hydrogen and syngas generation. EX1018, p.23-24. In addition, it was well-known to a POSA prior to the critical date to employ cryogenic CO₂ separation on a residual gas stream exiting a PSA hydrogen separation. EX1018, p.24.

41. In the '805 Patent, H₂ in residual gas from cryogenic CO₂ separation is separated using a membrane separation process. It was well-known to a POSA prior to the critical date to generally employ hydrogen selective membranes for H₂ separation. EX1018, p.42. Also, it was well-known to a POSA prior to the critical date in particular to separate H₂ from a residual gas stream exiting cryogenic CO₂ separation. For example, in Cryocap™ H₂-Cryogenic CO₂ separation (L'Air Liquide, France) noncondensed gases from

cryogenic separation “are recycled through a membrane system to recover H₂”.

EX1018, p.24.

VI. THE '805 PATENT (EX1001)

A. Overview of the '805 Patent

42. The '805 Patent is titled “Process and Plant for Preparation of Hydrogen and Separation of Carbon Dioxide” and generally relates to a process for making hydrogen from hydrocarbons, particularly methane, where specific emission levels of CO₂ are purportedly reduced. EX1001, col.2, ln.34-37. The process includes reforming a hydrocarbon feed gas, e.g., natural gas or methane, to form carbon monoxide, carbon dioxide and hydrogen. EX1001, col.4, ln.52–57. The process also uses a water gas shift process to convert carbon monoxide into carbon dioxide and additional hydrogen. EX1001, col.5, ln.4–9. Hydrogen is separated/purified using a pressure swing adsorption process (PSA). EX1001, col.11, ln.65 to col.12, ln.7. Carbon dioxide is separated from residual product gas from the PSA using cryogenic carbon dioxide separation. EX1001, col.5, ln.34-37.

43. The '805 Patent characterizes the process shown in FIG. 1 (annotated below) as being prior art. EX1001, col.9, ln.26–27. This is consistent with my own independent understanding of the prior art.

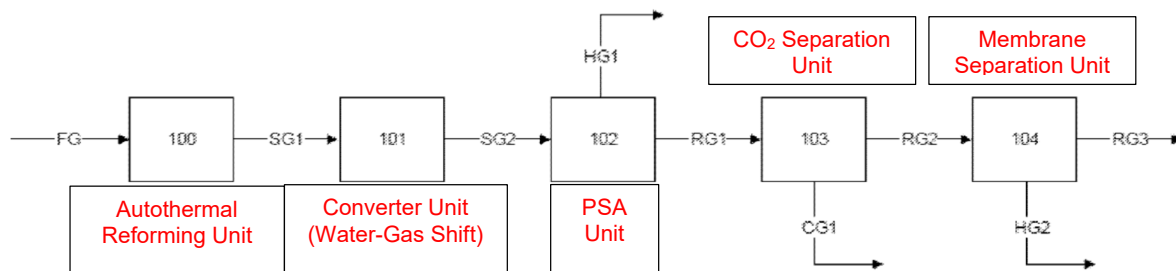
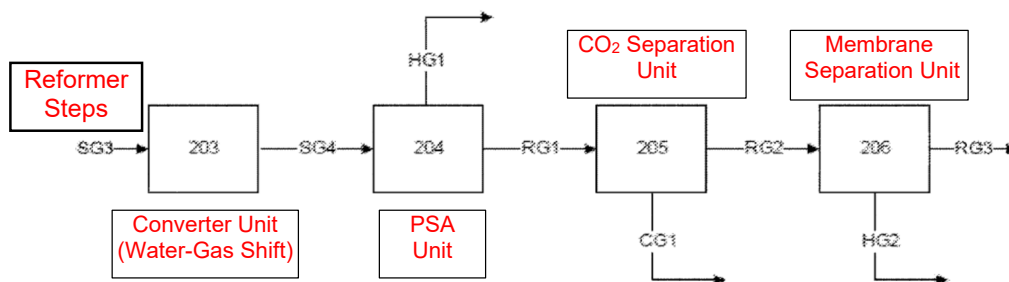


FIG. 1 (Annotated excerpt)

The prior art process of FIG. 1 as described in the '805 Patent combines an ATR step (**100**) and a WGS conversion step (**101**) to generate hydrogen enriched synthesis gas (SG2), i.e., shifted synthesis gas. As shown in the prior art process of FIG. 1, hydrogen is separated from hydrogen enriched synthesis gas (SG2) by a PSA process (**102**) to produce a first hydrogen rich gas stream (HG1) and a first residual gas (RG1). EX1001, col.9, ln.42–col.10, ln.15. The prior art process of FIG. 1 separates carbon dioxide in separation unit **103** from RG1 to produce carbon dioxide enriched gas (CG1) and a second residual gas stream RG2. EX1001, col.10, ln.6–26. An additional hydrogen separation of RG2 is also provided in FIG. 1, shown as using a hydrogen selective membrane (**104**), to produce hydrogen-rich stream HG2 and residual gas stream RG3. EX1001, col.10, ln.26–38.

44. FIG. 2 and FIG. 3 of the '805 Patent show processes characterized as the invention with a different reforming process that is included with the same downstream processes as in the prior art process of FIG. 1. Therefore, the process steps of FIG. 2 and FIG. 3, after the reforming step to produce synthesis

gas (SG3), are identical to the steps of the prior art process of FIG. 1. This downstream configuration of the FIG. 2 and FIG. 3 processes, identical to the prior art process of FIG. 1, are shown in the excerpt of these figures as annotated below:



FIGs 2 and 3 (annotated excerpt)

In FIG. 2 and FIG. 3, the stream SG3 relates to SG1 in FIG. 1. The process steps of FIG. 2 and FIG. 3 after reforming to produce SG3, therefore, are the same as those in the prior art process and include a Converter Unit (203) to carry out water gas shift reaction to produce hydrogen-enriched synthesis gas (SG4, shifted synthesis gas), a PSA Unit (204) to separate hydrogen producing hydrogen stream (HG1), a CO₂ Separation Unit (205), which carries out cryogenic CO₂ separation, to generate carbon dioxide enriched stream (CG1) and a Membrane Unit (206, hydrogen selective membrane) to separate additional hydrogen rich stream (HG2) from residual gas (RG2), thereby also generating residual gas (RG3). EX1001, col.12, ln.1–15 and ln.20–29; col.13, ln.22–24.

45. The processes shown in FIG. 3 and FIG. 2 of the '805 Patent, provide two alternative reformer configurations each of which includes an endothermic reforming step and an autothermal reforming step: a series configuration (A) and a parallel configuration (B), shown below in annotated excerpts from FIG. 3 (series configuration) and FIG. 2 (parallel configuration), respectively. In both series and parallel reformer configurations, heat generated in the autothermal reforming reaction is utilized as heat (element **202**) for the endothermic reforming step.

Series Configuration:

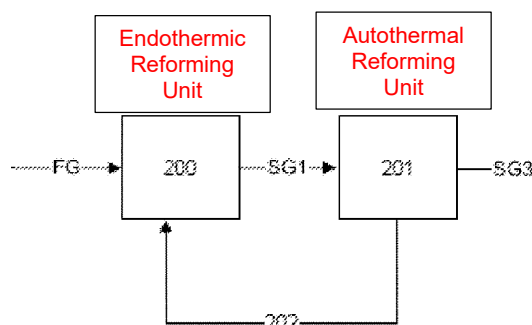
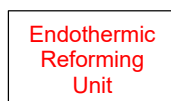


FIG. 3 (annotated excerpt)

Parallel Configuration:



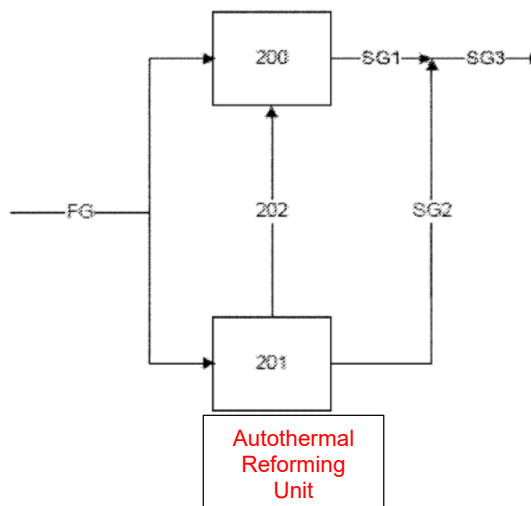


FIG. 2 (annotated excerpt)

46. In the series configuration (FIG. 3, excerpt), FG containing steam and hydrocarbon first enters the endothermic reforming step to generate a first synthesis gas stream (SG1) which is then provided as input to the autothermal reforming step. The series reformer configuration produces synthesis gas stream SG3 which is shown as exiting the autothermal reforming step. EX1001, col.4, ln.27–44. In the parallel configuration (FIG. 2 excerpt), FG is split into two portions and sent separately to the endothermic reforming step and the autothermal reforming step. In the parallel configuration, the synthesis gas streams SG1 and SG2 result from endothermic reforming and autothermal reforming steps, respectively, and are shown as combined external to the reforming units to form synthesis gas stream SG3. EX1001, col. 4, ln.16–23. I note that FIGs. 2 and 3 are both described in the '805 Patent as “a highly simplified block flow diagram” (EX1001, col.11, ln.14–15 and col.12, ln.65–

67) and, therefore, in my assessment these figures only conceptually illustrate the process described without illustrating the specific detailed flow of gas streams in the processes. FIG. 2 shows separate synthesis gas streams, SG1 and SG2, exiting the endothermic step and the autothermal step, respectively, and illustrates heat flow **202** (EX1001, col.11, ln.28–31) separate from gas flow from the autothermal step to the endothermic step. FIG. 3 shows SG1 exiting the endothermic step and entering the autothermal step and SG3 exiting the autothermal step and again illustrates heat flow **202** (EX1001, col.13, ln.13–17) separate from gas flow from the autothermal step to the endothermic step. The schematics of FIGs. 2 and 3 imply some form of indirect heat transfer from the autothermal step to the endothermic step with a heat transport medium. However, the gas streams SG2 and SG3 are stated to be preferably used directly for heating in the endothermic reforming step. EX1001, col.11, ln.31–38; col.13. ln.17–22. In my opinion, direct heating, described as preferred in the '805 Patent requires that gas streams SG2 and SG3 be introduced to the endothermic reforming step **200** to accomplish direct heat transfer without any further heat transport medium. As a consequence, the flow of these gases must both exit the endothermic reforming step which is consistent with known series and parallel dual reformer configurations discussed in ¶¶37-38, *supra* and understood by a POSA at the critical date. While some form of indirect heat transfer might be used, direct heat transfer by passing hot product gas into the

endothermic reforming step would in my opinion be the most practical and preferred method to achieve the heat transfer as described in the '805 Patent. I also note that I can find no specific description of indirect heat transfer with a heat transport medium in the '805 Patent. I interpret the illustration of gas flow in FIGs. 2 and 3 as schematic showing the principle of heat transfer to the endothermic step, but not showing the details of actual gas flows.

47. The '805 Patent states that it is an object of the invention to reduce carbon dioxide emissions from certain processes which in the context of the patent are processes used to generate hydrogen by reforming hydrocarbons with steam, e.g., autothermal reforming of methane. EX1001, col. 2, ln.34–37. I interpret, reduction of carbon dioxide emissions in these statements to refer to reduction of carbon dioxide emissions to the atmosphere, which I consider to be the conventional meaning of “carbon emissions” as understood by a POSA as of the critical date. The '805 Patent, however, expresses a more specific object “to reduce the amount of carbon dioxide formed per unit weight of hydrogen produced.” EX1001, col. 2, ln.31–33. Apparently related to this object, the '805 Patent defines “specific carbon dioxide emissions” as “the mass of CO₂ formed throughout the process per standard cubic meter of hydrogen produced.” EX1001, col.3, ln.44–47. This definition is different from my understanding of the conventional definition of “carbon emissions.” I interpret this definition, and in my view a POSA as of the critical date would interpret this definition, as

including CO₂ being generated within the process (e.g., produced via the process steps of steam reforming, partial oxidation and water-gas shift reaction and any other sources of CO₂ generation required for the process). Based on this definition, “specific carbon dioxide emissions” does not provide a specific measure of carbon dioxide emitted to the atmosphere by the process as some of the carbon dioxide formed will be captured (e.g., cryogenic CO₂ separation) as opposed to being emitted to the atmosphere. It is now and was at the critical date known by a POSA that hydrocarbon reforming with steam typically generates additional CO₂ in several heat generating firing units, often labelled as fired heaters, for example, used to superheat steam or to preheat feed gas going to the reformers. In my opinion, it is unclear if any CO₂ generation from external heaters has been included in the “specific carbon emissions” reported in the ’805 Patent.

48. The ’805 Patent reports simulations for specific carbon dioxide emission of 0.128 kg of CO₂ per standard cubic meter (m³ (STP)) of hydrogen produced and 0.118 kg of CO₂ per standard cubic meter (m³ (STP)) of hydrogen produced for the parallel configuration and the series configuration, respectively. EX1001, col.12, ln.59–62; col.13, ln.50–53. The ’805 Patent reports specific carbon emission from a process employing a single autothermal reforming step (Table 1) of 0.140 kg of CO₂ per standard cubic meter (m³ (STP)) of hydrogen produced. EX1001, col.11, ln.1–4. A comparison of specific

carbon emission simulations for the “prior art” process of Table 1 (ATR alone) with those of the processes of Table 2 and Table 3, gives reductions in specific carbon dioxide emissions relative to a process using only a single autothermal reforming step of 9% and 15% for the parallel configuration and series configuration using the dual reforming steps, respectively. EX1001, col. 12, lines 62–64; col. 13 lines 53–55. Based on these simulations, the ’805 Patent asserts that the series and parallel combinations of the endothermic reforming step and the autothermal reforming step “surprisingly significantly lowers the specific carbon dioxide emissions with regard to the overall process.” EX1001, col.3, lines 42–49. In my opinion, the data presented do not support this assertion.

49. Review of the simulation values given in Table 3 of the ’805 Patent, which are used to calculate specific carbon dioxide emissions, shows a clear error in that the mass balance values for gas streams RG1 and RG2 are indicated to be the same. This is clearly not possible because RG2 results from removal of carbon dioxide from RG1. While this may be a clerical error, a more important inconsistency in the simulation results of the ’805 Patent is present. The overall component mass balances in both Tables 2 and 3 are incorrect. For example, from the values in Table 3 (series configuration), elemental carbon input in FG is calculated to be 2649 kmol/h (based on molar flow rate and mol% given in Table 3), while elemental carbon output (HG1 + CG1 + HG2

+RG3) is 3207 kmol/h (based on combined molar flow rates and mol% given in Table 3). Using the same calculation for the values in Table 2 (parallel configuration), elemental carbon input in the FG is calculated to be 3374 kmol/h, while elemental carbon output (HG1 + CG1 + HG2 +RG3) is 4094 kmol/h. The carbon output in Table 2 and Table 3 is about 20% higher than the carbon input. As no carbon is added to the process other than in FG, carbon output higher than carbon input indicates significant discrepancies and clear errors in the values given in Table 2 and Table 3, and as such it is uncertain that the calculation of specific carbon emissions based on these values is accurate or reliable. Additionally, I find that the data in Table 1 used for the comparison do not have the same discrepancy in mass balance. This suggests to me that the processes compared in Table 1 and Tables 2 and 3 have differences in addition to the replacement of an ATR with an endothermic reforming step and an ATR with heat transfer. I also note that there are likely differences in external heating requirements between the prior art process of Table 1 and the processes of Tables 2 and 3, at least because of the different reforming steps employed. It is unclear to me based on the description given in the '805 Patent that the comparative values of specific carbon emissions used to support "surprisingly significant" improvement result from a valid comparison including such required heating. The data given in Table 1, Table 2 and Table 3, and calculations based on these data, as well as the uncertainty of what sources

of CO₂ are or are not included in the calculations, do not in my opinion, provide credible support for the assertions in the '805 Patent that a combination of an endothermic reforming step and an autothermal reforming step provides a surprising lowering of specific carbon dioxide emissions. EX1001, col. 3, ln.42–44. For these reasons, the simulations reported in the '805 Patent do not, in my opinion, allow for any meaningful comparison with the prior art and fail to provide technical support for the reported reductions in specific carbon emissions.

B. The Prosecution History of the '805 Patent (EX1002)

50. It is my understanding that the application leading to the '805 Patent (U.S. application no. 17/399,277 (“the '277 application”)), was filed on August 11, 2021, and claimed priority to European application 20020366 (“the '366 priority application”), filed on August 11, 2020 (the critical date). The '277 application originally presented 13 claims directed to a process for preparing hydrogen by reforming hydrocarbons with steam (claims 1-11) and a plant for preparation of hydrogen by reforming hydrocarbons with steam (claims 12-13).

51. A first Office Action was issued on October 12, 2022, allowing claims 1-12 and rejecting claim 13 as indefinite. EX1002, p.133–137. The Patent Office provided the following examiner's statement of reasons for

allowance of claims 1-12 citing to US 20150321914 (corresponding to EX1007 and EX1008):

Regarding claims 1, 12 and 13, US 20150321914 teaches a method of converting hydrocarbons (Abstracts). However, there is no teaching or suggestion regarding a first endothermic reforming unit configured to be heated by a second autothermal reforming unit nor is there for a plant comprising same. (Office Action of October 12, 2022, EX1002, p. 136)

52. The applicant filed a response dated January 12, 2023 cancelling claim 13. EX1002, p.155–159. A Notice of allowance issued on February 3, 2023 allowing claims 1-12. EX1002, p.162–166. The '277 application issued as the '805 Patent (EX1001) on June 13, 2023.

C. The Challenged Claims of the '805 Patent

53. The '805 Patent includes 12 claims. In this Declaration, I have assessed the validity of claims 1, 2, 3, 4, 5, 6, 11 and 12 (the “Challenged Claims”), whose limitations are listed in the chart below:

<i>Reference</i>	<i>Claim 1</i>
1.pre	A process for preparing hydrogen by reforming hydrocarbons with steam, and for separation of carbon dioxide, comprising:
1.1	(a) providing a feed gas stream,
1.1.1	wherein the feed gas stream comprises a hydrocarbon component and steam,
1.1.2	wherein the hydrocarbon component comprises at least methane;

1.2	(b) reforming at least a portion of the feed gas stream in an endothermic reforming step over a reforming catalyst thereby producing a first synthesis gas stream,
1.2.1	wherein the first synthesis gas stream comprises hydrogen, carbon monoxide, carbon dioxide, and unreacted methane;
1.3	(c) reforming a portion of the feed gas stream in an autothermal reforming step thereby producing a second synthesis gas stream, and combining the first synthesis gas stream and the second synthesis gas stream thereby producing a third synthesis gas stream, or reforming the first synthesis gas stream in an autothermal reforming step thereby producing a third synthesis gas stream,
1.3.1	wherein the autothermal reforming step comprises exothermic partial oxidation and endothermic reforming with steam over a reforming catalyst,
1.3.2	wherein the second synthesis gas stream and the third synthesis gas stream comprise hydrogen, carbon monoxide, carbon dioxide, and unreacted methane, and
1.3.3	wherein heat generated by the autothermal reforming step is utilized for heating in the endothermic reforming step of step (b);
1.4	(d) converting the carbon monoxide present in the third synthesis gas stream with steam thereby producing hydrogen and carbon dioxide thereby producing a fourth synthesis gas stream,
1.4.1	wherein the fourth synthesis gas stream comprises hydrogen, carbon dioxide, unreacted methane, and carbon monoxide unconverted in step (d);
1.5	(e) separating hydrogen from the fourth synthesis gas stream by pressure swing adsorption, thereby producing a first hydrogen-rich stream and a first residual gas stream,
1.5.1	wherein the first residual gas stream comprises carbon dioxide, carbon monoxide unconverted in step (d), hydrogen not separated off in step (e), and unreacted methane;
1.6	(f) separating carbon dioxide from the first residual gas stream obtained in step (e) by cryogenic carbon dioxide separation, thereby producing a first carbon dioxide-rich stream and a second residual gas stream,
1.6.1	wherein the second residual gas stream comprises carbon monoxide unconverted in step (d), hydrogen not separated off in step (e), carbon dioxide not separated off in step (f), and unreacted methane.

<i>Reference</i>	<i>Claim 2</i>
2	The process according to claim 1, wherein hydrogen is separated from the second residual gas stream, thereby producing a second hydrogen-rich stream and a third residual gas stream.
<i>Reference</i>	<i>Claim 3</i>
3	The process according to claim 2, wherein the hydrogen is separated from the second residual gas stream by membrane separation.
<i>Reference</i>	<i>Claim 4</i>
4	The process according to claim 2, wherein the second hydrogen-rich stream is supplied to the fourth synthesis gas stream for separation of hydrogen by pressure swing adsorption in step (e).
<i>Reference</i>	<i>Claim 5</i>
5	The process according to claim 2, wherein gases present in the second hydrogen-rich stream are utilized as fuel gases for heating in the autothermal reforming step of step (c).
<i>Reference</i>	<i>Claim 6</i>
6	The process according to claim 1, wherein the first carbon dioxide-rich stream comprises unreacted methane, and the first carbon dioxide-rich stream is subjected to a thermal separation process for separation of methane, thereby producing a second carbon dioxide-rich stream.
<i>Reference</i>	<i>Claim 11</i>
11	The process according to claim 1, wherein the first residual gas stream, for cryogenic carbon dioxide separation in step (f), is subjected to at least one compression step and at least one cooling step, thereby producing the first carbon dioxide-rich stream at least partly in the form of a condensed carbon dioxide stream.
<i>Reference</i>	<i>Claim 12</i>
12	A plant configured for performance of the process according to claim 1.

54. It is my opinion that the Challenged Claims of the '805 Patent are unpatentable as anticipated and/or obvious over the prior art references discussed below.

VII. LEVEL OF ORDINARY SKILL IN THE ART

55. A person of ordinary skill in the art ("POSA") pertinent to the '805 Patent as of its priority date of August 11, 2020 ("critical date") would have at least: (1) 3–5 years of experience with steam reforming of methane, partial oxidation, water gas shift process and synthesis gas separation, and (2) a graduate degree in chemical engineering (M.Sc. level).

VIII. CONSTRUCTION OF CLAIM TERMS

56. I understand that Petitioner has not proposed constructions for any of the claim terms of the '805 Patent. For my analysis and opinions, I have applied the plain and ordinary meaning of the claim terms to a POSA at the time of the invention.

IX. OVERVIEW OF THE PRIOR ART

A. Reinertsen (EX1005)-See Petition I (Grounds 1, 2 and 4)

57. I understand that U.S. Patent Publication No. 2023/0119784 (Reinertsen) is a publication of the U.S. National Stage of PCT International Application PCT/EP2021/054441, filed on Feb. 23, 2021, and claims priority to Great Britain Application Nos. 2003317.1 and 2010174.7, filed on Mar. 6, 2020 and Jul. 2, 2020, respectively. The '371.1 priority application and the '174.7

priority application correspond to GB application 2003317.1, as filed (EX1022) and GB application 2010174.7, as filed (EX1023), respectively.

58. I have examined Reinertsen, the '317.1 priority application (EX1022) and the '174.7 priority application (EX1023), and have found that the relevant aspects of Reinertsen's description, as discussed below, are fully supported by both priority applications, both of which were filed prior to the August 11, 2020 critical date of the '805 Patent. Accordingly, it is my understanding that Reinertsen qualifies as prior art to the '805 Patent.

59. Reinertsen discloses a process for producing hydrogen; e.g., annotated FIGURE 4 below (EX1005, FIGURE 4; EX1022, FIGURE 4; EX1023, FIGURE 4).

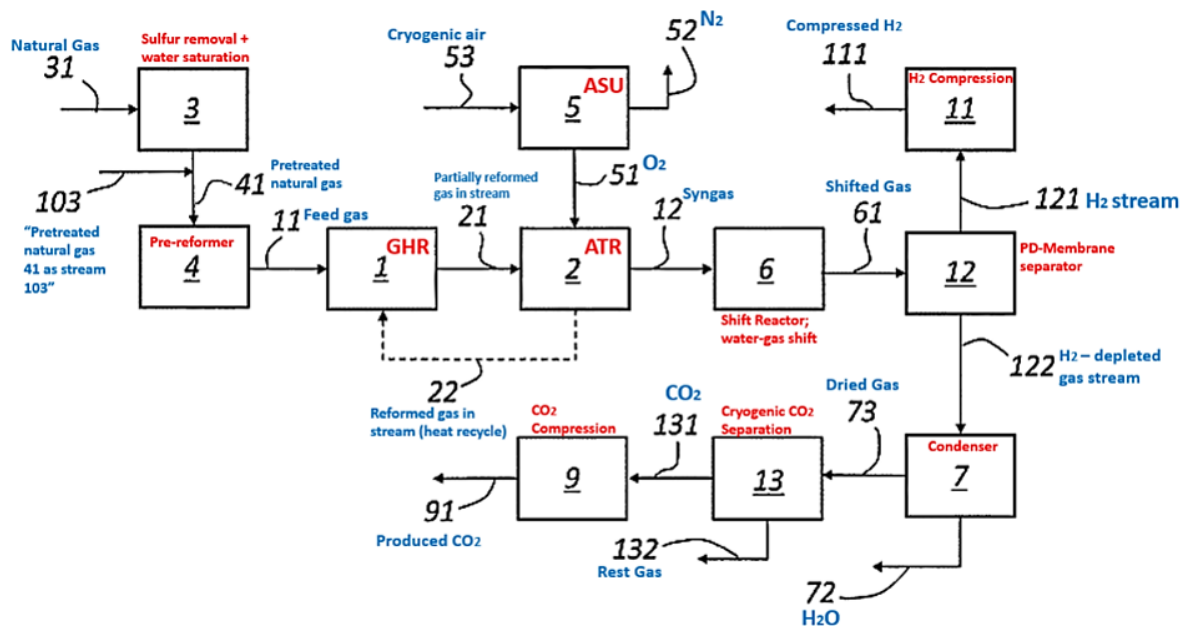


FIGURE 4 (annotated)

60. The method of Reinertsen includes an endothermic reforming step using a GHR (1) and an autothermal reforming step using an ATR (2), where the GHR and ATR components are in a series configuration and where heat generated (e.g., heated gas) from the ATR (22) is utilized for heating the GHR. EX1005, ¶¶[0009], [0074], [0102], FIGURE 1 and FIGURE 4; EX1022, p.4, ln.13–16, p.12, ln.14–23, p.18, ln.27–29, FIGURE 1 and FIGURE 4. The method of Reinertsen also includes conversion of CO and water to CO₂ and hydrogen generating shifted synthesis gas **61** downstream of the ATR and GHR processes via water gas shift conversion (6) and hydrogen separation (12) from shifted synthesis gas provided upstream of cryogenic CO₂ separation (13). A Pd-membrane separator (12) is shown in FIGURE 4, but PSA is disclosed as a process for hydrogen separation useful for some applications. EX1005, ¶¶[0056], [0063], [0085], [0103], [0147] and claim 7. Reinertsen reports an advantage of providing high carbon capture efficiency for the disclosed processes and systems for producing hydrogen from natural gas. EX1005, ¶¶[0057]–[0058].

61. The method of Reinertsen clearly teaches the series configuration of an ATR with a GHR with heat transfer from the ATR to the GHR, followed by WGS. For example, Reinertsen in FIGURE 1 (EX1005) illustrates a dual reforming process comprising a GHR (1) and an ATR (2):

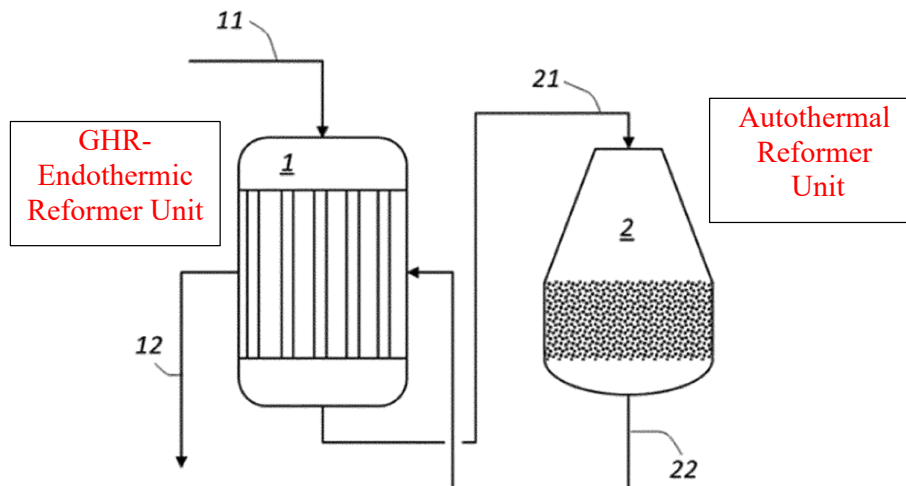


FIGURE 1. (annotated)

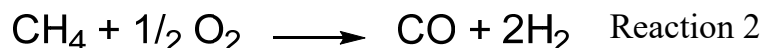
62. In the series reformer configuration illustrated in FIGURE 1, feed gas **11** passes through a catalyst in a GHR (**1**) to partially reform natural gas (methane). The output stream **21** (synthesis gas) then passes to an ATR (**2**) to generate further reformed gas in stream **22**. In this series configuration, gas stream **22** passes through the heating side of the GHR to provide heat for the endothermic reforming reaction in the GHR. Synthesis gas stream **12** from combined reforming in **1** and **2**, is shown in FIGURE 1 as exiting the GHR (**1**). I note that in FIGURES 2-9, stream **12** is schematically shown as exiting the ATR (**2**). Formally, in the dual reformer series configuration with direct heat transfer the combined synthesis gas stream **12** exits the GHR as shown in FIGURE 1. The illustrations of the series reformer configuration and gas flows in FIGURES 2-9 with streams **12** and **22** should be considered schematic showing the principle of heat transfer to the GHR and not to show the detail of specific gas stream flows in the reformer configuration.

63. Synthesis gas stream **12** is generated by combined reforming in the GHR and the ATR. EX1005, ¶[0074]. In a GHR process, methane is at least reacted with water (typically as steam) to generate carbon monoxide and hydrogen (synthesis gas) (as in the steam reforming Reaction 1, (*see*: ¶28, *supra*)).



The GHR unit carries out at least this reaction which is an endothermic reforming step. Thus, methane reforming in a GHR comprises an endothermic reforming step and this would have been understood by a POSA at the critical date.

64. In an ATR, feed gas (methane) is fed together with oxygen where in addition to Reaction 1, at least partial oxidation of methane Reaction 2 takes place (*see*: ¶29, *supra*) to generate additional carbon monoxide and hydrogen (synthesis gas):



Partial oxidation is exothermic and provides the energy required for the endothermic step of Reaction 1. As noted above (*see*: ¶29, *supra*), Reaction 2 is a simplified way of describing the partial oxidation of methane that occurs in an ATR, for example, some level of the highly exothermic full oxidation occurs to generate CO₂. Furthermore, some of the generated H₂ is oxidized to water. At

the end of the ATR process, all oxygen is reacted and the gas from partial oxidation has a high temperature.

65. The reformer combination of FIGURE 1 (EX1005) provides an example of a series combination of an endothermic reforming step (1) and an autothermal reforming step (2) in which FG containing methane and steam is converted to synthesis gas produced by the combined endothermic reformer step (GHR) and the autothermal reformer (ATR) step (2). In this configuration, hot autothermally reformed gas **22** exiting the ATR transfers heat to the endothermic reformer step of the GHR.

66. Reinertsen also provides a WGS (6), hydrogen separation (12, illustrated as a Pd-membrane) and cryogenic CO₂ separation processes downstream of the GHR and ATR reforming processes, as shown in process flow charts which are provided in FIGURES 2-9 (EX1005). In all Reaction 3 charts of Reinertsen, synthesis gas produced by the combined endothermic and autothermal reformers (2) is first converted in a shift reactor (a water-gas shift reactor 6) by Reaction 3 (EX1005):



converting carbon monoxide and water to carbon dioxide and additional hydrogen.

67. Reinertsen provides a process for separating pure hydrogen downstream of the ATR, GHR and WGS processes. EX1005, ¶[0005]. The

hydrogen separation process includes a Pd-membrane, an example of a hydrogen selective membrane. EX1005, ¶[0011]. Alternatively, preferably the hydrogen separation process comprises a PSA process. EX1005, ¶[0012]. At various places in the application, PSA is disclosed as an alternative process option for separation of hydrogen. EX1005, ¶¶[0056], [0063], [0081], [0085], [0147] and claim 7. A POSA, therefore, would view a PSA as readily interchangeable with a Pd-membrane in the process of Reinertsen. The interchangeability of the membrane separation and PSA separation is also shown in Table 1 of Figure 10 of Reinertsen, which provides a summary of process schemes in which either the Pd membrane or PSA are options. EX1005, FIGURE 10. Reinertsen also teaches that embodiments preferably use cryogenic separation to separate CO₂. EX1005, ¶[0059].

68. Reinertsen cites to J. R. Rostrup-Nielsen I (EX1027), and J. R. Rostrup-Nielsen II (EX1028). EX1005, ¶[0055].

69. The J. R. Rostrup-Nielsen I and Rostrup-Nielsen II references (EX1027 and EX1028) provide an overview of syngas (synthesis gas) production, applications, and its significance in the field of catalysis and energy and describe in detail methane reforming, water gas shift conversion, and the composition of resulting synthesis gas.

70. As noted above, it is my opinion that Reinertsen is supported by the '317.1 and the '174.7 priority applications because the '317.1 the '174.7

priority applications describe the aspects of Reinertsen corresponding to the subject matter of at least claims 1-6, 11 and 12 of the '805 Patent. Therefore, it is my understanding that Reinertsen is entitled to the earlier March 6, 2020 and July 2, 2020 filing dates of the '317.1 the '174.7 priority applications, respectively. In the comments that follow, therefore, I include parallel references to both Reinertsen and the '317.1 the '174.7 priority applications. Reinertsen and the '317.1 and '174.7 priority applications are all directed to a process for preparing hydrogen by reforming hydrocarbons with steam, and providing for separation of carbon dioxide. EX1005, FIG. 4, ¶¶[0005], [0064], [0109]; EX1022, FIG. 4, p.4, ln.10–20, p.10, ln.4–9, p.20, ln.24–p.21, ln.2; EX1023, p.4, ln.9–20, p.10, ln.10–15, p.21, ln.5–12. Reinertsen and the '317.1 and '174.7 priority applications disclose dual reformers (i.e., GHR and ATR) provided in a series configuration, in which it is explained that heat generated from the ATR is used for heating an endothermic reforming process in the GHR. EX1005, ¶¶[0067]–[0068], [0070], [0074], [0102]; EX1022, p.10, ln.18–22, p.11, ln.10–22, p.12, ln. 14–23, p.18, ln.24–p.19, ln.4; EX1023, p.10, ln.24–p.11, ln. 3, p.11, ln.17–29, p.12, ln. 20–29, p.19, ln.4–14. Reinertsen and the '317.1 and '174.7 priority applications disclose converting CO in synthesis gas generated from the dual reformers via a water gas shift reaction for the purpose of generating synthesis gas enriched with CO₂ and hydrogen. EX1005, ¶¶[0103], [0067]; EX1022, p.17, ln.5-21, p.10, ln.5–21, p.10, ln.18-19; EX1023,

p.17, ln.15-31, p.19, ln.15-31. Reinertsen and the '317.1 and '174.7 priority applications disclose separating hydrogen from synthesis gas, including description of PSA as an option for hydrogen separation preferred for some processes. EX1005, ¶¶[0012], [0063], [0081], [0085], [0106]; EX1022, p.15, ln.9-21, p.20, ln.4-11 and EX1023, p.5, ln.10, p.10, ln.7-9, p.15, ln.11-31, p.20, ln.15-22. Reinertsen and the '317.1 and '174.7 priority applications disclose cryogenic separation for capture of CO₂, including the use of compression and cooling steps such that CO₂ is liquefied. EX1005, ¶¶[0059], [0107], [0110], EX1022, p.9, ln.14-16, p.20, ln.12-16, p.21, ln.3-5, EX1023, p.9, ln.16-19, p.20, ln.23-27, p.21, ln.13-15. Reinertsen and the '317.1 and '174.7 priority applications disclose a plant for carrying out a process for preparing hydrogen by reforming hydrocarbons with steam, and providing for separation of carbon dioxide. EX1005, ¶[0029], EX1022, p.6, ln.18-19, EX1023 p.6, ln.19-20.

B. Martin (EX1006)-See Petition II (Grounds 1-3)

71. I understand that the article, “Progress Update on the Allam Cycle: Commercialization of NET Power and the NET Power Demonstration Facility”, by Martin et al., is a Conference proceedings of the 14th International Conference on Greenhouse Gas Control Technologies, GHGT-14 held on the 21st -25th October 2018, Melbourne, Australia. (EX1006, page 1). I have been informed that Martin was uploaded to SSRN’s online database by April 11,

2019 and that this reference was publicly accessible at least as early as this date (EX1013). I therefore understand that Martin qualifies as prior art to the '805 Patent because it was publicly accessible prior to the critical date.

72. Martin discloses a process for producing hydrogen; e.g., annotated Figure 7, below. The method of Martin includes an endothermic reforming step using a GHR and an autothermal reforming step using an ATR (or a POX), where GHR and ATR components are provided in a parallel configuration and where heated gas from the ATR is utilized for heating the GHR. EX1006, Figure 7, p.12. The method of Martin also includes conversion of CO to CO₂ via water gas shift (WGS) conversion (“SYNGAS/COOLING & SHIFT” in Figure 7). The process in Martin further provides a pressure swing adsorption (PSA) process (“H₂ PSA” in Figure 7) for hydrogen separation that is provided upstream of a CO₂ separation and purification system (“LOW TEMPERATURE CO₂ SEPARATION” in Figure 7). The CO₂ separation and purification system in Martin is a cryogenic carbon dioxide separation system, which operates by cooling the gas stream near the CO₂ triple point to give at least partial condensation of CO₂ to separate and purify a CO₂ product stream. EX1006, p.12. Martin references additional purification of carbon dioxide using a distillation column. EX1006, p.12. Further, to improve process efficiency and CO₂ capture, Martin describes that residual gas from the CO₂ separation and purification system can be recycled back to the reforming reactors. EX1006,

p.12. Martin Figure 7 also shows a FIRED HEATER upstream of the parallel reformers to heat natural gas feed prior to reforming, as I have explained ¶35 *supra*, preheating of feed gas into an ATR increases process efficiency.

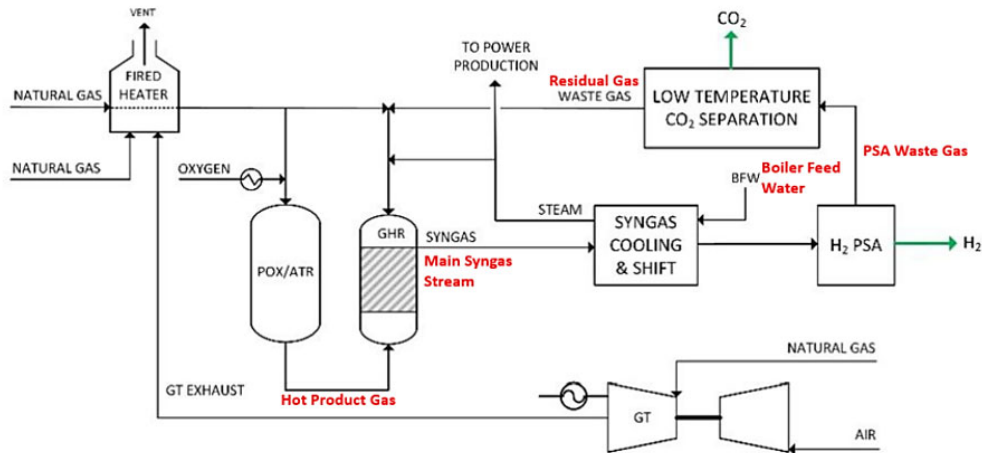


Figure 7 (annotated)

On review of Figure 7, I note an obvious omission of a steam feed into the ATR, when the autothermal reforming step is an ATR. As I explained, ¶33, Figure 55, *supra*, the feed into the ATR should include steam and this in my opinion would have been obvious to a POSA reviewing Figure 7. In contrast, when a POX is employed a large amount of steam input is not required.

C. Rytter (EX1009)-See Petition I (Ground 3)

73. I understand that WIPO Publication No. WO 19162236 (Rytter) corresponds to PCT international application PCT/EP2019/053986, filed on February 18, 2019, and published on August 29, 2019. I understand that Rytter

qualifies as prior art because it published prior to the critical date of the '805 Patent.

74. Rytter discloses a process for producing hydrogen as illustrated in annotated Figure 3 below. The method of Rytter includes an endothermic reforming step using a GHR (1) and an autothermal reforming step (2) using an ATR. The GHR and ATR components are provided in a series configuration and heat generated 22 (e.g., heated gas) from the ATR is used for heating the GHR. EX1009, p.4, ln.20–24.

75. Rytter, in Figure 1, shows a reforming process comprising an endothermic reformer step (GHR 1) and an autothermal reforming step (ATR 2):

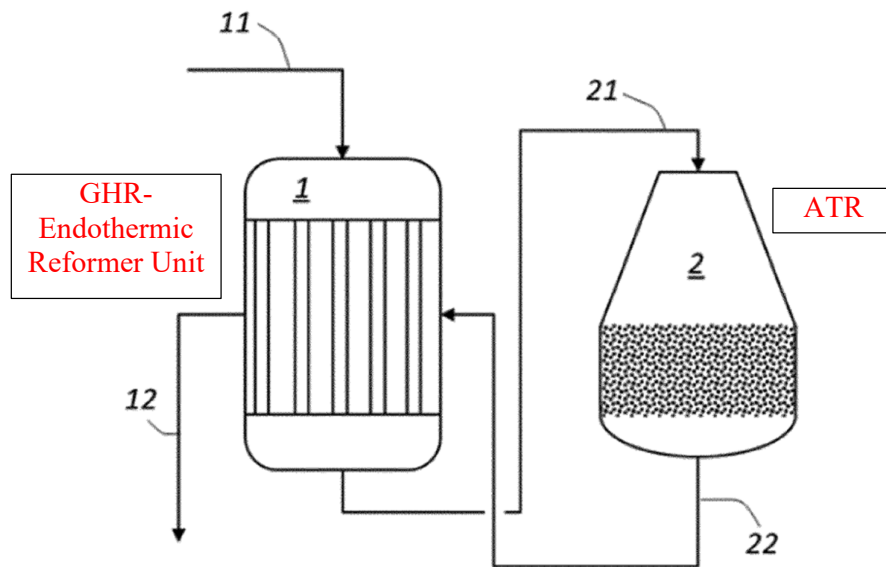


Figure 1 (annotated)

76. In the combined reformer configuration of Rytter, feed gas 11 passes through a catalyst in a GHR (1) to partially reform natural gas (methane)

in stream 21. Stream 21 then passes to an ATR (2) to generate further reformed gas in stream 22. Gas stream 22 passes through the heating side of the GHR to provide heat for the reforming reaction in the GHR. Gas stream 12 contains synthesis gas generated by combined reforming in the GHR and the ATR.

EX1009, p.10, ln.23–24. The combined reformer combination of Figure 1 provides a series configuration including an endothermic reforming step (1) and an autothermal reforming step (2) in which FG containing methane and steam is converted to synthesis gas produced by the combined endothermic reformer step and the autothermal reformer steps (1 and 2). In this configuration, hot synthesis gas exiting the autothermal reformer (22) transfers heat to the endothermic reformer step.

77. Figure 3 of Rytter illustrates an example process for production of hydrogen and carbon dioxide using the combined GHR and ATR of Figure 1. Synthesis gas produced in the combined reformers is treated with a shift reactor (12) (by water-gas shift reaction) to convert carbon monoxide to carbon dioxide and additional hydrogen (Reaction 3 above) to produce synthesis gas enriched in carbon dioxide and hydrogen (121, shifted gas). The process of Rytter also includes downstream processes for CO₂ separation and hydrogen separation (EX1009, p.15, ln.13–17). PSA is disclosed as an alternative for hydrogen separation and cryogenic CO₂ separation is disclosed as an alternative for CO₂ separation. EX1009, p.6, ln.18–23. For example, Figure 3 of Rytter

shows a separation process 13 which separates carbon dioxide (131) from the shifted gas where carbon dioxide is compressed in a compressor 14 and optionally liquefied (EX1009, p.15, ln.13–14) and hydrogen 151 is separated in process 15, for example by PSA (EX1009, p.15, ln.16–17). In Rytter, the rest gas of the PSA 152 can be utilized for fuel or recycled back into the process (e.g., the GHR or ATR) to maximize carbon efficiency. Similar to the description of Reinertsen above (Section IX.A. *supra*), the illustration of the series reformer configuration and gas flows in FIGURE 3 of Rytter with streams 12 and 22 should be considered schematic showing the principle of heat transfer to the GHR and not to show the detail of specific gas stream flows in the reformer configuration.

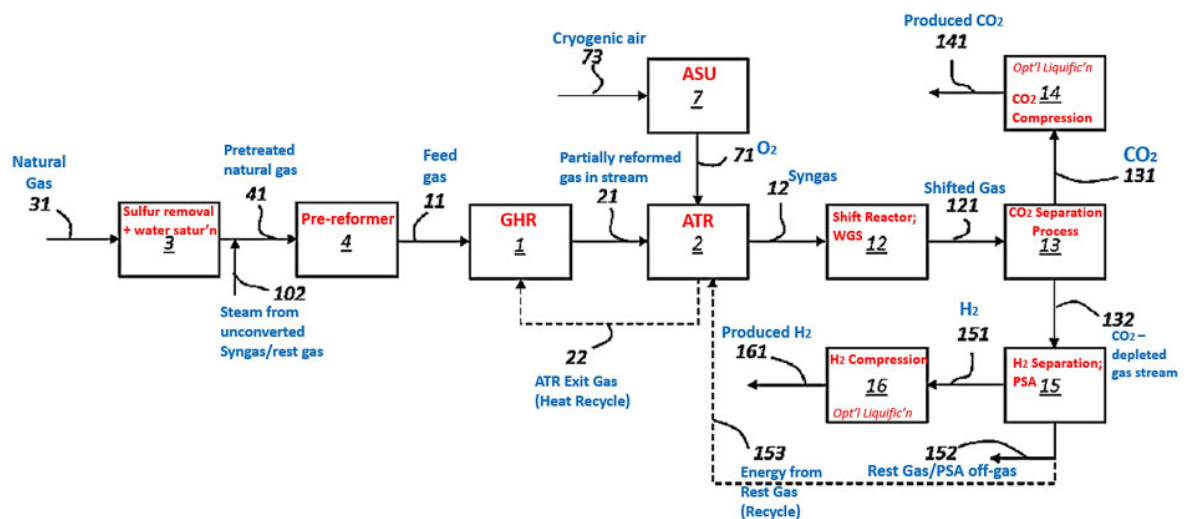


Figure 3 (annotated)

D. Darde (EX1008)-See Petition I (Grounds 2-4)

78. I understand that WIPO Publication No. WO 2014/091098 A1 (Darde, in French EX1007) corresponds to PCT international application PCT/FR/0552613, filed on October 31, 2013, and published on June 10, 2015. I have been informed that the Darde reference is available as prior art to the '805 Patent because it published prior to the '805 Patent, given its publication date predates the August 11, 2020 filing date of the '366 priority application serving as the earliest priority of the '805 Patent. I have reviewed the certified English translation of EX1007, which is EX1008. EX1008 contains one figure which is not numbered.

79. Darde discloses a process for producing hydrogen; *see*. e.g., annotated Figure. The method of Darde includes an endothermic reforming step using steam methane reforming (SMR). EX1008, p.7, ln.22–25. The process in Darde is implemented by addition of natural gas (1) and steam (22 and 27) to a SM (4) with production of synthesis gas (5). EX1008, p.15, ln.26–31. Synthesis gas (5) contains H₂ and CO (but also CO₂, CH₄, water and impurities). EX1008, p.15, ln.30–31. This synthesis gas (5) is cooled and introduced into a conversion module (7) in which CO in the presence of water is converted by water-gas shift reaction to CO₂ and additional H₂, producing a synthesis gas enriched in CO₂ and H₂. EX1008, p.15, ln.29–36. This CO-enriched synthesis gas is cooled to condense water and saturated synthesis gas is produced (9). EX1008, p.15, ln.37–p.16, ln.5. The synthesis gas (9) is then passed to a PSA unit (11) to

produce a stream of highly purified hydrogen (shown exiting PSA **11**) and a residual gas (**12**) containing all of the carbon dioxide, unconverted methane, unconverted carbon monoxide, nitrogen and hydrogen. EX1008, p.16, ln.6–10. The residual gas (**12**) is then sent to a cryogenic purification unit CPU (**13**) for separation of at least one stream of carbon dioxide (**14**), as well as a gaseous stream rich in hydrogen (**15**) which is recycled to the PSA (**11**) for additional recovery of hydrogen. EX1008, p.16, ln.11–22. The CPU unit is also described as producing a non-condensable CPU offgas (**16**) comprising methane, hydrogen, carbon monoxide, non-condensable carbon dioxide, nitrogen and water which is recycled as fuel to the reformer (EX1008, p.16, ln.14–16) and a liquid stream (**17**) of CPU condensates which are primarily water with dissolved impurities. EX1008, p.16, ln.17–20. Darde also discusses separation of condensates (C1 and C2) from the process streams and the combination of various water containing streams (**20**) to ultimately generate impure steam (**22**). EX1008, p.16, ln.23–32.

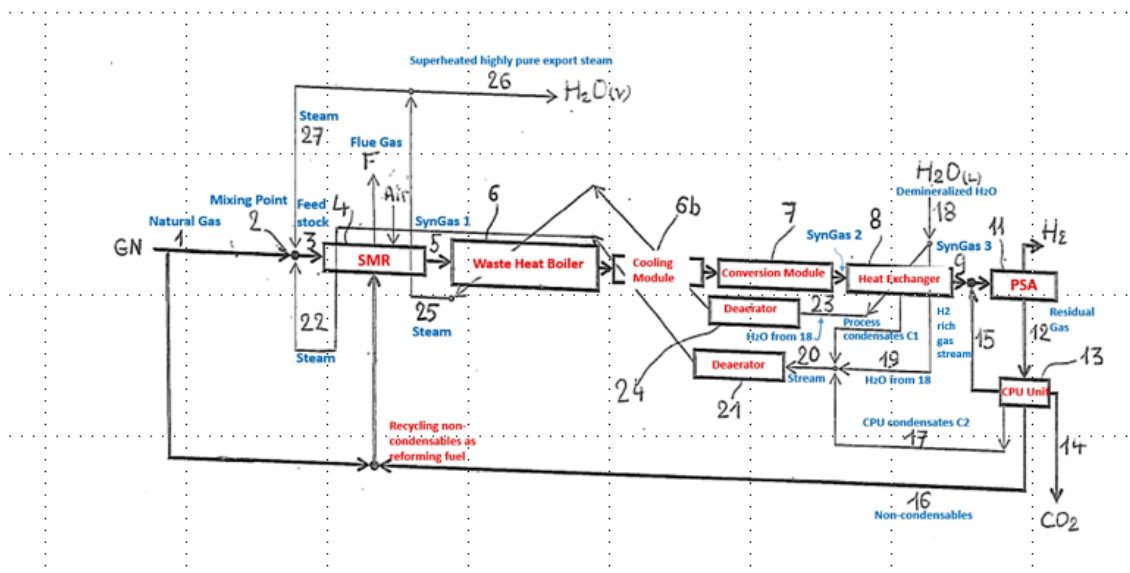


Figure (annotated)

80. Darde explicitly discloses the compositions of feed gas streams including methane and steam (“...the hydrocarbons to be reformed are mixed with steam so as to produce the feedstock for the reforming...”). EX1008, p.4, ln.7–8. Darde explicitly discloses the compositions of the synthesis gas stream produced by reforming including carbon monoxide, hydrogen, carbon dioxide and unreacted methane (“...a synthesis gas is generated, comprising essentially hydrogen, carbon monoxide, carbon dioxide and also excess steam, residual methane, water and impurities...”). EX1008, p.4, ln.8–10. Darde discloses a shift reaction processes for producing a synthesis gas composition comprising carbon dioxide, carbon monoxide unconverted, hydrogen not separated off, and unreacted methane; for example (“...a synthesis gas enriched with H₂ and CO₂ and depleted of CO compared with the gas 5 produced by reforming, and also containing additional impurities generated during the conversion - of about 65%

to 85% of H₂, 11 % to 22% of CO₂, 0.5% to 6% of unconverted CO and 3% to 10% of CH₄. EX1008, p.15, ln.33–36. Darde explicitly discloses the almost pure hydrogen rich stream (H₂ with arrow) from PSA (**11**) and the residual stream (**12**) of the PSA as including carbon dioxide, carbon monoxide, methane, hydrogen not separated in the PSA step and unreacted methane. EX1008, p.8, ln.1-4. Darde also states that the PSA offgas (e.g., **12**) is usually sent as fuel to the burners of the SMR or the dedicated boiler of an ATR. EX1008, p.8, ln.5–7. Darde explicitly discloses the compositions of the residual non-condensable stream CPU off gas (**16**) produced by cryogenic CO₂ separation as including hydrogen, unreacted methane, carbon monoxide and carbon dioxide not separated in the cryogenic CO₂ separation step. EX1008, p.16, ln.11–16.

81. The specific means by which the gaseous stream **15** rich in hydrogen is produced from CPU unit **13** is not explicitly described in the Figure or the more general description of cryogenic CO₂ capture in the reference. EX1008, p.8, ln.22–30. I note that the CPU unit would normally separate a stream containing CO₂ (e.g., i.e., a liquid stream) from at least a capture offgas that would contain non-condensable gases. The CPU unit **13** is shown in the Figure as generating two non-condensable streams **15** (hydrogen-rich stream) and **16** which is called the CPU offgas. EX1008, p.16, ln.11–16. The gases of both of these non-condensable streams must be generated in the CPU and represent the actual capture offgas of the CO₂ separation. In my opinion, there

must be some form of hydrogen separation that is employed in Darde to generate the hydrogen rich gas stream **15**. Darde does state that the condensation/separation steps in the CPU unit can be supplemented by membrane separation steps. EX1008, p.8, ln.29–30. I note that it was well-known in the art prior to the critical date to separate H₂ from a residual gas stream exiting cryogenic CO₂ separation. As I discussed ¶41, *supra*, in the Cryocap™ H₂-Cryogenic CO₂ separation (L’Air Liquide, France) non-condensable gases from cryogenic separation “are recycled through a membrane system to recover H₂”. EX1018, p.24. Thus, in my opinion hydrogen-rich stream **15** is generated by some form of hydrogen separation, likely a H₂ membrane is in the CPU unit, that is not explicitly described in Darde.

E. Rafati (EX1015)-See Petition II (Grounds 1- 3)

82. I understand that U.S. Patent Publication No. 2019/0135626 (Rafati) published on May 9, 2019. I have been informed that the Rafati reference is available as prior art to the ’805 Patent because it published prior to the critical date of the ’805 Patent.

83. Rafati describes a method for the production and separation of hydrogen and carbon dioxide from hydrocarbon fuel streams in the presence of steam using two-stage reforming processes. EX1015, ¶[0014]. In particular, Rafati discloses:

“... [T]he present disclosure further can relate to systems and methods for generation of H₂+CO in a PDX or ATR reactor followed by the use of a

gas heated reformer (GHR) in either a series or parallel mode to the PDX or ATR reactor to produce additional H₂ and CO (i.e., synthesis gas) by utilizing the exhaust sensible heat in the PDX and/or ATR reactor system to provide the heat for endothermic catalytic steam plus hydrocarbon reforming reactions taking place in the GHR.” EX1015, ¶[0016] (emphasis added).

Thus, before the critical date, a POSA understood that production and separation of H₂ and CO₂ from a hydrocarbon component in the presence of steam could be accomplished using a dual reforming process having an ATR step and a GHR step where the heat from the ATR provides heat for the endothermic reforming reaction in the GHR.

84. Rafati discusses certain advantages of using a two-stage reforming process:

“An advantage of the two reactor configuration is that the yield of H₂+CO from hydrocarbon feed is maximized, and all CO₂ formed in the reactions is contained within the high-pressure system.” EX1015, ¶[0115] (emphasis added).

In view of Rafati, a POSA wishing to enhance H₂ yield from a hydrocarbon source would, thus have been motivated to use such a dual reforming process.

85. Rafati illustrates an example of a reforming process comprising a PDX and/or ATR reactor system and a GHR process in annotated Fig. 2:

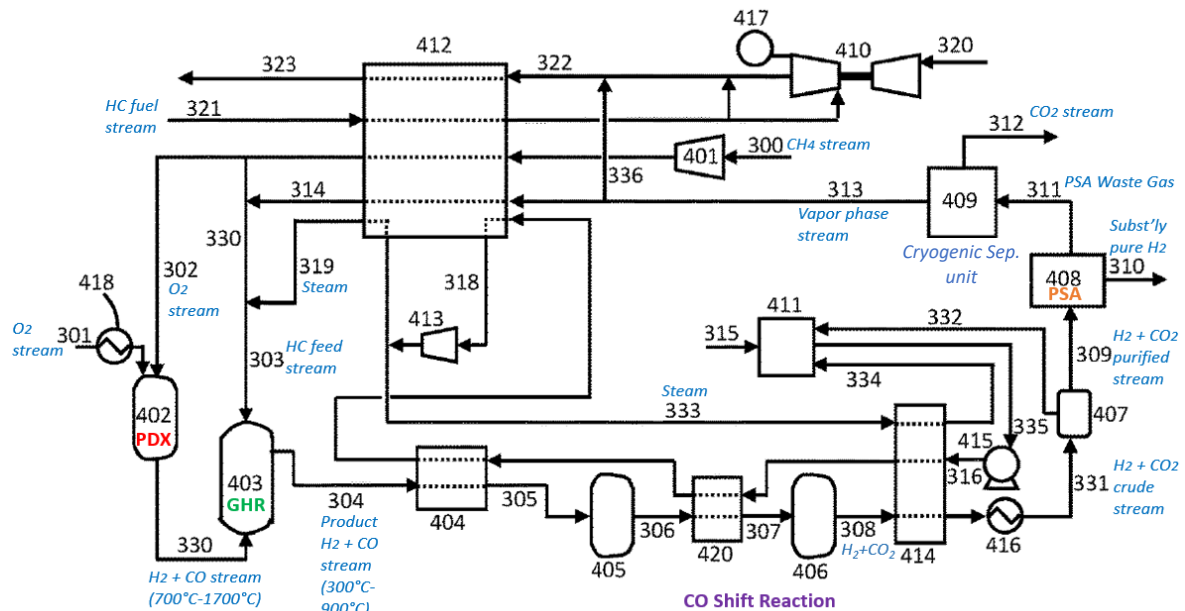


Fig. 2 (annotated)

86. Fig. 2 shows an example dual reforming system and a CO conversion system (405, 420, 406, and 414) paired with a downstream pressure swing adsorption (PSA) unit (408) and a cryogenic separation unit (409) to enhance hydrogen yield and facilitate CO₂ capture. EX1015, ¶[0083]:

“...processing the stream comprising H₂+CO₂ in a pressure swing adsorber to provide a product stream formed of substantially pure hydrogen and also provide waste gas stream comprising CO₂; forming a liquid CO₂ product stream in a cryogenic separation unit operating with auto-refrigeration by passing the waste gas stream comprising CO₂ therethrough such that at least 50 mol % of the CO₂ in the waste gas stream comprising CO₂ is separated into the liquid CO₂ product stream.”.

87. Rafati gives further details of the reactions occurring at each step of the process, including the compositions of the feed streams, syngas streams, and waste streams. EX1015, ¶¶[0114], [0116], [0124], [0126]–[0129]. Thus, prior to the critical date, a POSA understood the reactions and compositions of the streams present at each stage of the process for producing H₂ and separating CO₂ using a dual reforming system, a CO conversion step, a PSA separating step, and a cryogenic CO₂ separating step.

88. Rafati also describes the provision of heat to the hydrogen production system (EX1015, ¶[0019]) and specifically states that heat can be added to the system for “superheating one or both of a fuel stream (e.g., natural gas) and a stream feed to an H₂ + CO synthesis gas generation reaction system(e.g., any one or more of an SMR, a PDX, an ATR, a PDX+CIHR, or an ATR+GHR).” EX1015, ¶[0019]. Rafati further explains that “efficiency of the H₂ + CO generation in the two reactors can be significantly increased by preheating the hydrocarbon and steam feeds” preferably using an external heat source. EX1015, ¶[0118]. This description in Rafati confirms that it was known in the art prior to the critical date that preheating of feed gas to reformers generally increased the efficiency of production of synthesis gas.

F. Gauthier (EX1014)-See Petition II (Ground 2)

89. Gauthier is U.S. patent application 20090298957, published on December 3, 2009 that issued as U.S. patent 8,021,464 on September 20, 2011.

I have been informed that Gauthier is prior art to the '805 Patent because it was published before the critical date.

90. Gauthier describes "...a method for combined production of hydrogen and carbon dioxide from a mixture of hydrocarbons...." EX1014, Abstract. For example, Figure 1 illustrates a method of separating H₂ from shifted synthesis gas (13) using a PSA (14). EX1014, ¶[0143].



Figure 1

91. Compressed and purified PSA offgas (19, residual gas), after condensation and drying (21), is cooled and a condensate containing CO₂ (23) is separated by cryogenic purification (22). EX1014, ¶¶[0144]–[0146]. The cryogenically separated CO₂ is then subjected to distillation (25) to remove light impurities and generate food grade CO₂ (26). EX1014, ¶[0147]. PSA offgas, when feed gas is natural gas, is described as typically containing “...close to: CO₂: 45%-CO: 12%-H₂: 23%-CH₄: 17%-H₂O: 1 %-N₂: 2%.” EX1014, ¶[0143]. Light impurities to be distilled from CO₂ (molar mass of 44.01 g/mol) would be expected by a POSA to include methane (molar mass of 16.84 g/mol). With respect to the distillation process described, CH₄ is a light impurity in the cryogenically separated CO₂ that can be removed from CO₂ by distillation. Distillation is an example of a thermal process as defined in the '805 Patent that can be used to separate CH₄ from CO₂.

92. A gas mixture resulting from the cryogenic separation (24), a purge gas containing CO₂, CO, H₂, CH₄ and N₂ (EX1014, ¶[0149]) is treated in a permeation module (29) to generate a hydrogen-rich permeate (31) rich in hydrogen and a purge waste (30) containing CH₄ and CO (EX1014, ¶[0150]). Generation of hydrogen-rich permeate (31) indicates that the permeation module is a hydrogen selective membrane. The hydrogen-rich permeate is recycled to the inlet of the hydrogen purification unit (14, PSA type). EX1014, ¶[0151]. The purge waste (30) from this membrane purification is recycled in

part to reformer feed (**30a**, prereformed mixture) and also to be used as fuel in the reforming burners (**30b**). EX1014, ¶[0152]. The purge waste can be used as fuel in “the combustion chamber of a nonadiabatic reformer, or in a feed preheating furnace in the case of an autothermal reformer or a partial oxidation.” EX1014, ¶¶[0075], [0152].

G. Terrien (EX1024)-See Petition I (Ground 4) and Petition II (Ground 3)

93. Terrien (EX1024) is U.S. 2012/0291484, published November 22, 2012, based on U.S. application 13/169,241, filed June 27, 2011. I have been informed that Terrien is prior art to the '805 Patent because it was published before the critical date.

94. Terrien describes a method to more efficiently recover hydrogen and carbon dioxide from a syngas stream of a hydrogen generation plant. EX1024, Abstract. Recovery of hydrogen and capture of carbon dioxide is increased by balancing recycle of hydrogen-rich permeates from hydrogen membrane separation units to the process unit. EX1024, Abstract.

95. Terrien FIG. 5 (EX1024, ¶¶[0077]–[0078]) provides an exemplary process for improved recovery of hydrogen and carbon dioxide from a process stream (**1**) obtained from a process unit (**9**). The process unit (**9**) is any unit which includes a purification step that results in a hydrogen rich fraction, e.g., stream **27** (EX1024, ¶[0018]), preferably a pressure swing adsorption (PSA) unit that is used to recover and purify hydrogen. EX1024, ¶[0020]. Process

stream **1** thus can be residual gas from a PSA unit and typically contains at least methane, carbon monoxide, carbon dioxide, water and any unrecovered hydrogen. EX1024, ¶[0021].

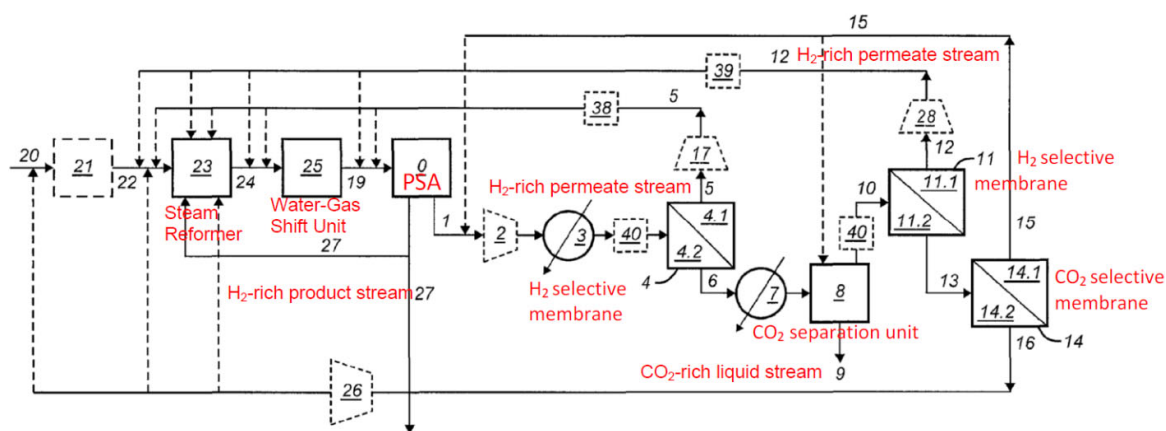


FIG. 5 (annotated)

96. The process of FIG. 5 is an exemplary hydrogen generation plant (EX1024, ¶[0078]) in which hydrocarbon feed stream **20** is optionally pre-reformed to form pre-reformed gas **22** which is reformed in a steam reformer unit **23** to generate syngas **24**. EX1024, ¶[0073]–[0074]. Syngas **24** is then shifted in a water gas shift reactor **25** to generate feed gas **19** (i.e., shifted synthesis gas). EX1024, ¶[0075]. Hydrogen rich stream **27** is separated along with a process stream **1**, in process unit **0**, for example a PSA unit. EX1024, ¶[0076]–[0077].

97. FIG. 5 shows passage of process stream **1** (a hydrogen depleted stream after passage through process unit **9**) to a first hydrogen selective membrane separation unit **4** (EX1024, ¶[0077]) to generate hydrogen rich permeate stream **5** and residual stream **6** which is cooled and then subjected to separation/purification in carbon dioxide separation unit **8** (EX1024, ¶[0077]) to produce carbon dioxide rich liquid stream **9** and carbon dioxide lean non-condensable stream **10**. EX1024, ¶[0077]. Stream **10** is then passed through a second hydrogen selective membrane separation unit **11** forming a second hydrogen rich permeate stream **12** and a second hydrogen lean residue stream **13**. EX1024, ¶[0077].

98. The carbon dioxide separation unit is described as typically carried out utilizing single or multi-step partial liquefaction as depicted in FIGs. 2-4. EX1024, ¶[0038]. For example, the carbon dioxide separation unit of FIG. 4 (shown below) includes two liquefaction units (**18.1** and **18.2**), one distillation column (**29**), one compressor (**30**) and one heat exchanger (**31**). EX1024, ¶[0038]. The liquefaction temperature is said to be generally between about -10° C and -57° C, and preferably between about -30° C and -57° C. EX1024, ¶[0038]. The description of the carbon dioxide separation unit (EX1024, ¶[0038]–[0041]) refers to unit **8** and the CO₂-rich liquid stream as **9** (as in FIG. 5, above), but the carbon dioxide separation units and the CO₂-rich

liquid stream are clearly erroneously labelled unit 4 and stream 6 in FIGs. 2-4.

The errors in labeling are corrected in the annotated excerpt of FIG. 4 below.

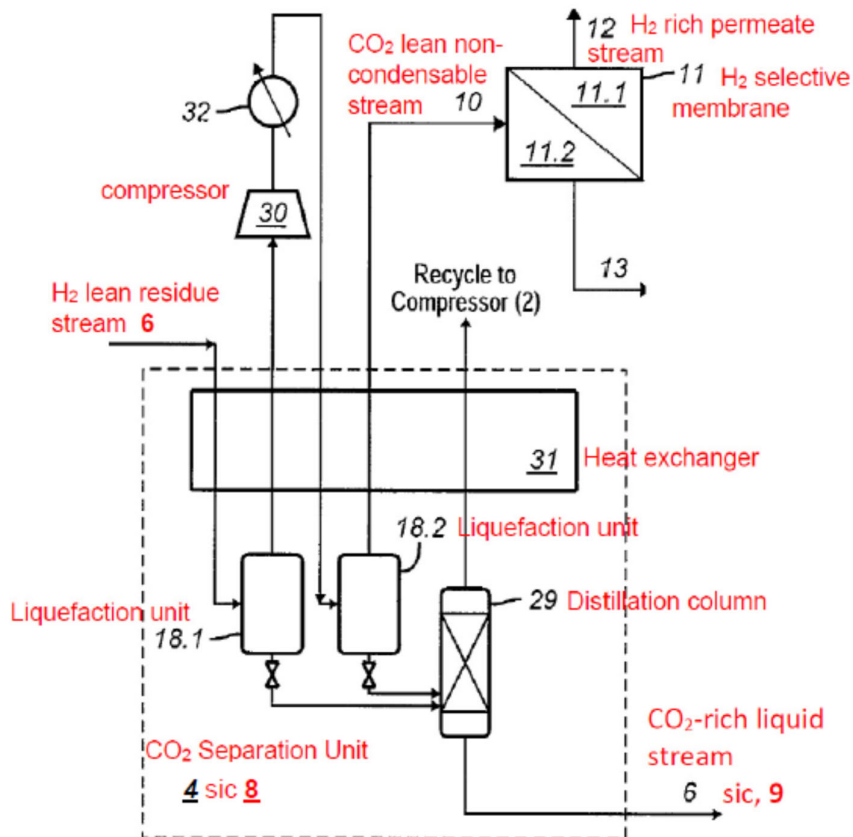


FIG. 4 (excerpt, annotated, labeling corrected)

99. While, Terrien does not specifically call the carbon dioxide separation 8 of FIG. 5 (as further illustrated in FIGs. 2-4) a cryogenic carbon dioxide separation, the process units and steps described require cooling of the gas stream apparently to a low temperature above the triple point of carbon dioxide with at least one compression step and a distillation step. A POSA in view of the description in Terrien and what is known and understood in the art,

would have recognized the carbon dioxide separation unit **8** of Terrien to be a cryogenic carbon dioxide separation unit.

100. Terrien specifically exemplifies the use of a steam reformer in the processes of FIGS. 5 and 6. However, Terrien describes that feed gas **19** can be generated from a reformer unit/water gas shift unit, a partial oxidation unit (POx), an autothermal reformer unit (ATR), syngas from a coal gasification unit, refinery off gas or any other gas mixture that contains hydrogen, carbon monoxide and carbon dioxide. EX1024, ¶[0019]. Thus, Terrien teaches that the steam methane reforming unit of the processes of FIGs. 5 and 6 can be replaced, for example, with an ATR.

101. An important feature of the Terrien process is recycling of hydrogen-rich streams for use as feeds and/or fuel. EX1024, Abstract. A portion of hydrogen rich product stream **27** can be recycled for use for hydrogen fueling of steam methane reformer **23**. EX1024, ¶[0077]. Similarly, hydrogen rich permeate stream **5** can be recycled for supplemental feed to the steam methane reformer, the water gas shift reactor or the PSA of the process unit **0** or as fuel for steam reformer **23**. EX1024, ¶[0077]. Additionally, hydrogen rich permeate stream **12** can be used as supplemental feed for process unit **0** (i.e., the PSA). EX1024, ¶[0077]. However, when the method is employed in a hydrogen generation plant as shown in FIG. 5, hydrogen rich permeate stream **12** is

preferably used as a fuel to steam methane reformer **23** resulting in a boost in carbon dioxide capture. EX1024, ¶[0078].

102. Feed gas **19** from which process stream **1** is generated can be any feed stream that typically will be subjected to treatment for removal of hydrogen. EX1024, ¶[0019]. Exemplary feed gas **19** can be any mixture containing hydrogen, carbon monoxide and carbon dioxide. EX1024, ¶[0019]. More specifically, feed gas **19** will be the product of a hydrocarbon containing feed stream **20** that has been subjected to a reformer unit/water gas shift unit, an ATR unit, a POx unit or a gasification unit. EX1024, ¶[0019]. Terrien states that feed gas **19** can be shifted synthesis gas when the process is implemented in a hydrogen generation plant. EX1024, ¶[0075].

103. Thus, Terrien demonstrates that it was known in the art at the critical date to employ hydrogen rich permeate streams generated from hydrocarbon reforming by passage through hydrogen selective membranes as feeds to various process steps, including to a PSA for additional H₂ capture or as fuel in the reforming process. More specifically, Terrien demonstrates that it was known in the art prior to the critical date with respect to a process for hydrogen generated from shifted syngas to follow a first step of hydrogen separation by PSA (unit **9**) with a step of cryogenic CO₂ separation (**8**) and thereafter to perform at least one step of H₂ membrane separation (**11**) to generate a second hydrogen rich gas stream (**12**). As noted above, Terrien

teaches that this second hydrogen rich gas stream can be recycled as feed to a PSA unit or as fuel to a reformer unit, for example, to improve efficiency of hydrogen and carbon dioxide recovery.

**X. FACTS AND OPINIONS RELEVANT TO THE
UNPATENTABILITY GROUNDS ASSERTED IN PETITION I
AND PETITION II**

104. It is my firm belief and opinion that at least claims 1, 11 and 12 of the '805 Patent are anticipated by Reinertsen, because the reference discloses each of the limitations of those patent claims, either explicitly or inherently.

105. It is also my firm belief and opinion that at the time of the alleged invention in the timeframe just prior to the critical date, at least claims 1, 2, 3, 4, 5, 6, 11 and 12 of the '805 Patent would have been obvious to a POSA. Prior to the critical date, a POSA would have known that prior art methane reformer and separation processes, such as described in Reinertsen, Rytter, Martin, Darde, Rafati, Gauthier and Terrien could have readily been modified in obvious ways that would clearly satisfy the limitations of the '805 Patent claims, such as by combining a dual reformer configuration, for example a GHR combined with an ATR, with downstream separation processes using PSA and cryogenic CO₂ separation for the well-recognized purpose and goal of producing hydrogen by methane reforming while reducing carbon dioxide emissions. For example, a POSA would have been motivated to make such a combination of a dual reformer configuration with downstream separation processes using PSA and

cryogenic CO₂ separation to achieve hydrogen production with a purified coproduct of carbon dioxide and/or to reduce environmental impacts of hydrogen generation from hydrocarbons by providing an effective means of separating and capturing the carbon dioxide generated in the process. My specific opinions regarding particular combinations of references, as provided in the two Petitions, are given below.

XI. INVALIDITY ANALYSIS FOR PETITION

A. Petition I Ground 1: Claims 1, 11 and 12 are Anticipated by Reinertsen

106. As discussed in more detail below, Reinertsen discloses each and every element of independent claim 1 and dependent claims 11 and 12. In my opinion, therefore, Reinertsen anticipates claims 1, 11 and 12 of the '805 Patent.

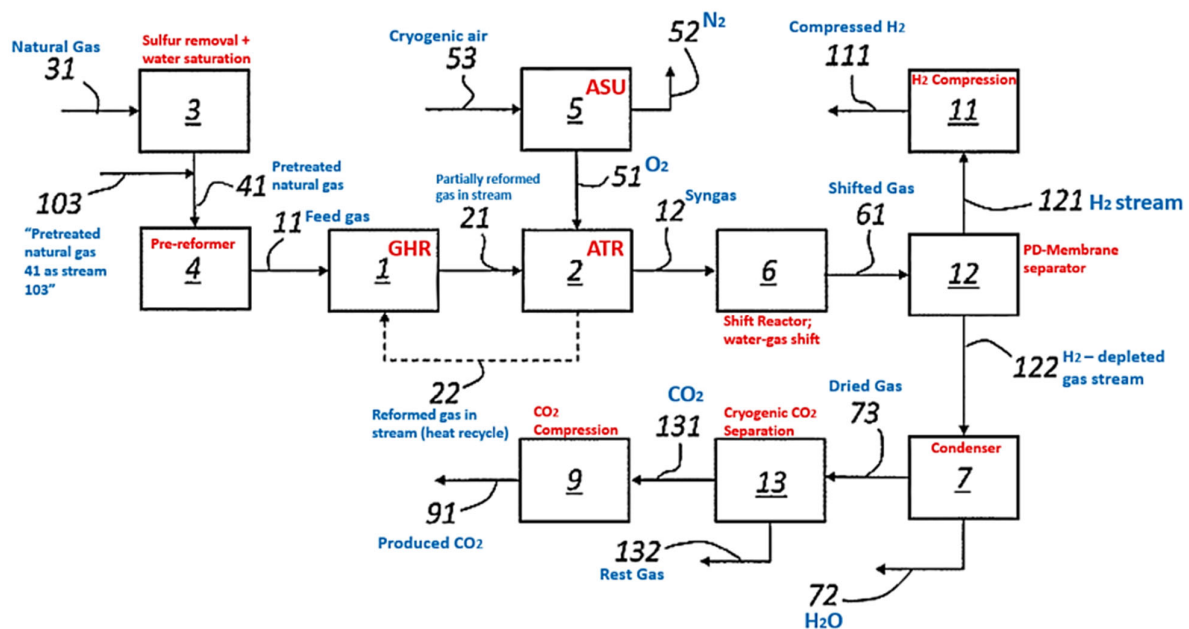
107. I provide discussion below regarding my analysis.

1. Element 1.pre: Preamble

108. The preamble of claim 1 recites “[a] process for preparing hydrogen by reforming hydrocarbons with steam, and for separation of carbon dioxide.” It is my opinion that Reinertsen discloses a process for preparing hydrogen by reforming hydrocarbons with steam, and for separation of carbon dioxide.

109. Reinertsen discloses a process for producing hydrogen by reforming hydrocarbon, such as natural gas, with steam and for separation of carbon dioxide. EX1005, FIGURE 4, ¶¶[0005], [0064], [0109]; EX1022, p.4,

ln.10–20, p.10, ln.4–9, p.20, ln.24–p.21, ln.2; EX1023, p.4, ln.9–20, p.10, ln.10–15, p.21, ln.5–12. For example, Reinertsen states that “FIGURE 4 shows a method for production of hydrogen from natural gas with separation of CO₂ according to embodiment example 2.” EX1005 ¶[0109]; EX1022, p.20, ln.24–25; EX1023, p.21, ln.5–6. Reinertsen further discloses providing a feed gas stream, where the feed gas stream contains a hydrocarbon component and steam, where the hydrocarbon component contains natural gas which includes at least methane. EX1005, ¶[0017], [0102], [0067]; EX1022, p.5, ln.17–18, p.18, ln.24–p.19, ln.4, p.10, ln.18–20; EX1023, p.5, ln.17–18, p.19, ln.4–14, p.10, ln.24–28.



EX1005, FIGURE 4, annotated

110. FIGURE 4 of Reinertsen shows syngas production, by the ATR 2, GHR 1 and with oxygen provided by the ASU 5, and natural gas treatment by removing sulfur 3 and pre-reforming 4, and water-gas shift 6, which are further

described in FIGURES 1 and 2. EX1005, ¶[0109]; EX1022, p.20, ln.24–25; EX1023, p.21, ln.5–6. The process of FIGURE 4 also includes cryogenic separation of CO₂ **13**. EX1005, ¶[0110]; EX1022, p.21, ln.3–5; EX1023, p.21, ln.13–15. As noted above, Reinertsen further specifically discloses providing a feed gas stream, where the feed gas stream contains a hydrocarbon component and steam. EX1005, ¶[0067]; EX1022, p.10, ln.18–20; EX1023, p.10, ln.24–28. Therefore, the process exemplified in FIGURE 4 of Reinertsen, provides an example of a process for preparing hydrogen by reforming hydrocarbons with steam, and for separation of carbon dioxide as recited in claim 1 of the '805 Patent.

1. Element 1.1: providing a feed gas stream

111. Element 1.1 recites a step of “providing a feed gas stream”. It is my opinion that Reinertsen discloses providing a feed gas stream.

112. Reinertsen describes, “The feed gas **11** first passes through the catalyst in the GHR **1**, then the partially reformed gas in stream **21** passes through the ATR **2**, and finally the reformed gas in stream **22** passes through the heating side of the GHR **1** to provide the heat for the initial reaction.” EX1005, ¶[0074]; EX1022, p.12, ln.14–23; EX1023, p.12, ln.20–29. Reinertsen further demonstrates providing a feed gas in the following schematic at feed gas **11**. Therefore, the feed gas passed through the GHR in Reinertsen is an example of providing a feed gas stream as recited in claim 1 of the '805 Patent.

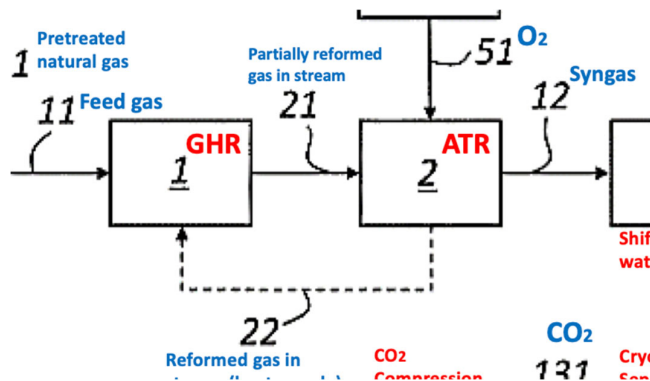


FIGURE 4, excerpt

2. Element 1.1.1: hydrocarbon component and steam

113. Element 1.1.1 recites “wherein the feed gas stream comprises a hydrocarbon component and steam”. It is my opinion that Reinertsen discloses a feed gas stream comprising a hydrocarbon and steam.

114. Reinertsen discloses the feed gas stream comprises a hydrocarbon component and steam. EX1005, Claim 1, ¶[0010], ¶[0017], ¶[0067]; EX1022, p.4, ln.30–p.5, ln.4, p.5, ln.17–18, p.10, ln.18–20; EX1023, p.4, ln.26–p.5, ln.2, p.5, ln.17–18, p.10, ln.24–28. Reinertsen explicitly discloses a hydrocarbon component in the feed gas stream. For example, Claim 1 of EX1005 recites “receiving a feed gas comprising hydrocarbons” and ¶[0017] discloses that “the feed gas is a hydrocarbon-rich gaseous stream.” *See*, also EX1022, Claim 1, p.5, ln.17; EX1023, Claim 1, p.5, ln.17.

115. Reinertsen further discloses the inclusion of steam in the feed gas stream. Reinertsen expressly discloses that steam is used in the reformer process by referring to use of a “steam reformer” and by describing the chemical

reactions, including steam reforming, that are involved in production of synthesis gas. EX1005, ¶¶[0054], [0066]–[0067]; EX1022, p.10, ln.12–20; EX1023, p.10, ln.18–28. Additionally, Reinertsen discloses that the feed gas can be saturated with water before pre-reforming. EX1005, ¶[0010]; EX1022, p.5, ln.1–2; EX1023, p.4, ln.30–p.5, ln.1. When the water-saturated feed gas is heated as required by the processes described, such as a GHR (EX1005, ¶[0055]), a POSA would have understood that the temperature of these processes are sufficiently high that steam is necessarily present as a part of the feed gas stream. Further, a POSA would know that steam must be a component of the feed gas of the disclosed GHR and ATR processes, because steam makes up one of the reactants for these processes. See my discussion in ¶¶29 and 30 *supra*. Reinertsen clearly teaches this, for example, by referring to the chemical reaction “ $\text{CH}_4 + \text{H}_2\text{O} \rightleftharpoons \text{CO} + 3\text{H}_2$ ” of the reforming process as “Steam reforming.” EX1005, ¶[0067]; EX1022, p.8, ln.16–26; EX1023, p.8, ln.19–29. Reinertsen’s description of the processes of the invention requires the presence of steam in the feed gas and a POSA would clearly recognize in view of that description that steam must necessarily be present in the feed gas. The feed gas passed through the heated GHR and ATR, which is treated by saturation with water or addition of vaporized water (steam), in Reinertsen is an example of a feed gas comprising a hydrocarbon component and steam as recited in claim 1 of the ’805 Patent.

3. Element 1.1.2: methane

116. Element 1.1.2 recites “wherein the hydrocarbon component comprises at least methane.” It is my opinion that Reinertsen discloses a feed gas stream with a hydrocarbon component comprising at least methane.

117. Reinertsen discloses a feed gas stream with a hydrocarbon component containing methane. EX1005, ¶[0064]; EX1022, p.10, ln.4–7; EX1023, p.10, ln.10–13. Specifically, Reinertsen discloses that “(e)mbodiments include receiving a supply of natural gas, or more generally a hydrocarbon containing gas from any source.” EX1005, ¶[0064]; EX1022, p.10, ln.4–7; EX1023, p.10, ln.10–13. Reinertsen also discloses that “natural gas may be cleaned and pre-treated in a suitable manner so that the gas feed mainly comprises methane after treatment.” EX1005, ¶[0064]; EX1022, p.10, ln.4–7; EX1023, p.10, ln.10–13. It is general knowledge that methane is the main component of natural gas. Further, I emphasize that a POSA has understood long before the critical date that synthesis gas was prepared by certain reactions of methane, as I have described above. Therefore, the feed gas passed through the GHR and ATR in Reinertsen, such as natural gas, is an example of a feed gas having a hydrocarbon component containing methane as recited in claim 1 of the ’805 Patent.

4. Element 1.2: endothermic reforming step

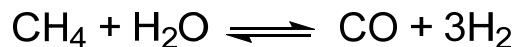
118. Element 1.2 recites a step of “reforming at least a portion of the feed gas stream in an endothermic reforming step over a reforming catalyst

thereby producing a first synthesis gas stream”. It is my opinion that Reinertsen discloses reforming at least a portion of the feed gas stream in an endothermic reforming step over a reforming catalyst thereby producing a first synthesis gas stream.

119. Reinertsen discloses a process including an endothermic reforming step over a reforming catalyst using a GHR for reforming a feed gas to produce a first synthesis gas stream. EX1005, 1 of FIGURE 1, 1 of FIGURE 4, and ¶¶ [0067]–[0068], [0074]; EX1022, p.10, ln.18–22, p.12, ln.14–23; EX1023, p.10, ln.24–p.11, ln.3, p.12, ln. 20–29.

120. In more detail, Reinertsen discloses producing a first synthesis gas stream (partially reformed gas in stream **21** of FIGURE 4) from an endothermic reforming step (GHR 1 of FIGURE 4). Reinertsen describes the “steam reforming” chemical reaction “ $\text{CH}_4 + \text{H}_2\text{O} \rightleftharpoons \text{CO} + 3\text{H}_2$ ” of the feed gas stream reforming process as being “strongly endothermic.” EX1005, ¶¶[0067]–[0068]; EX1022, p.10, ln.18–22; EX1023, p.10, ln.24–p.11, ln.3. Reinertsen further discloses that this endothermic reforming step is performed in a gas heated reformer (GHR) using a catalyst. EX1005, ¶[0074]; EX1022, p.12, ln.14–23; EX1023, p.12, ln. 20–29. Reinertsen specifically discloses that heat must be provided for the reforming reactions and that heat from the ATR is provided to heat the initial reaction in the GHR. EX1005, ¶[0074]; EX1022, p.12, ln.14–23; EX1023, p.12, ln. 20–29. I also emphasize that a POSA would have understood

long before the critical date that a GHR process to make synthesis gas includes at least the endothermic steam reforming reaction (Reaction 1, above):



which is also disclosed in Reinertsen. EX1005, ¶¶[0067]–[0068]; EX1022, p.10, ln.18–22; EX1023, p.10, ln.24–p.11, ln.3. Therefore, the GHR process in Reinertsen is an example of an endothermic reforming step over a reforming catalyst as recited in claim 1 of the '805 Patent.

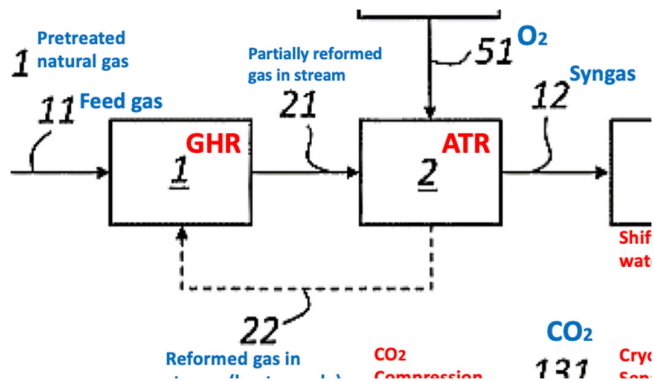


FIGURE 4, excerpt

5. Element 1.2.1: first synthesis gas

121. Element 1.2.1 recites “wherein the first synthesis gas stream comprises hydrogen, carbon monoxide, carbon dioxide, and unreacted methane”. It is my opinion that Reinertsen discloses a first synthesis gas stream comprising hydrogen, carbon monoxide, carbon dioxide, and unreacted methane.

122. The first synthesis gas (SG1) is produced in an endothermic reforming step. As described above, Reinertsen describes an endothermic

reforming step in a GHR. Reinertsen describes “[t]he gas mixture from the reformer reactor contains mainly the gas components CO, H₂, H₂O, CO₂ and some CH₄.” EX1005, ¶[0076]; EX1022, p.12, ln.24–26; EX1023, p.13, ln.3-5. Moreover, Reinertsen describes chemical reactions that occur during the production of synthesis gas by reforming of natural gas (methane). EX1005, ¶[0067]; EX1022, p.10, ln.18–20; EX1023, p.10, ln.24–28.

123. The GHR of Reinertsen therefore naturally results in conversion of a portion of the reactants, methane (CH₄) and steam (H₂O), into reaction products carbon monoxide (CO) and hydrogen (H₂), as well as some carbon dioxide which will always be present to at least some degree in view of the various reactions that accompany methane reforming, ¶30, *supra*. Given that the steam reforming reaction is an equilibrium controlled reaction, both reaction products carbon monoxide (CO) and reagents hydrogen (H₂) and reactants methane (CH₄) and steam (H₂O) are necessarily present in the output of the GHR process to at least some degree. At least, because steam reforming is accompanied by some level of the water-gas shift reaction, CO₂ will also be present, ¶28, *supra*. Thus, to the extent Reinertsen may not expressly disclose the composition of the first synthesis gas stream as including the gases enumerated in the claim, the composition of gases is at least inherently present in the first synthesis gas stream in view of Reinertsen’s disclosure of the feed gas used and the process used to make the first synthesis gas stream.

Considering the reactants methane (CH_4) and steam (H_2O) to the GHR, the law of constant atoms of species C, H and O forces that only components which consist of these species form the synthesis gas at the outlet of the GHR: reaction products H_2 , CO and CO_2 and any unreacted reactants CH_4 and H_2O . The extent of maximal conversion to products is determined by the chemical equilibrium of each chemical reaction. This is a natural law and is based on the second law of thermodynamics. At the end the chemical equilibrium is determined by the inlet conditions to the GHR as well as the pressure and temperature at the outlet. The activity of the catalyst in the GHR determines how close the actual approach to chemical equilibrium conversion is, but it can never be higher than the value determined by chemical equilibrium. In my opinion, therefore, the synthesis gas output of the GHR process disclosed in Reinertsen would necessarily include hydrogen, carbon monoxide, carbon dioxide, and unreacted methane as recited in claim 1 of the '805 Patent.

6. Element 1.3: reforming the first synthesis gas

124. Element 1.3 recites a step of “reforming the first synthesis gas stream in an autothermal reforming step thereby producing a third synthesis gas stream”. It is my opinion that Reinertsen discloses reforming the first synthesis gas stream in an autothermal reforming step thereby producing a third synthesis gas stream, and therefore this element of claim 1.

125. Reinertsen discloses a process including an autothermal reforming step using an ATR for reforming the first synthesis gas stream from the endothermic reforming step to produce a third synthesis gas stream. EX1005, 2 and stream 12 of FIGURE 4, and ¶[0070].

126. Reinertsen discloses reforming the first synthesis gas stream in an autothermal reforming step thereby producing a third synthesis gas stream. EX1005, FIGURE 4, ¶[0070], [0074]; EX1022, p.11, ln.10–22; EX1023, p.11, ln.17–29. As shown in FIGURE 4 excerpt below, Reinertsen discloses a series arrangement passing a reformed gas (“partially reformed gas in stream 21”) through an autothermal reformer (“ATR 2”) to produce a new synthesis gas stream (“syngas 12”). EX1005, ¶[0074]; EX1022, p.12, ln.18–21; EX1023, p.12, ln.24–27. Therefore, the ATR process in Reinertsen is an example of a process for reforming a synthesis gas stream in an autothermal reforming step as recited in claim 1 of the ’805 Patent.

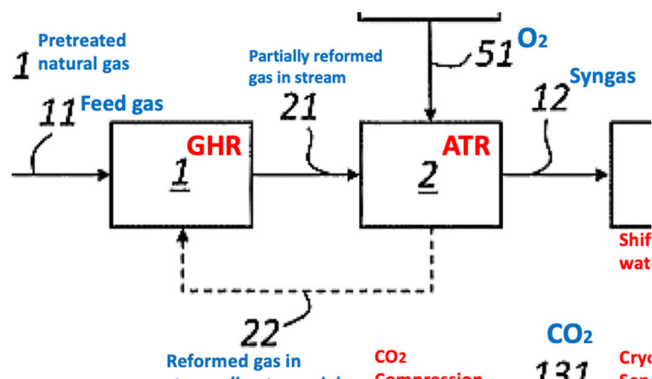


FIGURE 4, excerpt, annotated

7. Element 1.3.1: exothermic partial oxidation and endothermic reforming

127. Element 1.3.1 recites “wherein the autothermal reforming step comprises exothermic partial oxidation and endothermic reforming with steam over a reforming catalyst”. It is my opinion that Reinertsen discloses the autothermal reforming step comprising exothermic partial oxidation and endothermic reforming with steam over a reforming catalyst.

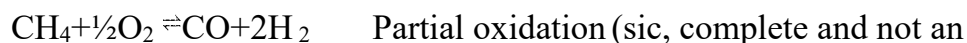
128. Reinertsen discloses the use of an ATR, which by its nature operates in this manner. EX1005, ¶[0068]; EX1022, p.10, ln.20–22; EX1023, p.11, ln.1–3 (“The heat of reaction for the strongly endothermic steam reforming can be provided either by external heating, as in a steam reformer, or by partial oxidation in an autothermal reformer”). EX1005, ¶[0070]; EX1022, p.11, ln.10–22; EX1023, p.11, ln.17–29 (“In an ATR, natural gas (methane) is fed together with oxygen or air into a combustion chamber. The energy which is required to operate the endothermic steam reforming reactions is provided by the exothermic reactions between hydrocarbons and/or hydrogen and oxygen. The temperature in the combustion chamber can reach more than 2000° C. After the combustion chamber the reactions may be driven to equilibrium over a catalyst bed before the synthesis gas leaves the reactor at approximately 1000–1050° C”). Reinertsen also expressly describes the use of a steam reformer with an autothermal reformer. EX1005, ¶[0072]; EX1022, p.11, ln.26–27; EX1023, p.12, ln.1–2 (“Reforming of natural gas can also be made by combined

reforming (CR) which is a combination of a steam reformer (SR) and an autothermal reformer (ATR”). Therefore, the ATR process in Reinertsen is an example of a process including exothermic partial oxidation and endothermic reforming with steam over a reforming catalyst as provided in claim 1 of the ’805 Patent.

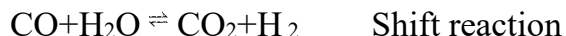
8. Element 1.3.2: second and third synthesis gas

129. Element 1.3.2 recites “wherein the second synthesis gas stream and the third synthesis gas stream comprise hydrogen, carbon monoxide, carbon dioxide, and unreacted methane”. It is my opinion that Reinertsen discloses the third synthesis gas stream comprising hydrogen, carbon monoxide, carbon dioxide, and unreacted methane. I note that Reinertsen only discloses the series arrangement of reformers, a second synthesis gas as described in the ’805 Patent resulting from a parallel reformer configuration is not present.

130. Reinertsen discloses the chemical reactions for production of synthesis gas streams produced from an ATR, alone or in combination with a GHR, and water gas shift processes showing first, third and fourth synthesis gas stream components of hydrogen, carbon monoxide, carbon dioxide and unreacted methane. EX1005, ¶¶[0067]–[0068]; EX1022, p.10, ln.18–22; EX1023, p.10, ln.24–p.11, ln.3:



equilibrium controlled chemical reaction)



131. Reinertsen shows hydrogen, carbon monoxide, and carbon dioxide as products of these reactions. Unreacted methane is also necessarily present in the synthesis gas streams of Reinertsen due to the thermodynamic constraints of the equilibrium condition for the steam reforming reaction as shown by the double arrow (equilibrium) in the steam reforming reaction in Reinertsen, which limits this reaction from going to completion. The partial oxidation reaction is not equilibrium controlled (the reaction is shown incorrectly in equilibrium in Reinertsen) which leads to completed reaction. Oxygen supplied to the ATR is limited such that not all methane can be fully oxidized to carbon dioxide and water (¶¶ 29, 34, 64, *supra*). Thus, the conversion of O₂ is complete, but unconverted CH₄ remains after the non-catalytic partial oxidation section of the ATR.

132. Reinertsen discloses syngas stream **12** of FIGURE 4 which exits the reformer and feeds into the shift reactor **6** of FIGURE 4. The syngas of FIGURE 4 thus comprises hydrogen, carbon monoxide, carbon dioxide, and unreacted methane. The gas mixture from the reformer reactor “contains mainly the gas components CO, H₂, H₂O, CO₂ and some CH₄.” EX1005, ¶[0076]; EX1022, p.12, ln.24–28; EX1023, p.13, ln.3–8. As I have explained above, and as shown correctly in FIGURE 1 of Reinertsen, syngas **12** should exit the GHR

in the series configuration of the GHR and ATR shown when direct heat transfer is employed, as is preferred. The illustration of syngas **12** exiting the ATR in Figure 4 is schematic. Syngas stream **12** (FIGURE 4, excerpt below) is the combined synthesis gas stream formed in the endothermic (GHR) reforming step and the autothermal reforming step, the same as SG3 in the '805 Patent.

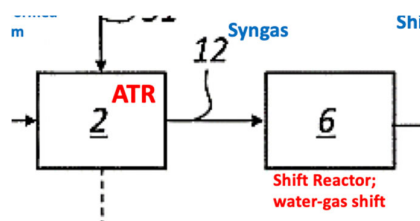


FIGURE 4, excerpt, annotated

133. The ATR process of Reinertsen, therefore, naturally results in conversion of a portion of the methane (CH_4) and steam (H_2O) as reactants into carbon monoxide (CO), carbon dioxide (CO_2) and hydrogen (H_2) as reaction products. Given that the steam reforming reaction is an equilibrium controlled reaction, both reaction products carbon monoxide (CO) and hydrogen (H_2) and reactants methane (CH_4) and steam (H_2O) are necessarily present in the output of the ATR process to at least some degree. Thus, to the extent Reinertsen may not expressly disclose the composition of the first synthesis gas stream as including the gases enumerated in the claim, such composition of gases are at least inherent in the processes disclosed by Reinertsen. I also emphasize that a POSA would have understood at least at the critical date, in view of the process description of Reinertsen and what was known in the art, that the enumerated

components would have been present in syngas stream **12**. In my opinion, therefore, the synthesis gas output of the ATR, provided in a series configuration with a GHR process, disclosed in Reinertsen would necessarily include hydrogen, carbon monoxide, carbon dioxide, and unreacted methane.

9. Element 1.3.3: heat generated by the autothermal reforming step

134. Element 1.3.3 recites a limitation of “wherein heat generated by the autothermal reforming step is utilized for heating in the endothermic reforming step of step (b)”. It is my opinion that Reinertsen discloses that heat generated by the autothermal reforming step is utilized for heating in the endothermic reforming step of step (b), and therefore discloses this element of claim 1.

135. Reinertsen discloses endothermic reforming via a GHR and an autothermal reforming process via an ATR provided in a series configuration, where synthesis gas from the autothermal reforming steps is provided to the ATR for autothermal reforming. *See, e.g.,* EX1005, **1**, **2** and **22** of FIGURES 1 and 4, ¶[0074]; EX1022, p.12, ln.18–21; EX1023, p.12, ln.24–27. Reinertsen discloses endothermic reforming via GHR and autothermal reforming ATR processes, where heat generated (e.g., heated gas) from the ATR is utilized for heating the GHR. EX1005, **1**, **2** and **22** of FIGURES 1 and 4, ¶[0074], [0102]; EX1022, p.12, ln.18–21, p.18, ln.24–p.19, ln.4; EX1023, p.12, ln.24–27, p.19, ln.4–14.

136. Reinertsen discloses heat (via recycled exit gas **22** from the ATR **2**, FIGURE 4 excerpt below) “passes through the heating side of the GHR **1** to provide heat for the initial reaction”. EX1005, ¶¶[0074], [0102]; EX1022, p.12, ln.18–21, p.18, ln.24–p.19, ln.4; EX1023, p.12, ln.24–27, p.19, ln.4–14 (“The heat recycle **22** is the exit gas from the ATR used to heat the GHR”).

Therefore, the thermally coupled ATR and GHR in Reinertsen is an example of a dual reformer configuration where heat generated by an autothermal reforming step (i.e. the ATR) is utilized for heating in the endothermic reforming step (i.e. the GHR) as provided in claim 1 of the '805 Patent.

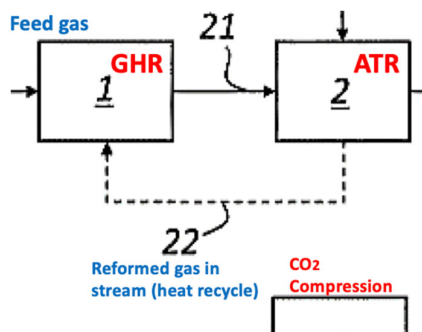


FIGURE 4, excerpt

10. Element 1.4: converting the carbon monoxide

137. Element 1.4 recites a step of “(d) converting the carbon monoxide present in the third synthesis gas stream with steam thereby producing hydrogen and carbon dioxide thereby producing a fourth synthesis gas stream”. It is my opinion that Reinertsen discloses converting the carbon monoxide present in a third synthesis gas stream with steam thereby producing hydrogen and carbon dioxide thereby producing a fourth synthesis gas stream.

138. Reinertsen discloses converting the carbon monoxide present in the third synthesis gas stream with steam conversion via water gas shift reaction, thereby producing hydrogen and carbon dioxide and a fourth synthesis gas stream. EX1005, 6 of FIGURE 4, ¶¶[0067], [0103]; EX1022, p.10, ln.18–20, p.19, ln.5–21; EX1023, p.10, ln.24–28, p.19, ln.15–31. Reinertsen also explicitly discloses the shift gas reaction providing conversion “ $\text{CO} + \text{H}_2\text{O} \rightleftharpoons \text{CO}_2 + \text{H}_2$ Shift reaction.” EX1005, ¶[0067]; EX1022, p.10, ln.18–20; EX1023, p.10, ln.24–28.

139. In more detail, Reinertsen describes using a water-gas shift reactor 6 of FIGURE 4 (excerpt below) to convert carbon monoxide in a third synthesis gas stream using steam. Syngas **12** (i.e., the third synthesis gas stream) is “shifted to increase the content of hydrogen and CO_2 in one or more shift reactors 6, i.e. water-gas-shift reactor(s) 6, to produce the shifted gas **61**.” EX1005, ¶[0103]; EX1022, p.19, ln.5–9; EX1023, p.19, ln.15–19. Reinertsen further adds that “(s)team may be added to the gas mixture before” it enters the shift reactor. EX1005, ¶[0103]; EX1022, p.19, ln.5–9; EX1023, p.19, ln.15–19. The addition of steam is also described as increasing the efficiency of the shift reaction. EX1005, ¶[0103]; EX1022, p.19, ln.5–9; EX1023, p.19, ln.15–19. Reinertsen discloses that the result of this shift produces hydrogen and carbon dioxide in a shifted gas **61** of FIGURE 4 (i.e., a fourth synthesis gas stream). Therefore, the water gas shift process in Reinertsen is an example of a process

for converting the carbon monoxide present in a synthesis gas stream (i.e., a third syngas stream) with steam for producing hydrogen and carbon dioxide in a shifted syngas stream (i.e., a fourth syngas stream) as recited in claim 1 of the '805 Patent.

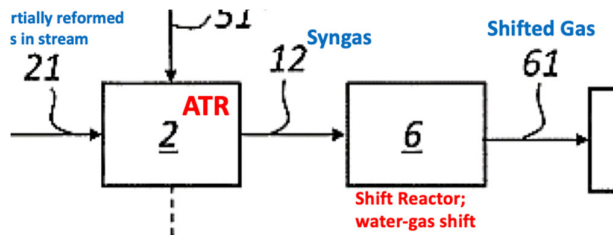
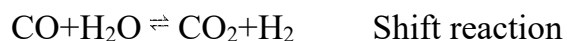
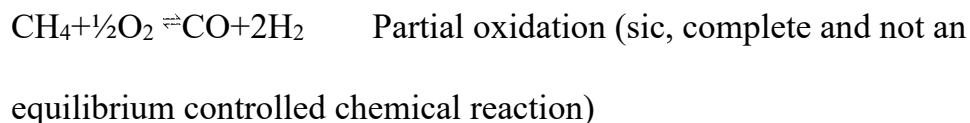
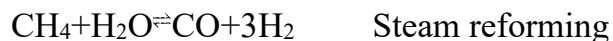


FIGURE 4, excerpt, annotated

11. Element 1.4.1: fourth synthesis gas

140. Element 1.4.1 recites “wherein the fourth synthesis gas stream comprises hydrogen, carbon dioxide, unreacted methane, and carbon monoxide unconverted in step (d).” It is my opinion that Reinertsen discloses a fourth synthesis gas stream comprising hydrogen, carbon dioxide, unreacted methane, and carbon monoxide unconverted in step (d).

141. As described above, Reinertsen (EX1005, ¶[0067]; EX1022, p.10, ln.18–20; EX1023, p.10, ln.24–28) discloses the chemical reactions for production of synthesis gas streams produced from combined GHR, ATR and water gas shift processes showing first, third and fourth synthesis gas stream components of hydrogen, carbon monoxide, carbon dioxide and unreacted methane:



142. Reinertsen shows hydrogen, carbon monoxide, and carbon dioxide as products of these reactions. Any methane that is unreacted by steam reforming and partial oxidation is also necessarily present in the synthesis gas streams of Reinertsen. The thermodynamic constraints of the equilibrium condition for the steam reforming reaction (as shown by the double arrow in the steam reforming reaction above and as was known in the art), limit this reaction and prevent it from going to completion. The partial oxidation reaction, in contrast, is not equilibrium controlled (the use of the double arrow in the partial oxidation reaction above was a technical/clerical error). Oxygen is supplied to the process in an under-stoichiometric ratio with respect to full methane oxidation (as explained ¶¶29, 34, 64, *supra*). Thus, O₂ is completely converted, but unconverted CH₄ remains after the non-catalytic partial oxidation section of the ATR.

143. Reinertsen discloses “performing a water-gas-shift process on the reformed gas so as to generate a shifted gas comprising hydrogen and carbon dioxide”. EX1005, ¶[0005]; EX1022, p.4, ln.16-18; EX1023, p.4, ln.12-14. Reinertsen further discloses that the shifted gas contains carbon monoxide,

carbon dioxide, methane and hydrogen. EX1005, ¶[0085]; EX1022, p.15, ln.18–20; EX1023, p.15, ln.28–30.

144. FIGURE 2 (comparative example) of Reinertsen describes a flow sheet for production of hydrogen and CO₂ by combination of an ATR (2) and GHR (1) to generate syngas (**12**) which is then shifted to produced shifted syngas (**61**). EX1005, ¶¶[0101]–[0103]; EX1022, p.18, ln.21–p.19, ln.7; EX1023, p.19, ln.1–17. Reinertsen further discloses that the rest gas (**102** of FIGURE 2), which is downstream from the shifted gas (**61** of FIGURE 2) in the process, “contains remnants of CO and CH₄”, indicating that unreacted methane and unconverted carbon monoxide are present in the shifted gas (**61**) of FIGURE 2. EX1005, ¶[0103]; EX1022, p.19, ln.5–21; EX1023, p.19, ln.15–31. Shifted gas (**61**) of FIGURE 2 is produced using the same process as shifted gas (**61**) of FIGURE 4. EX1005, FIGURES 2 and 4; EX1022, FIGURES 2 and 4; EX1023, FIGURES 2 and 4. Reinertsen thus indicates that unreacted methane and unconverted carbon monoxide are present in the shifted gas (**61**) of FIGURE 4.

145. The water gas shift (WGS) reaction of Reinertsen results in conversion of a portion of the carbon monoxide (CO) and steam (H₂O) as reactants into carbon dioxide (CO₂) and hydrogen (H₂) as reaction products. This was and is well known in the art. Given that the WGS reaction is an equilibrium controlled reaction, both reaction products carbon monoxide (CO)

and reagents hydrogen (H₂) and carbon dioxide (CO₂) are necessarily present in the output of the water gas shift reaction process. Thus, to the extent Reinertsen may not expressly disclose the composition of the first synthesis gas stream as including the gases enumerated in the claim, such composition of gases is at least inherent in the process description of Reinertsen. In my opinion, therefore, the output of the water gas shift reaction process disclosed in Reinertsen necessarily includes hydrogen, carbon dioxide, unreacted methane, and carbon monoxide unconverted as recited in claim 1 of the '805 Patent.

12. Element 1.5: separating hydrogen

146. Element 1.5 recites a step of “(e) separating hydrogen from the fourth synthesis gas stream by pressure swing adsorption, thereby producing a first hydrogen-rich stream and a first residual gas stream.” It is my opinion that Reinertsen discloses separating hydrogen from a fourth synthesis gas stream by pressure swing adsorption, thereby producing a first hydrogen-rich stream and a first residual gas stream, and therefore discloses this element of claim 1.

147. Reinertsen discloses separating hydrogen from shifted syngas stream **61** (i.e., the fourth synthesis gas stream) to produce a first hydrogen-rich stream and a first residual gas stream, where hydrogen separation is provided upstream of cryogenic CO₂ separation. EX1005, **12** and **13** of FIGURE 4, ¶¶[0103], [0107], [0109]–[0110]; EX1022, p.19, ln.5–21, p.20, ln.12–16, p.20, ln.24–29, p.21, ln.3–5; EX1023, p.19, ln.15–31, p.20, ln.23–27, p.21, ln.5–15.

In the process illustrated in FIGURE 4 (excerpt below), the “shifted gas” enters “a hydrogen separation vessel that comprises a Pd-membrane 12.” EX1005, ¶[0106]; EX1022, p.20, ln.6–7; EX1023, p.20, ln.17–18. The Pd-membrane 12 (i.e., a hydrogen separation membrane) separates a “hydrogen stream 121 and a hydrogen-depleted gas stream 122.” EX1005, ¶[0107]; EX1022, p.20, ln.12–16; EX1023, p.20, ln.23–27. The hydrogen depleted stream 122 is dried and the dried stream 73 is introduced into cryogenic separation 13 to separate CO₂ stream 131 and rest gas 132. EX1005, ¶[0107],[0110]; EX1022, p.20, ln.4-14, p.21, ln.3-5; EX1023, p.20, ln. 15-27; p.21, ln.113-15.

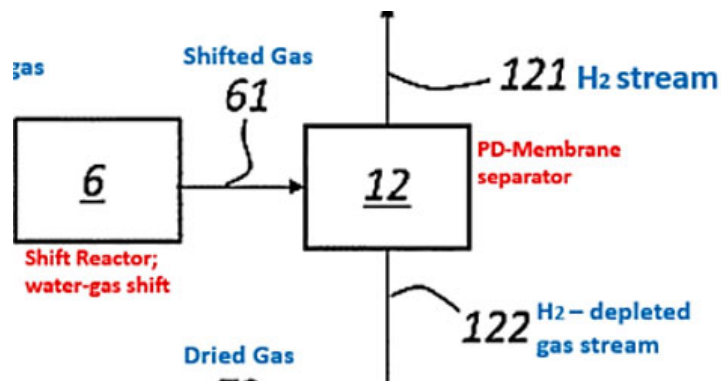


FIGURE 4, excerpt, annotated

148. However, Reinertsen describes that a pressure swing adsorption (PSA) process may be used in place of the Pd-membrane. EX1005, ¶[0012]; EX1023, p.5, ln.10 (“Preferably, the hydrogen separation process comprises a PSA process”); EX1005, ¶[0063]; EX1023, p.10, ln.7-9 (“Embodiments also include using a PSA process, or PSA processes, to separate hydrogen from the

reformed natural gas; or more generally from a reformed gas containing hydrocarbons.”); EX1005, ¶[0081] (“If a different to Pd-membrane hydrogen separation process is used, like PSA, it still might be feasible to operate only one WGS reactor, e.g., an isothermal shift reactor.”); and EX1005, ¶[0085];; EX1023, p.15, ln.21–31 (“Embodiments may alternatively use PSA to separate hydrogen from the gas output from the WGS reactor. PSA is a technology used to separate some gas species from a mixture of gases under pressure according to the species' molecular characteristics and affinity for an adsorbent material”). Additionally, Reinertsen teaches that water is condensed before the PSA unit, to avoid overloading the adsorbents. EX1005, ¶[0085]; EX1022, p.15, ln.14–21; EX1023, p.15, ln.24–31 (“It operates at near-ambient temperatures and differs significantly from cryogenic distillation techniques of gas separation. Specific adsorptive materials (e.g., zeolites, activated carbon, molecular sieves, etc.) are used as a trap, preferentially adsorbing the target gas species at high pressure. The process then swings to low pressure to desorb the adsorbed material. In the present mixture of gases, CO, CO₂ and CH₄ are adsorbed, letting the hydrogen pass through at process pressure. Not to overload the adsorbent, water is condensed before the PSA unit. Alternatively, the temperature can swing instead of the pressure.”). In my opinion, a POSA at the critical date, in view of the descriptions in Reinertsen of the use of PSA hydrogen separation as an alternative to Pd-membrane separation, particularly as illustrated in FIGURE 4,

along with specific teaching that water is condensed before the PSA and in view of what was known in the art about hydrogen separation employing a PSA, would have considered PSA separation as a suitable alternative to the Pd-membrane separation of FIGURE 4. Therefore, the PSA process in Reinertsen is an example of separating hydrogen from a synthesis gas stream by pressure swing adsorption so as to produce a hydrogen-rich stream and a residual gas stream as provided in claim 1 of the '805 Patent.

13. Element 1.5.1: first residual gas

149. Element 1.5.1 recites a limitation of “wherein the first residual gas stream comprises carbon dioxide, carbon monoxide unconverted in step (d), hydrogen not separated off in step (e), and unreacted methane”. It is my opinion that Reinertsen discloses a first residual gas stream comprising carbon dioxide, carbon monoxide unconverted in step (d), hydrogen not separated off in step (e), and unreacted methane, and therefore this element of claim 1.

150. Reinertsen discloses that the input to the PSA uses a “mixture of gases, CO, CO₂, and CH₄” which are adsorbed to allow hydrogen to pass through. EX1005, ¶[0085]; EX1022, p.15, ln.18–19; EX1023, p.15, ln.28–30. Pressure swing adsorption (PSA) is a technique used to separate gas species on the basis of the affinity for an adsorbent material. In the PSA process for hydrogen as described in Reinertsen, components other than hydrogen are adsorbed whereas the major part of the hydrogen is allowed to pass, thereby providing a hydrogen rich gas stream. A residual gas is also produced that

necessarily will contain all of the CO₂, unconverted CO, and unreacted CH₄ supplied to the PSA. These components are almost completely adsorbed in the PSA process. Hence, almost all required high H₂ purities can be obtained in a PSA. However, a 100% separation of hydrogen is not practically achievable and, therefore, the residual gas from this process will necessarily include hydrogen to at least some degree. In my opinion, therefore, the output of the PSA process disclosed in Reinertsen necessarily includes carbon dioxide (CO₂), unconverted carbon monoxide (CO), hydrogen (H₂) not separated by the PSA, and unreacted methane (CH₄) as provided in claim 1 of the '805 Patent.

14. Element 1.6: separating carbon dioxide

151. Element 1.6 recites a step of “(f) separating carbon dioxide from the first residual gas stream obtained in step (e) by cryogenic carbon dioxide separation, thereby producing a first carbon dioxide-rich stream and a second residual gas stream RG2”. It is my opinion that Reinertsen discloses separating carbon dioxide from a first residual gas stream obtained in step (e) by cryogenic carbon dioxide separation, thereby producing a first carbon dioxide-rich stream and a second residual gas stream RG2, and therefore this element of claim 1.

152. Reinertsen discloses separating carbon dioxide from the first residual gas stream by cryogenic carbon dioxide separation. EX1005, **13** of FIGURE 4, ¶¶[0027], [0059]; EX1022, p.9, ln.14–16; EX1023, p.9, ln.16–19 (“Although embodiments may separate CO₂ by using an amine washing

process, or other process, embodiments preferably use cryogenic separation to separate CO₂. That is to say, the gas stream is cooled to a temperature, and at a pressure, where CO₂ is liquefied.”). Reinertsen discloses producing a first carbon dioxide-rich stream and a second residual gas stream. EX1005, **131** and **132** of FIGURE 4. Reinertsen necessarily provides for a residual gas stream from the cryogenic CO₂ separation comprising carbon monoxide unconverted in **6**, hydrogen not separated off in **12** and unreacted methane, because the cryogenic CO₂ separation process in Reinertsen, like that of the '805 Patent, operates on the principle that process gases have different condensation temperatures, and when CO₂ is condensed, CO, H₂ and CH₄ will largely remain in the gas phase to be released as residual gas. EX1005, FIGURE 4. While CO₂ is separated in cryogenic CO₂ separation, the separation is never 100%, and therefore, there will necessarily be CO₂ in the residual gas stream **132** that is not separated off.

153. In more detail, Reinertsen discloses that the dried gas (**73** of FIGURE 4, excerpt below) is obtained from drying unit (**7** of FIGURE 4) by condensation of water **72**. The dried gas **73** is supplied to a cryogenic CO₂ separation unit (**13**) to produce a CO₂ rich stream (**131**) and rest gas (**132**). EX1005, ¶¶[0109], [0110]; EX1022, p.21, ln.3-5; EX1023, p.21. ln.13-15. In addition, Reinertsen in FIGURE 3 shows that CO₂ can be separated from the dried gas with an amine separator **8**. “The dried gas **73** is then input into the

amine separator 8 that outputs CO₂ stream **81** and a rest gas **83**. CO₂ stream **81** is compressed 9, and the CO₂ depleted rest gas **83** may be used for providing energy in fired heater(s)”). EX1005, ¶[0107]; EX1022, p.20, ln.13–16; EX1023, p.20, ln.24–27. Reinertsen then also discloses “...the amine unit 8 of embodiment example 1 (FIGURE 3) is replaced with cryogenic separation 13 of CO₂ **131** from the dried gas **73**, giving the rest gas **132** for use in fired heater(s)”. EX1005, ¶[0110]; EX1022, p.21, ln.3–5; EX1023, p.21, ln.13–15. Therefore, the cryogenic separation process in Reinertsen (either as in FIGURE 4 or as in FIGURE 3 with replacement of 8 with 13) is an example of separating carbon dioxide by cryogenic carbon dioxide separation to produce a carbon dioxide-rich stream and residual gas stream as provided in claim 1 of the ’805 Patent.

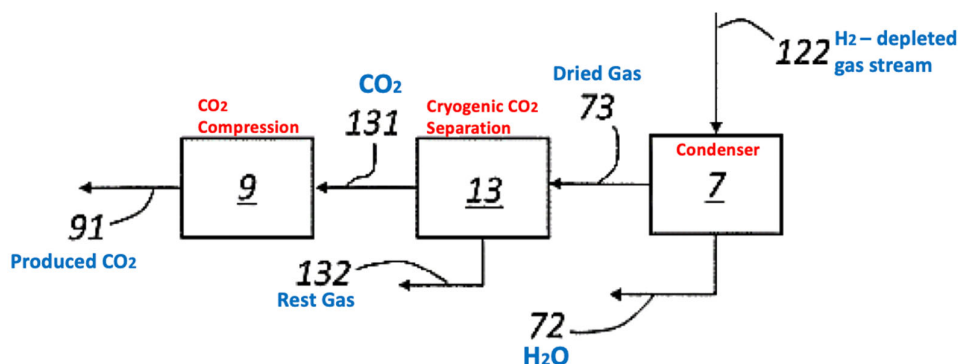


FIGURE 4, excerpt, annotated

15. Element 1.6.1: second residual gas

154. Element 1.6.1 recites a limitation of “wherein the second residual gas stream comprises carbon monoxide unconverted in step (d), hydrogen not

separated off in step (e), carbon dioxide not separated off in step (f), and unreacted methane”. It is my opinion that Reinertsen discloses a second residual gas stream comprising carbon monoxide unconverted in step (d), hydrogen not separated off in step (e), carbon dioxide not separated off in step (f), and unreacted methane, and therefore this element of claim 1.

155. Reinertsen discloses separating carbon dioxide from the first residual gas stream by cryogenic carbon dioxide separation. EX1005, **13** of FIGURE 4, ¶¶[0027], [0059]; EX1022, p.9, ln.14–16; EX1023, p.9, ln.16–19. Reinertsen discloses producing a first carbon dioxide-rich stream and a second residual gas stream. EX1005, **131** (CO₂) and **132** (Rest Gas) of FIGURE 4. Reinertsen necessarily provides for a residual gas stream from the cryogenic CO₂ separation that includes carbon monoxide that is unconverted in **6**, hydrogen not separated off in **12** and unreacted methane. EX1005, FIGURE 4. This residual composition including unconverted carbon monoxide, unseparated hydrogen and unreacted methane is present because the cryogenic CO₂ separation process in Reinertsen, like that of the ’805 Patent, operates on the principal that process gases have different condensation temperatures , and therefore, when CO₂ is condensed, carbon monoxide, hydrogen and methane provided to the cryogenic separation process will largely remain in the gas phase to be released as residual gas. EX1005, ¶¶[0027], [0059]; EX1022, p.9, ln.14–16; EX1023, p.9, ln.16–19.

156. Gaseous mixtures consisting of several components can be separated at cryogenic temperatures, for example, as achieved in Reinertsen using cryogenic CO₂ separation. In this process, if the boiling temperatures of the gaseous components of a mixture are far apart, cryogenic separation can be achieved by partial condensation. This principle of partial condensation is applied in Reinertsen to a mixture of CO₂, H₂, CO and CH₄. The boiling points of the noncondensing components H₂, CO and CH₄ (-253 °C, -191.5 °C, -161.5 °C, respectively at atm. pressure) are far apart from the condensing CO₂ (-78.5 °C, at atm. pressure). Although the boiling points are far apart, not all CO₂ will be condensed, and some of the lighter boiling gases will dissolve in the liquid CO₂.

157. Therefore, while CO₂ is separated in cryogenic CO₂ separation, the separation is never 100%, and therefore, there will necessarily be CO₂ in the residual gas stream that is not separated off. This property of the residual gas is explicitly acknowledged in Reinertsen which states “The rest gas **132** contains remnants of CO and CH₄ together with unseparated CO₂ and hydrogen”.

EX1005, FIGURE 4, ¶[0103]; EX1022, p.19, ln.17–18; EX1023, p.19, ln.27–28. In my opinion, therefore, the output of the PSA process disclosed in Reinertsen necessarily includes carbon dioxide, unconverted carbon monoxide, hydrogen not separated by the PSA, and unreacted methane as provided in claim 1 of the '805 Patent.

16. Element 11: compression step and cooling step

158. Element 11 recites a limitation of “[t]he process according to claim 1, where the first residual gas stream, for cryogenic carbon dioxide separation in step (f), is subjected to at least one compression step and at least one cooling step, thereby producing the first carbon dioxide-rich stream at least partly in the form of a condensed carbon dioxide stream.”. It is my opinion that Reinertsen discloses a first residual gas stream, for cryogenic carbon dioxide separation in step (f), is subjected to at least one compression step and at least one cooling step, thereby producing the first carbon dioxide-rich stream at least partly in the form of a condensed carbon dioxide stream, and therefore this element of claim 11.

159. I emphasize that cryogenic CO₂ separation was known in the art long before the critical date to include compression and cooling steps. See ¶40, supra; EX1018; EX1020; EX1021. More specifically, Reinertsen discloses “separating CO₂ by cryogenic cooling” (EX1005, ¶[0141]), and “[t]he CO₂ separation process may be a cryogenic process” (EX1005, ¶[0147]). Reinertsen further discloses, “[a]lthough embodiments may separate CO₂ by using an amine washing process, or other process, embodiments preferably use cryogenic separation to separate CO₂. That is to say, the gas stream is cooled to a temperature, and at a pressure, where CO₂ is liquefied.” EX1005, ¶[0059]; EX1022, p.9, ln.14–16; EX1023, p.9, ln.16–19. Thus, Reinertsen explicitly mentions a cooling step that is to be carried out at a pressure where CO₂ is

liquefied, which a POSA would view as corresponding to a compression step. In my opinion, a POSA at the critical date would have understood that the cryogenic CO₂ separation process of Reinertsen necessarily includes the at least one compression step and at least one cooling step of claim 11 of the '805 Patent. In addition, the terms "cryogenic cooling" and "cryogenic process", as used in Reinertsen, would have been readily understood by a POSA as examples of a process including cooling and condensation steps of a gas stream containing CO₂ in a manner necessarily producing a CO₂ rich stream and residual gas stream. A POSA would have understood at the critical date that the condensation of CO₂ occurs at cryogenic temperatures in combination with increased pressure. A POSA would have understood that CO₂ containing residual gas from PSA hydrogen separation is at low pressure and would have understood that compression of the residual gas is a required process step. Without a compression step, the required cryogenic temperatures would be below the sublimation point of CO₂ and CO₂ could not be liquified as required for cryogenic separation. The alternative of having the residual gas outlet of the PSA at elevated pressure (i.e., compressed) before cryogenic separation would have been recognized as economically impractical because it would dramatically reduce the hydrogen recovery rate of the PSA process. A POSA would not have selected this alternative. Reinertsen also teaches that hydrogen separation is possible with a Pd membrane process where the hydrogen product

stream is obtained at low pressure but the CO₂ containing residual gas is at higher pressure (i.e., already compressed). A POSA would have understood at the critical date that the residual gas pressure from a Pd membrane separation could be high enough to achieve a downstream cryogenic CO₂ separation without any further compression step. Efficient and economical application of a PSA separation step generating a lower pressure residual gas would require at least a compression step for cryogenic CO₂ separation in addition to cryogenic cooling.

160. In more detail, Reinertsen discloses a first residual gas stream, for cryogenic carbon dioxide separation in step (f), is subjected to at least one compression step and at least one cooling step, thereby producing the first carbon dioxide-rich stream at least partly in the form of a condensed carbon dioxide stream. EX1005, FIGURE 4, ¶¶[0103], [0110]; EX1022, p.19, ln.5–21, p.21, ln.3–5; EX1023, p.19, ln.15–31, p.21, ln.13–15. For example, Reinertsen discloses “[t]he shifted gas **61** is subsequently cooled in the condenser 7 to remove its water content **72**, thus obtaining dry shifted syngas **71**. Amine type separation process **8** separates CO₂ **81** from the shifted gas”,”: EX1005, ¶[0103]; EX1022, p.19, ln.9–12; EX1023, p.19, ln.19–22. Reinertsen further discloses that amine type CO₂ separation may be replaced with cryogenic CO₂ separation. EX1005, ¶[0110]; EX1022, p.21, ln.3–5; EX1023, p.21, ln.13–15 (“A difference between embodiment example 2 and embodiment example 1 is

that the amine unit 8 of embodiment example 1 is replaced with a cryogenic separation 13 of CO₂ 131 from the dried gas 73, giving the rest gas 132 for use in fired heater(s)”). As I have explained, Reinertsen discloses hydrogen separation using a PSA and subsequent cryogenic separation of CO₂ from the residual gas exiting the PSA, ¶¶147-155, *supra*. As explained ¶159, *supra*, cryogenic separation of CO₂ from PSA residual gas requires compression and cooling of the residual gas. Therefore, cryogenic CO₂ separation applied to residual gas from a PSA hydrogen separation in Reinertsen is an example of a process including at least one compression step and at least one cooling step for generating a carbon dioxide-rich stream at least partly in the form of a condensed carbon dioxide stream as provided in claim 1 of the ’805 Patent.

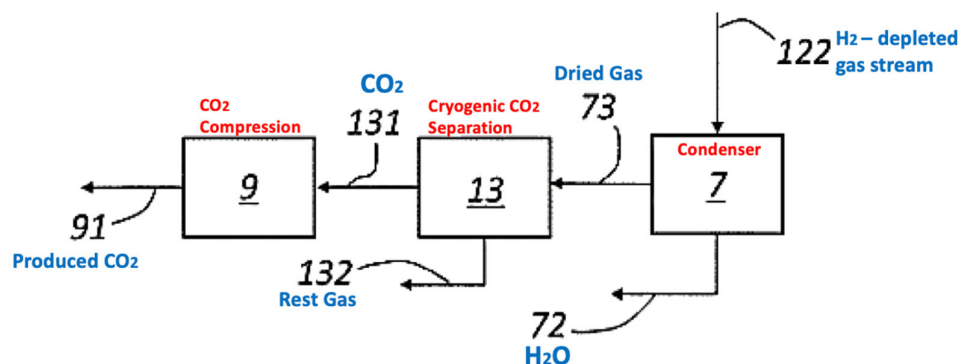


FIGURE 4, excerpt

17. Element 12: a plant configured for performance of the process

161. Element 12 recites “a plant configured for performance of the process according to claim 1.” It is my opinion that Reinertsen discloses each element of the process of claim 1 as detailed in Section XI.A.1–15, *supra*, from

which claim 12 depends. Reinertsen discloses a plant configured for performance of the hydrogen separation process. EX1005, ¶[0029]; EX1022, p.6, ln.18–19; EX1023, p.6, ln.19–20 (“there is provided a hydrogen production plant arranged to perform the method of the first aspect.”). In my opinion, the plant of claim 12 is defined by the process of claim 1. No additional elements of the plant are provided. Thus, Reinertsen in disclosing each and every element of the process of claim 1 and the further disclosure of providing a plant for performing the disclosed process, in my opinion, teaches each and every element of claim 12 as recited in the ’805 Patent.

18. The Claims are Anticipated by Reinertsen

162. Reinertsen discloses all of the features of claims 1, 11 and 12, either explicitly or inherently. It is my opinion that these claims are anticipated, and not patentable under 35 U.S.C. §102.

B. Petition I: Ground 2: Claims 1-6 , 11 and 12 are Obvious over Reinertsen and Darde

163. It is my opinion that claims 1-6, 11 and 12 would have been obvious to a POSA at the critical date under 35 U.S.C. § 103 in view of Reinertsen and Darde.

1. Reinertsen and Darde—Motivation to Combine

164. Reinersten and Darde share a common, recognized purpose of producing high purity hydrogen via methane reforming while maximizing process efficiency for separating and capturing CO₂ thereby decreasing CO₂

emissions. The common goal, as would be recognized by a POSA at the critical date, is achieved in Reinersten and Darde via a cryogenic capture solution, which allows for sequestration of CO₂ and/or generation of a CO₂ coproduct, in both cases reducing or preventing CO₂ emissions; e.g., to the environment.

165. Reinertsen achieves this commonly recognized purpose using a process for producing hydrogen from a hydrocarbon-rich feed gas involving reforming the feed gas to generate a reformed gas containing hydrogen and carbon monoxide, followed by a water-gas shift process to convert carbon monoxide into hydrogen and carbon dioxide. Subsequently, hydrogen is separated from the shifted gas, optionally using a PSA, and CO₂ is separated through cryogenic CO₂ separation. *See*, Sections IX.A, ¶¶57-70 and X1.A.1-17, ¶¶106-161, *supra*. Methane reforming, water-gas-shift conversion, PSA and cryogenic separation as described in Reinertsen are carried out by processes that were known to a POSA and subject to commercial implementation in the field of gas reforming as of the critical date. *See*, Sections IX.A, ¶¶57-70 and V., ¶¶26-41, *supra*.

166. Darde (EX1008) similarly achieves this commonly recognized purpose using steam reforming of hydrocarbons, integrated with carbon dioxide capture and steam generation via a process similar to Reinertsen. *See*, Section IX.D, ¶¶77-79, *supra*. The process in Darde involves mixing hydrocarbons with steam to produce a reforming feedstock, generating syngas, enriching it in

hydrogen and CO₂ via water-gas shift conversion, and separating process condensates for reuse. Section IX.D, ¶78, *supra*. The syngas is then treated using PSA to produce high-purity hydrogen, where the residual gas containing CO₂ is purified in a cryogenic purification unit (CPU). Section IX.D, ¶78, *supra*. Methane reforming, water-gas-shift conversion, PSA and cryogenic purification as described in Darde are carried out by processes that were known to a POSA and subject to commercial implementation in the field of gas reforming as of the critical date. *See*, Sections IX.D, ¶¶77-79 and V., ¶¶26-41, *supra*.

167. As would have been recognized by a POSA as of the critical date, the methane reforming processes of Reinertsen and Darde overlap significantly in that they utilize a feed gas stream including methane and steam to generate a synthesis gas stream by gas reforming reactions well known and understood by a POSA at the critical date. The water-gas-shift processes of Reinertsen and Darde function in the same manner in that they receive a synthesis gas stream as input and generate a shifted synthesis gas stream by a water gas shift reaction, which was well known and understood by a POSA at the critical date. The PSA processes of Reinertsen and Darde function in the same manner in that they receive a gas stream as input and generate a hydrogen-rich stream and residual gas stream, which was well known and understood by a POSA at the critical date. The cryogenic separation process of Reinertsen and the cryogenic

purification process of Darde function in the same manner in that they receive a gas stream as input and generate a carbon dioxide-rich stream and residual gas stream via a cryogenic separation process, which was well known and understood by a POSA at the critical date. In my opinion, therefore, a POSA would have been motivated to combine the disclosures from Darde with those of Reinertsen because both references implement a similar combination of known and compatible processes to achieve a common purpose of producing a hydrogen product stream with reduced carbon dioxide emissions.

168. Reinertsen describes processes for hydrogen separation using a Pd-membrane provided upstream of cryogenic CO₂ separation. EX1005, ¶¶[0105]–[0110], FIGURE 4. PSA is disclosed in Reinertsen as an alternative, preferred process for hydrogen separation preferred for some embodiments. EX1005, ¶[0012]. Therefore, a POSA would view the hydrogen separation process in Reinertsen as including only two options, a Pd-membrane and PSA, as alternatives for use in the overall hydrogen generation process. Darde discloses advantages of the specific process configuration incorporating the PSA option provided upstream of cryogenic CO₂ separation, such that PSA offgas rich in CO₂ is cryogenically captured. EX1008, p.8, ln.8–11. These advantages in Darde would have been easily recognized by a POSA at the critical date as useful for implementing in the hydrogen generation process of Reinertsen. For example, Darde demonstrates a PSA process configuration achieving a “gaseous

stream of highly pure (greater than 99%) hydrogen” (EX1008, p.16, ln.7) and an offgas having a high CO₂ content such that “[b]y virtue of this relatively high CO₂ content, the cryogenic CO₂ capture solution may be applied to the PSA offgas.” EX1008, p.8, ln.10–11. In my opinion, therefore, a POSA would have been motivated to make a selection of PSA as one of the finite number of options disclosed in Reinertsen (i.e., PSA and Pd-membrane) to achieve benefits of generating a highly pure hydrogen stream with reduced emissions by energetically efficient CO₂ capture, as described in Darde. A POSA would also have been motivated to incorporate PSA upstream of cryogenic CO₂ separation, in the manner shown in Darde, into the process of Reinertsen because both references use PSA operating in a normal and expected way to separate hydrogen and generate a residual gas stream that would have been viewed by a POSA as compatible with cryogenic CO₂ separation to reduced CO₂ emissions.

169. Moreover, in my opinion, a POSA at the critical date would have recognized additional advantages of using PSA, in the manner shown in Darde, in the process of Reinertsen. For example, a POSA at the critical date would have recognized a benefit as shown in the process of Darde of PSA producing a purified hydrogen stream having a pressure sufficiently high for use in certain industrial applications, such as energy applications, use as a recycle stream and/or electrochemical applications. For hydrogen production in both Reinertsen and Darde, the reformer outlet gas is routed to a water gas shift

reaction (WGS). After WGS the hydrogen rich synthesis gas, also labelled as shifted gas, is preferably purified directly in a pressure swing absorption unit (PSA) in order to separate unconverted methane, carbon dioxide and carbon monoxide from the purified hydrogen product. By virtue of the PSA process, components other than hydrogen are selectively adsorbed, whereas hydrogen is allowed to pass, thereby generating a purified hydrogen product stream having a pressure which is close to the inlet pressure of the PSA minus a minor pressure drop due to the adsorbers. Therefore, in my opinion, a POSA would have recognized the capability of PSA to generate a hydrogen output stream having a sufficiently high pressure as an advantage for subsequent use of the hydrogen generated in industrial applications. Generating a high purity hydrogen gas product stream at such pressures via PSA would have been viewed by a POSA as providing an advantage over other hydrogen purification processes, such as membrane-based hydrogen separation approaches for which a decrease of pressure across the membrane is unavoidable. In my opinion, therefore, a POSA would have been motivated to select PSA for hydrogen separation, as shown in Darde, for hydrogen separation in the processes of Reinertsen to achieve a high purity hydrogen product stream having a useful pressure.

170. In my opinion, another aspect in favor of selection of PSA, as described in both Reinertsen and Darde, is the recognition by a POSA at the critical date of the high degree of technical readiness, practical robustness and

ease of implementation of PSA in combination with gas reforming and cryogenic CO₂ separation. For example, a person having ordinary skill in the art would view PSA as having potential for a high useful, practical lifetime and performance attributes in an industrial application, such as hydrogen production, as compared to an alternative Pd-membrane process which would be recognized by a POSA at the critical date as susceptible to decreased performance in certain industrial settings due to acknowledged problems such as membrane fouling. Motivation for the selection of PSA for hydrogen separation also includes compatibility of the process parameters, such as temperature, pressure and flow rates, used in PSA with downstream cryogenic CO₂ capture, used in both Reinertsen and Darde, that would also be readily understood by a POSA at the critical date.

171. In my opinion, a POSA would also have been motivated to combine the teachings of Reinertsen with Darde because both processes are directed to hydrogen production capable of achieving a high purity hydrogen product stream. The reported high purity achieved in Darde (greater than 99%) would be viewed by a POSA at the critical date as meeting the criteria and objectives of Reinertsen, particularly when combined with the acknowledged reduction of CO₂ emission. The high purity hydrogen product and compatibility with cryogenic CO₂ capture as described in Darde, provide guidance and

motivation for a person having skill in the art to select PSA upstream of cryogenic CO₂ separation in the process of Reinertsen.

172. Darde also provides an explicit recitation of the stream components as described in claim 1 of the '805 Patent, including feed stream composition (EX1008, p.4, ln.7–9, p.13, ln.36–37); synthesis gas stream compositions as generated by gas reforming (EX1008, p.5, ln.13–34, p.9, ln.30–32, p.5, ln.29–31); downstream water-gas-shift conversion (EX1008, p.7, ln.30–32, p.15, ln.33–36); hydrogen-rich stream composition and residual gas stream composition from a PSA process (EX1008, p.7, ln.35–p.8, ln.4, p.8, ln.8–11); and a carbon-dioxide-rich stream composition and a residual gas stream composition from a cryogenic CO₂ separation (EX1008, p.8, ln.24–29, p.16, ln.11–22). In my opinion, the gas stream compositions of the '805 Patent are also explicitly or inherently present in the process of Reinertsen. To the extent that the gas stream compositions would not be viewed as provided in Reinertsen, a POSA at the critical date would have had sufficient knowledge and guidance to implement the process of Reinertsen to achieve the gas and product stream components explicitly disclosed in Darde. Selection of the stream compositions of Darde, would not change the principles of operation and/or process conditions in Reinertsen, as these compositions are established by the overlapping gas reforming processes and reactions (EX1005, ¶[0067]) and the downstream conversion and separation processes that are provided in

both Darde and Reinertsen. Therefore, the stream components in Darde would have been recognized by a POSA at the critical date as compatible with and readily adaptable to the process in Reinertsen. Moreover, a POSA at the critical date would have viewed the stream components of Darde as advantageous for the purpose recognized in Reinertsen of generating a high purity hydrogen stream with reduced carbon dioxide emissions. Given that Reinertsen and Darde use a similar combination of methane reforming with the same downstream conversion and separation processes, carrying out the process in Reinertsen to include the gas stream components in Darde would have been a matter of routine engineering within the skill of a POSA at the critical date.

2. Petition I: Ground 2 Reinertsen and Darde Combine to Disclose the Limitations of the Claims

173. My analysis of relevant aspects of Reinertsen is given above with respect to Ground 1. *See*, Sections XI.A.1-17, ¶¶106–161, *supra*.

(a) Element 1.pre: Preamble

174. *See*, Section XI.A.1, ¶¶108–110, *supra*.

175. Darde discloses hydrogen production by either an endothermic reforming process (SMR) or an ATR reforming process combined with hydrogen separation by PSA followed by cryogenic CO₂ capture using a CO₂-purifying CPU unit. EX1008, Abstract, p.4, ln.5–16. As I have stated above (Section XI.A, *supra*), Reinertsen provides an example of a process for preparing hydrogen by reforming hydrocarbons with steam, and for separation

of carbon dioxide as recited in claim 1 of the '805 Patent. A combination of the dual reformers of Reinertsen with the process for hydrogen production of Darde provides an example of a process for preparing hydrogen by reforming hydrocarbons with steam, and for separation of carbon dioxide as recited in claim 1 of the '805 Patent. It is my opinion that Reinertsen in view of Darde provides a process for preparing hydrogen by reforming hydrocarbons with steam, and for separation of carbon dioxide as recited in claim 1 of the '805 Patent.

(b) Element 1.1: providing a feed gas stream

176. See, Section XI.A.1.1, ¶¶111–112, *supra*. It is my opinion that Reinertsen alone or in view of Darde discloses providing a feed gas stream.

(c) Element 1.1.1: hydrocarbon component and steam

177. See, Section XI.A.1.1.1, ¶¶113–115, *supra*.

178. Darde discloses the compositions of feed gas streams including hydrocarbon and steam. EX1008, p.4, ln.5–8 (“... the hydrocarbons to be reformed are mixed with steam so as to produce the feedstock for the reforming.”). It is my opinion that Reinertsen alone or in view of Darde discloses a feed gas stream comprising a hydrocarbon and steam and therefore this element of claim 1.

(d) Element 1.1.2: methane

179. See, Section XI.A.1.1.2, ¶¶116–117, *supra*. It is my opinion that Reinertsen alone or in view of Darde discloses a feed gas stream with a

hydrocarbon component comprising at least methane and therefore this element of claim 1.

(e) Element 1.2: endothermic reforming step

180. See, Section XI.A.1.2, ¶¶118–120, *supra*. It is my opinion that Reinertsen alone or in view of Darde discloses reforming at least a portion of the feed gas stream in an endothermic reforming step over a reforming catalyst thereby producing a first synthesis gas stream.

(f) Element 1.2.1: first synthesis gas

181. See, Section XI.A.1.2.1, ¶¶121–123, *supra*.

182. Darde discloses reforming processes for producing synthesis gases comprising hydrogen, carbon monoxide, carbon dioxide, and unreacted methane; for example, EX1008, p.5, ln.29–34 (“always in the form of a mixture containing mainly hydrogen (H₂) and carbon monoxide (CO) and in smaller proportions carbon dioxide (CO₂), but also unreacted methane (CH₄), excess steam and traces of various compounds, impurities initially present but also generated during the reforming, and essentially due to the presence of the catalyst”) and EX1008, p.15, ln.30–31 (“comprising essentially hydrogen and CO, but also CO₂, CH₄, water and impurities”). It is my opinion that Reinertsen alone or in view of Darde discloses a first synthesis gas stream comprising hydrogen, carbon monoxide, carbon dioxide, and unreacted methane and therefore this element of claim 1.

(g) Element 1.3: reforming the first synthesis gas

183. See, Section XI.A.1.3, ¶¶124–126, *supra*. It is my opinion that Reinertsen alone or in view of Darde discloses reforming the first synthesis gas stream in an autothermal reforming step thereby producing a third synthesis gas stream, and therefore this element of claim 1.

(h) Element 1.3.1: exothermic partial oxidation and endothermic reforming

184. See, Section XI.A.1.3.1, ¶¶127–128, *supra*. It is my opinion that Reinertsen alone or in view of Darde discloses the autothermal reforming step comprising exothermic partial oxidation and endothermic reforming with steam over a reforming catalyst, and therefore this element of claim 1.

(i) Element 1.3.2: second and third synthesis gas

185. See, Section XI.A.1.3.2, ¶¶129–133, *supra*.

186. Darde discloses reforming processes for producing synthesis gases comprising hydrogen, carbon monoxide, carbon dioxide, and unreacted methane; for example, see ¶182 under “Element 1.2.1: first synthesis gas”, *supra*. It is my opinion that Reinertsen alone or in view of Darde discloses the third synthesis gas stream comprising hydrogen, carbon monoxide, carbon dioxide, and unreacted methane, and therefore this element of claim 1.

(j) Element 1.3.3: heat generated by the autothermal reforming step

187. See, Section XI.A.1.3.3, ¶¶134–136, *supra*. It is my opinion that Reinertsen discloses that heat generated by the autothermal reforming step is

utilized for heating in the endothermic reforming step of step (b), and therefore discloses this element of claim 1.

(k) Element 1.4: converting the carbon monoxide

188. See, Section XI.A.1.4, ¶¶137–139, *supra*. It is my opinion that Reinertsen alone or in view of Darde discloses converting the carbon monoxide present in a third synthesis gas stream with steam thereby producing hydrogen and carbon dioxide thereby producing a fourth synthesis gas stream, and therefore this element of claim 1.

(l) Element 1.4.1: fourth synthesis gas

189. See, Section XI.A.1.4.1, ¶¶140–145, *supra*.

190. Darde discloses a shift reaction processes for producing a synthesis gas composition comprising carbon dioxide, carbon monoxide unconverted, hydrogen not separated off, and unreacted methane; for example “a synthesis gas enriched with hydrogen and CO₂ and depleted of CO compared with the gas produced by reforming, and also containing additional impurities generated during the conversion of about 65% to 85% of H₂, 30 11 % to 22% of CO₂, 0.5% to 6% of unconverted CO and 3% to 10% of CH₄”; EX1008, p.15, ln.33–36. It is my opinion that Reinertsen alone or in view of Darde discloses a fourth synthesis gas stream comprising hydrogen, carbon dioxide, unreacted methane, and carbon monoxide unconverted in step (d) and therefore this element of claim 1.

(m) Element 1.5: separating hydrogen

191. See, Section XI.A.1.5, ¶¶146–148, *supra*.

192. Darde discloses PSA for producing a hydrogen-rich stream and a first residual gas stream, wherein the first residual gas stream comprises carbon dioxide, unconverted carbon monoxide, hydrogen not separated off, and unreacted methane; for example “... produces at least one gaseous stream of highly pure (greater than 99%) hydrogen, and also a residual gas **12** which for its part contains all the carbon dioxide, the vast majority of the unconverted methane and of the unconverted carbon monoxide, and a large part of the nitrogen and hydrogen, the quality of which depends on the yield from the H₂ PSA.” EX1008, p.16, ln.6–10. It is my opinion that Reinertsen alone or in combination with Darde, and for example in view of the additional disclosure in Darde regarding PSA H₂ separation, discloses separating hydrogen from a fourth synthesis gas stream by pressure swing adsorption, thereby producing a first hydrogen-rich stream and a first residual gas stream, and therefore this element of claim 1.

(n) Element 1.5.1: first residual gas

193. See, Section XI.A.1.5.1, ¶¶149–150, *supra*.

194. Darde explicitly discloses the compositions of the hydrogen rich stream including hydrogen and the residual stream produced by PSA hydrogen separation including carbon dioxide, carbon monoxide, hydrogen not separated in the PSA step and unreacted methane. EX1008, p.8, ln.1–4 (“At the same time

as almost pure hydrogen, the H₂ PSA unit produces a PSA offgas which contains the CO₂ contained in the synthesis gas feeding the PSA unit, and also the methane which did not react during the reforming, CO which did not react during the conversion step and a part of the hydrogen feeding the PSA unit”). It is my opinion that Reinertsen alone or in view of Darde discloses a first residual gas stream comprising carbon dioxide, carbon monoxide unconverted in step (d), hydrogen not separated off in step (e), and unreacted methane, and therefore this element of claim 1.

(o) Element 1.6: separating carbon dioxide

195. See, Section XI.A.1.6, ¶¶151–153, *supra*.

196. Darde discloses cryogenic CO₂ capture using a CO₂-purifying CPU unit for producing a first carbon dioxide-rich stream and a second residual gas stream RG2; for example, “The residual gas **12** is sent at **13** to a CPU unit for separation of the CO₂, the CPU unit producing at least one stream **14** of CO₂, a gaseous stream **15** rich in hydrogen which is recycled so as to feed the hydrogen PSA in order to recover the hydrogen contained and to thus improve the overall hydrogen yield of the facility, a stream **16** of non-condensables comprising methane, hydrogen, carbon monoxide, non-condensed carbon dioxide, nitrogen and water, constituting the CPU offgas which is recycled as reforming fuel to the reforming module **4**”. EX1008, p.16, ln.11–16. It is my opinion that Reinertsen in view of Darde discloses separating carbon dioxide from a first residual gas stream obtained in step (e) by cryogenic carbon dioxide separation,

thereby producing a first carbon dioxide-rich stream and a second residual gas stream RG2, and therefore this element of claim 1.

(p) Element 1.6.1: second residual gas

197. See, Section XI.A.1.6.1, ¶¶154–157, *supra*.

198. Darde discloses that the second residual gas stream RG2 comprises carbon monoxide unconverted, hydrogen not separated off, carbon dioxide not separated off, and unreacted methane; for example, see ¶196 under “Element 1.6: separating carbon dioxide”, *supra*. It is my opinion that Reinertsen alone or in view of Darde discloses a second residual gas stream comprising carbon monoxide unconverted in step (d), hydrogen not separated off in step (e), carbon dioxide not separated off in step (f), and unreacted methane, and therefore this element of claim 1.

(q) Element 2: hydrogen is separated from the second residual gas stream (RG2), thereby producing a second hydrogen-rich stream (HG2) and a third residual gas stream (RG3)

199. In my opinion, Reinertsen alone, or in combination of Darde, teaches each and every element of claim 1 from which claim 2 depends. Sections XI.A and XI.B, *supra*.

200. In the '805 Patent RG2 is residual gas exiting the cryogenic CO₂ separation unit (205) and is described as being subject to a separation process (e.g., membrane) to generate a stream enriched in hydrogen (e.g. HG2) and a third residual gas stream (RG3). EX1001, col.5, ln.53–63.

201. Reinertsen describes a process for generation of a stream enriched in hydrogen from a residual gas from cryogenic CO₂ separation. For example, FIGURE 4 in Reinertsen shows generation of a rest gas (**132**) shown by the annotated figure below.

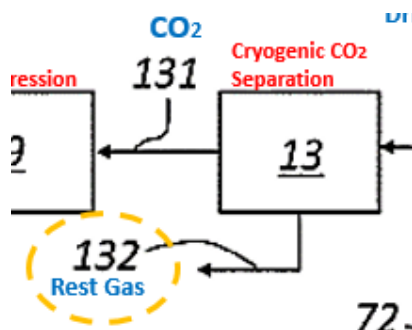
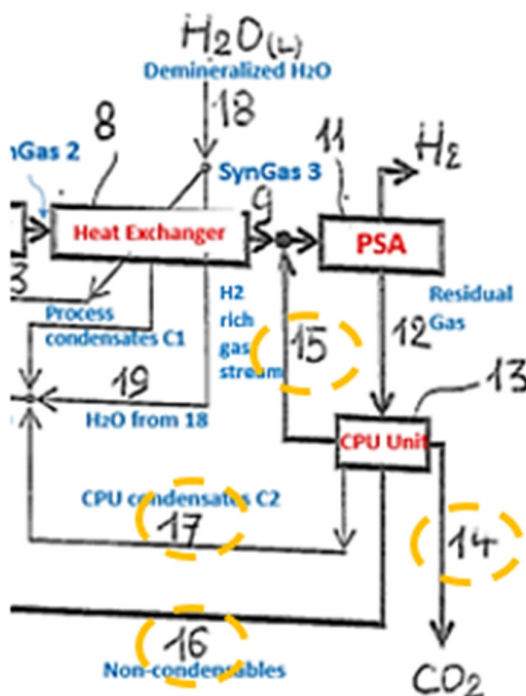


FIGURE 4, excerpt, annotated

In my opinion, a POSA would have recognized that rest gas **132** is enriched in hydrogen, particularly because it is described as useful for fuel for various recycle processes to increase hydrogen yield. EX1005, ¶¶[0123]–[0125], [0131]–[0136].

202. Darde describes a cryogenic CO₂ separation process carried out in a CPU unit **13** resulting in generation of a CO₂-enriched liquid stream **14**, a stream enriched in hydrogen **15**, a stream of non-condensables **16** and a liquid stream **17** of CPU condensates as shown by the annotated figure below:



EX1008, Figure, excerpt, annotated

In Darde, stream **16** is called CPU offgas which contains methane, hydrogen, carbon monoxide, non-condensed carbon dioxide, nitrogen and water. EX1008, p.16, ln.11–16. The means for generating the hydrogen-rich stream **15** is not explicitly described. However, the actual capture offgas from cryogenic separation would include all non-condensable gases generated in the CPU. Thus, non-condensable streams **15** and **16** must represent the combined capture offgas. As I have explained, ¶81, *supra*, it is my opinion that the CPU unit of Darde contains some form of H₂-separation, likely a H₂-separation membrane, through which the actual capture offgas of the CPU process is passed to generate the hydrogen rich stream **15** and the non-condensable stream **16**. In my

opinion, therefore, a POSA would have recognized that the hydrogen-rich stream **15** is the same as hydrogen-rich gas generated by hydrogen separation from the second residual gas stream **HG2** as described in the '805 Patent. The stream **16** of non-condensables in Darde which comprises methane, hydrogen, carbon monoxide, non-condensed carbon dioxide, and nitrogen would then be the residual non-condensable gases from the hydrogen separation. In my opinion, therefore, a POSA would have recognized stream **16** as the same as the third residual gas stream **RG3** generated by hydrogen separation from the second residual gas stream as described in the '805 Patent.

203. In my opinion, therefore, a POSA would have viewed the combination of Reinertsen and Darde as teaching a process including hydrogen separation of a second residual gas stream from cryogenic separation to produce a second hydrogen-rich stream and a third residual gas stream as described in the '805 Patent.

(r) Element 3: hydrogen is separated from the second residual gas stream (RG2) by membrane separation

204. In my opinion, Reinertsen in combination with Darde, teaches each and every element of claims 1 and 2 from which claim 3 depends. *See*, Sections XI.B.2(a)–(q), *supra*.

205. In the '805 Patent, **RG2** exits cryogenic CO₂ separation unit **(205)** and is described as being introduced into a membrane unit **206** to generate hydrogen-rich stream **HG2** and residual gas **RG3**. EX1001, col.10, ln.26–38.

206. Reinertsen describes processes including use of a Pd-membrane for separation of hydrogen from hydrogen containing gas streams to generate an H₂-enriched product stream. EX1005, ¶¶[0129], [0138].

207. Darde discloses that the condensation/separation steps of the CPU unit may be supplemented to include membrane separation steps. EX1008, p.8, ln.22–30. In my opinion, a POSA would have recognized the membrane separation steps described in Darde would include use of a membrane for separation of hydrogen from a residual gas, particularly given description in Darde of use of a separated hydrogen stream as a recycle to the PSA to increase overall all hydrogen production. In my opinion, a POSA would recognize the membrane separation steps described in Darde as applicable to the residual gas from cryogenic separation to generate a hydrogen-rich stream because membrane separation is specifically contemplated for the separation steps in the CPU Unit **13**, which is shown as generating hydrogen-rich gas stream **15**. EX1008, p.8, ln.22–30 and Figure.

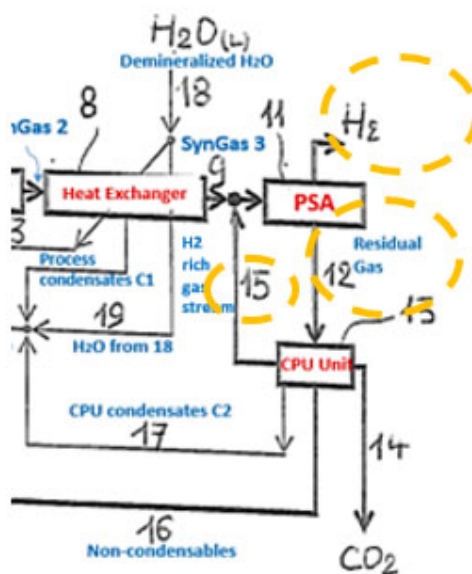
208. In my opinion, therefore, a POSA would view the combination of Reinertsen and Darde as teaching a process wherein membrane separation of a second residual gas stream from a cryogenic separation process is used to generate a hydrogen-rich stream as described in the '805 Patent.

(s) **Element 4: supplying the second hydrogen-rich stream to the fourth synthesis gas stream for separating hydrogen by PSA of step (e)**

209. In my opinion, Reinertsen in combination with Darde, teaches each and every element of claims 1 and 2 from which claim 4 depends. *See*, Sections XI.A.1-18 and XI.B.2(a)–(q), *supra*.

210. In the '805 Patent, hydrogen-rich gas HG2 exits membrane unit 206 (Figures 2 and 3) and is optionally recycled to the synthesis gas stream SG4 which enters PSA unit 204 to increase overall hydrogen yield. EX1001, col.12, ln.29–31.

211. Darde describes a process including recycle of a hydrogen-rich gas stream (15) from the CPU unit to the PSA unit, as depicted in the annotated Figure below.



EX1008, Figure, excerpt, annotated

212. In the configuration in Darde, the recycle of a hydrogen-rich gas stream (15) is combined with the synthesis gas stream 9 to be included as feed the PSA in order to recover the hydrogen contained and to thus improve the overall hydrogen yield of the facility. EX1008, p.16, ln.11–14. In my opinion, recycling stream 15 to the PSA in Darde corresponds to supplying the second hydrogen-rich stream to the fourth synthesis gas stream for separation of hydrogen by PSA as described in the '805 Patent. In my opinion, therefore, a POSA would have viewed the combination of Reinertsen and Darde as teaching a process including supplying a hydrogen-rich stream generated from the residual gas from cryogenic CO₂ separation to the synthesis gas stream from water-gas shift conversion for separation of hydrogen by pressure swing adsorption as described in the '805 Patent.

(t) *Element 5: gases present in the second hydrogen-rich stream are utilized as fuel gases for heating in the autothermal reforming step of step (c)*

213. In my opinion, as discussed in sections XI.A.1-18 and XI.B.2(a)-(q), *supra*, Reinertsen in combination of Darde, teaches each and every element of claims 1 and 2 from which claim 5 depends.

214. The '805 Patent states that “gases present in the hydrogen-rich stream HG2 are utilized as fuel gases for heating in the autothermal reforming step of step (c)”. EX1001, col.6, ln.9–11. It is explained in the '805 Patent that for the autothermal reforming step, the feed gas has to be warmed which can be done using a fired heating unit. EX1001, col. 6, lines 9–13. The fired heating

unit of the autothermal reformer may be heated at least partly with the aid of gases present in HG2 that have high calorific value (CO, methane and hydrogen). EX1001, col. 6, ln.11–18. Calorific value, as is known in the art, is the amount of heat energy present in a fuel determined by the complete combustion of a specified quantity of fuel at constant pressure and at normal conditions.

215. Darde discloses a separation step resulting in a second hydrogen-rich stream. XI.B.2.(q), *supra*. Reinertsen teaches the use of a residual gas from cryogenic separation as a fuel for providing energy in fired heater(s). EX1005, ¶¶[0053], [0103], [0110] (“The rest gas **102** contains remnants of CO and CH₄ together with unseparated CO₂ and hydrogen. The energy in the rest gas is utilized for fuel in fired heater(s)” and EX1005, Table 4 of Figure 13 (see below). In my opinion, description of fired heater(s) in Reinertsen would have been understood by a POSA as referring to a component of an ATR which provides a means of increasing the temperature of feed gases to a temperature that is operational and efficient for an autothermal reforming process. A POSA would also have recognized that rest gas **102** is enriched with hydrogen given that it is generated as a residual gas after separation of the CO₂. In my opinion, therefore, use of the residual gas as fuel in Reinertsen would have been viewed by a POSA at the critical date as corresponding to use of the hydrogen-rich stream as fuel gases for heating in the ATR step as described in the ’805 Patent.

Table 4. Effect of recycling rest-gas to ATR.

	Embodiment Example 2	Embodiment Example 5	Embodiment Example 6	Embodiment Example 7
Recycle option	No recycle	50%	80%	90%
O ₂ to ATR (kg/h)	4179	4316	4426	4482
H ₂ production (kmol/h)	752	777	794	799*
Carbon capture (%)	89.7	94.4	97.7	98.8
Heat available in Fired heater (MW)	4.6	2.5	1.0	0.5

EX1005, FIGURE 13, excerpt, annotated

(u) Element 6: first carbon dioxide-rich stream is subjected to a thermal separation process for separation of methane from carbon dioxide

216. In my opinion, claim 6 recites nothing more than applying methods known in the prior art. Reinertsen, alone or in combination with Darde, discloses all elements of claim 6.

217. Element 6 recites a limitation of “The process according to claim 1, wherein the first carbon dioxide-rich stream comprises unreacted methane, and the first carbon dioxide-rich stream is subjected to a thermal separation process for separation of methane, thereby producing a second carbon dioxide-rich stream.” It is my opinion that Darde discloses the first carbon dioxide-rich stream comprises unreacted methane, and the first carbon dioxide-rich stream is subjected to a thermal separation process for separation of methane, thereby producing a second carbon dioxide-rich stream.

218. Darde discloses liquid CO₂ generated by cryogenic CO₂ separation can be distilled to achieve CO₂ purities greater than 99%; for example, “Under

these temperature and pressure conditions, it will partially condense, the liquid phase being particularly enriched with CO₂ and the gas phase with non-condensable gases. The liquid obtained can then be distilled so as to achieve CO₂ purities greater than 99%.” EX1008, p.8, ln.18–21. In my opinion, a POSA would have understood that methane is a major impurity in the separated CO₂, and thus understood Darde to teach that methane from the CO₂-rich stream from cryogenic CO₂ separation may be removed by distillation, as provided by claim 6 the '805 Patent. It is my opinion that Reinertsen in view of Darde discloses the first carbon dioxide-rich stream is subjected to a thermal separation process for separation of methane, thereby producing a second carbon dioxide-rich stream, and therefore this element of claim 1.

(v) *Element 11: the first residual gas stream is subjected to at least one compression step and at least one cooling step*

219. In my opinion, Reinertsen teaches each and every element of claim 11. *See*, Section XI.A.1-16, *supra*.

220. I emphasize that cryogenic CO₂ separation was known in the art long before the critical date to include compression and cooling steps. *See* ¶40, *supra*; EX1018, EX1020; EX1021. In addition, Darde discloses that compression and cooling steps, as recited in claim 11, are steps in cryogenic CO₂ separation which is applied to residual gas from PSA separation. EX1008, p.8, ln.12–18 (“(CPU) calls for the partial condensation and/or the distillation of the CO₂ contained in the CO₂-rich gaseous stream in a cryogenic purification

unit (CPU) ...The gas, after drying and compression to a pressure between 20 and 100 bar(a), is cooled to a temperature close to that of the triple point of CO₂ (approximately -56 °C).). Based on this teaching in Darde, the teachings in Reinertsen (§159, *supra*) and the general skill and knowledge in the art, a POSA would have understood to implement cryogenic CO₂ separation following PSA for hydrogen separation using at least one compression step and at least one cooling step to produce a carbon dioxide-rich stream at least partly in the form of a condensed carbon dioxide stream. In my opinion, Reinertsen alone or in combination with Darde teaches all of the elements of claim 11 of the '805 Patent.

(w) *Element 12: a plant configured for performance of the process*

221. Element 12 recites “a plant configured for performance of the process according to claim 1.”

222. In my opinion, Reinertsen alone or in combination with Darde discloses all the elements of the process of claim 1. *See*, Section XI.A.1-18, XI.B.2(a)-(p) *supra*.

223. Darde further discloses a plant configured for performance of the hydrogen separation process, “[a]ccording to another aspect of this invention, the latter relates to a facility for producing hydrogen by reforming hydrocarbons using steam, combined with carbon dioxide capture and steam production.”

EX1008, p.13, ln.33–35. In my opinion, the plant of claim 12 is defined by the

process of claim 1. No additional elements of the plant are provided. In my opinion, therefore, the combination of Reinertsen and Darde teaches each and every element of claim 12 as recited in the '805 Patent.

3. Reasonable Expectation of Success

224. In my opinion, a POSA would have been readily able to adapt the teachings of Reinertsen with those of Darde to arrive at the process of claims 1–6, 11 and 12 of the '805 Patent with a reasonable likelihood of success.

225. The processes in Reinertsen and Darde are overlapping and used in a similar manner to achieve the same goal of hydrogen production with reduced CO₂ emissions. Reinertsen describes a process for producing hydrogen from a hydrocarbon-rich feed gas with reduced CO₂ involving i) a reforming process including an endothermic reforming step using a GHR and an autothermal reforming step using an ATR, wherein GHR and ATR components are provided in a series configuration and wherein heated gas from the ATR is utilized for heating the GHR; (ii) downstream conversion of CO to CO₂ via water gas shift conversion; and (iii) separation processes including a hydrogen separation (with the options of a Pd membrane or a PSA) provided upstream of cryogenic CO₂ separation. Darde discloses a similar process using methane reforming (e.g., SMR or ATR) with downstream water shift conversion, and subsequent PSA for hydrogen separation provided upstream of cryogenic CO₂ capture.

226. When combined with Reinertsen, which also provides for PSA as an alternative, the processes disclosed in Darde, provides sufficient teaching to

allow a POSA to select and implement PSA for hydrogen separation from the two options disclosed in Reinertsen (i.e., Pd-membrane and PSA). The guidance provided in Darde, therefore, would have been sufficient to allow for selection of PSA and successful integration into the process of Reinertsen to predictably arrive at the invention with a reasonable expectation of success, for example, to provide a hydrogen product stream at moderate cost with sufficient purity for applications including clean energy and manufacturing of high value chemicals such as ammonia. In my opinion, any modification or other adaptation needed to successfully combine Reinertsen with Darde in this manner would have been a matter of routine engineering and, thus readily within the skill of a POSA at the critical date.

227. A POSA would also expect that the reforming and separation processes of Reinertsen and Darde would be compatible with each other. Reinertsen and Darde include known reforming and separation processes having similar and overlapping process conditions, such as, pressures, flow rates and temperatures; similar process inputs such as feed stream; and similar process outputs such the synthesis gas streams, hydrogen-rich stream, residual streams and CO₂ rich stream. Therefore, a POSA would require no more than ordinary skill to adapt the teachings of Reinertsen and Darde to implement any necessary adaptation or modification using a practical selection of known processes and process conditions used in their ordinary manner. Given that these well-known

processes are used in Reinertsen and Darde in the same manner to achieve the same technical solution of generating hydrogen with reduced CO₂ emissions, a POSA would have been readily able to adapt the teachings of these references to arrive at the claimed invention with a reasonable likelihood of success.

228. In my opinion, the benefits of the use of a dual reformer configuration as shown in Reinertsen with hydrogen separation using PSA provided upstream of cryogenic carbon dioxide separation, as described by Darde, would have been recognized by a POSA at the critical date. For example, the article Clean Hydrogen. Part 1: Hydrogen from Natural Gas Through Cost Effective CO₂ Capture Cotton; March 2019 (EX1010) provides evidence that a combination of a GHR with an ATR in series for hydrogen production with heat transfer from the ATR to the GHR was known at the time of the invention to provide the benefits of high level energy recycle, minimized CO₂ emissions and CO₂ capture at process pressure. *See*, generally, EX1010. Cotton provides simulations demonstrating use of dual reforming via a GHR and ATR combination in series with heat exchange and downstream hydrogen separation via PSA, to allow for hydrogen production. Cotton reports a reduction in CO₂ emissions of approximately 15% achievable via a GHR and ATR combination in series configuration relative to a process using steam methane reforming only, e.g., without heat exchange. *See*, EX1010, p.4, Table 1, line “CO₂ emitted” and columns “SMR FLOWSHEET” and “LCH

FLOWSHEET”. The simulations reported in Cotton, therefore, provide evidence that the benefits and extent of reduction in CO₂ emissions alleged in the ’805 Patent are not surprising or unexpected, but rather would have been recognized by a POSA at the critical date. Thus, implementing the process of Reinertsen with the CO₂ capture processes of Darde would have been recognized by a POSA at the critical date to provide a process for hydrogen production having an improved degree of a reduction of CO₂ emissions.

4. The Claims are Obvious over the Combination of Reinertsen and Darde

229. The combination of Reinertsen and Darde discloses all of the features of claims 1–6, 11, and 12. As shown above, it would have been obvious for a POSA to combine those references and to have had a reasonable expectation of success to obtain the processes as claimed. It is my opinion that these claims are obvious, and not patentable under 35 U.S.C. § 103.

C. Petition I Ground 3: Claims 1, 6, 11 and 12 are Obvious over Rytter and Darde

230. It is my opinion that claims 1, 6, 11 and 12 would have been obvious to a POSA under 35 U.S.C. § 103 in view of Rytter and Darde.

1. Rytter and Darde—Motivation to Combine

231. Rytter and Darde share a common, recognized purpose of producing hydrogen via methane reforming with cryogenic separation of carbon

dioxide to generate a carbon dioxide coproduct and/or achieve to reduce carbon dioxide emissions; e.g., to the environment.

232. Rytter achieves this commonly recognized purpose by a process with steps including methane reforming using GHR and ATR processes provided in a series configuration with a downstream water-gas shift process, hydrogen separation and cryogenic separation of carbon dioxide. Methane reforming, water-gas-shift conversion, hydrogen separation and cryogenic separation as described in Rytter are carried out by processes that were known to a POSA and subject to commercial implementation in the field of gas reforming as of the critical date.

233. Darde similarly achieves this commonly recognized purpose through steam reforming of hydrocarbons, integrated with carbon dioxide capture and steam generation. The process involves mixing hydrocarbons with steam to produce a reforming feedstock, generating syngas, enriching it in hydrogen and CO₂ via water-gas shift conversion, and separating process condensates for reuse. The syngas is then treated using PSA to produce high-purity hydrogen, where the residual gas containing CO₂ is purified in a cryogenic purification unit (CPU). Methane reforming, water-gas-shift conversion, PSA and cryogenic purification as described in Darde are carried out by processes that were known to a POSA and subject to commercial implementation in the field of gas reforming as of the critical date. In my

opinion, therefore, a POSA would have been motivated to incorporate disclosure from Darde into Rytter because these references each achieve a common goal of a reduction of CO₂ emissions by implementing gas reforming using a similar solution of known hydrogen separation and cryogenic CO₂ separation processes for energy efficient capture of CO₂.

234. Rytter describes process steps for the series configuration of claims 1, 6, and 11, including the combination of downstream H₂ separation by PSA and cryogenic CO₂ separation. EX1009, p.6, ln.18–23. While Rytter discloses CO₂ separation 13 occurs upstream of H₂ separation 15 in the example process shown in FIGURE 3, it also describes H₂ separation (e.g. using a membrane) upstream of CO₂ separation as an alternative process option. EX1009, p.12, ln.19–20 (“By combining solid membranes and liquid membranes it is also possible to achieve a rapid permeation of CO₂, while H₂ is kept back”). A POSA at the critical date would, therefore, have recognized the specific use of PSA hydrogen separation positioned upstream of cryogenic CO₂ separation as a process contemplated and described explicitly in Rytter. To the extent this process sequence is not explicitly in Rytter, a POSA would have been motivated to select the sequence in view of description in Darde which clearly shows the advantages of a process configuration of PSA provided upstream of cryogenic CO₂ separation to provide for cost effective carbon capture. Darde provides sufficient teaching and motivation to allow a POSA to modify the sequence in

Rytter's FIGURE 3 to provide for hydrogen separation via PSA upstream of cryogenic CO₂ separation, for example, to provide a CO₂ product stream at moderate cost with sufficient purity for efficient carbon sequestration and/or use as a reagent stream for an industrial application; e.g. manufacturing.

235. In my opinion, a POSA would readily comprehend that the order of H₂ and CO₂ separation steps are interchangeable depending on a number of factors including the availability of the specific process and resources used to perform each step (e.g., amine separation vs. cryogenic separation for CO₂), target product application, and/or CAPEX considerations. A POSA would have recognized the specific hydrogen and CO₂ separation processes and their sequence present a finite set of known design choices for achieving H₂/CO₂ separation. For example, Rytter discloses only six practical permutations for H₂/CO₂ separation from a shifted gas, summarized below in the Table titled, "Summary of H₂ and CO₂ Separation Processes". In evaluating these options, a POSA would not reasonably place a cryogenic CO₂ separation step upstream of a H₂ separation step recognizing that this configuration would require additional energy to cool and compress the hydrogen present in the shifted synthesis gas. Further, Rytter explains that different arrangements of H₂ and CO₂ separation are possible, but that "A Pd membrane requires a certain elevated temperature to operate efficiently. Therefore, only high-temperature WGS may be applied before the membrane." EX1009, p.11, ln.26–28.

Summary of H₂ and CO₂ Separation Processes:

<i>Separation Processes</i>	
H₂ Separation	CO₂ Separation
Membrane (e.g., Pd-membrane)	Amine Separation/Solvent Separation
PSA	Cryogenic Separation
<i>Practical Sequences of Separation Steps</i>	
H₂ Separation Upstream	CO₂ Separation Upstream
1. Membrane → Amine Separation 2. Membrane → Cryogenic Separation 3. PSA → Amine Separation 4. PSA → Cryogenic Separation	5. Amine Separation → Membrane 6. Amine Separation → PSA

236. Therefore, when presented with Rytter's specific example in FIGURE 3, a POSA at the critical date would have recognized the interchangeability of processes and sequence of H₂/CO₂ separation and would have been motivated to select the PSA → Cryogenic Separation alternative. Moreover, Darde provides guidance supporting selection of this specific process sequence by explaining that it is preferable to place a CO₂ separation step downstream of H₂ separation in providing:

“The PSA offgas is the gaseous stream richest in CO₂ of the process described above (typically 40-55% in the absence of capture upstream) ...

By virtue of this relatively high CO₂ content, the cryogenic CO₂ capture may be applied to the PSA offgas.” EX1008, p.8, ln.8–11.

237. Thus, a POSA would have recognized from this teaching in Darde that upstream PSA separation of H₂ provides a residual gas with beneficial properties, such as pressure and CO₂ content, allowing for effective cryogenic CO₂ capture. This guidance provides further motivation to a POSA to select the configuration of PSA provided upstream of cryogenic CO₂ separation in Darde for implementation in the process of Rytter. Therefore, a POSA would further be motivated to make this substitution by routine optimization of the finite number of design choices in Rytter, particularly in view of the explicit description in Darde of this configuration achieving cost effective and energetically efficient CO₂ capture.

238. Moreover, in my opinion, a POSA at the critical date would have recognized additional advantages of implementing hydrogen separation, such as by PSA, upstream of cryogenic CO₂ separation, in the manner shown in Darde, in the process of Rytter. For example, a POSA at the critical date would have recognized that separating hydrogen gas upstream reduces the total amount of gas provided for cryogenic CO₂ separation, thereby allowing for a physically smaller sized cryogenic unit(s) and beneficial reductions in capital expenditure (CAPEX) costs and operating expense (OPEX) costs associated with the overall process.

239. Darde also provides a verbatim recitation of the gas stream compositions as described in claim 1 of the '805 Patent, as discussed in ¶172, *supra*.

240. In my opinion, the gas stream compositions of the '805 Patent are also explicitly or inherently present in the process of Rytter. To the extent that the gas stream compositions would not be viewed as provided in Rytter, a POSA at the critical date would have sufficient knowledge and guidance to implement the process of Rytter to achieve the gas and product stream compositions explicitly disclosed in Darde. Selection of the stream compositions of Darde, would not change the principles of operation and/or process conditions in Rytter, as these compositions are established by the overlapping steam reforming processes and reactions and the downstream water-gas shift conversion and separation processes that are provided in both Darde and Rytter. Therefore, the stream compositions in Darde would have recognized by a POSA at the critical date as compatible with and readily adaptable to the process in Rytter.

241. Moreover, in my opinion, a POSA at the critical date would have viewed the stream compositions of Darde as advantageous for the purpose recognized in Rytter of generating a high purity hydrogen stream with reduced carbon dioxide emissions. Given that Rytter and Darde both use a similar combination of methane steam reforming with the same downstream water-gas

shift conversion and separation processes, carrying out the process in Rytter to include the gas stream compositions in Darde would have been a matter of routine engineering within the skill of a POSA at the critical date.

2. Rytter and Darde Combine to Disclose the Limitations of the Claims

242. The combination of Rytter and Darde provides all the features of claims 1, 6 and 11 of the '805 Patent.

243. My analysis of relevant aspects of Darde is summarized with respect to Ground 2. Sect. XI, B.2, ¶¶173-199, *supra*.

(a) Element 1.pre: Preamble

244. Rytter discloses a process for preparing hydrogen by reforming hydrocarbons with steam, and for separation of carbon dioxide. *See*, generally, Figure 3, as annotated below:

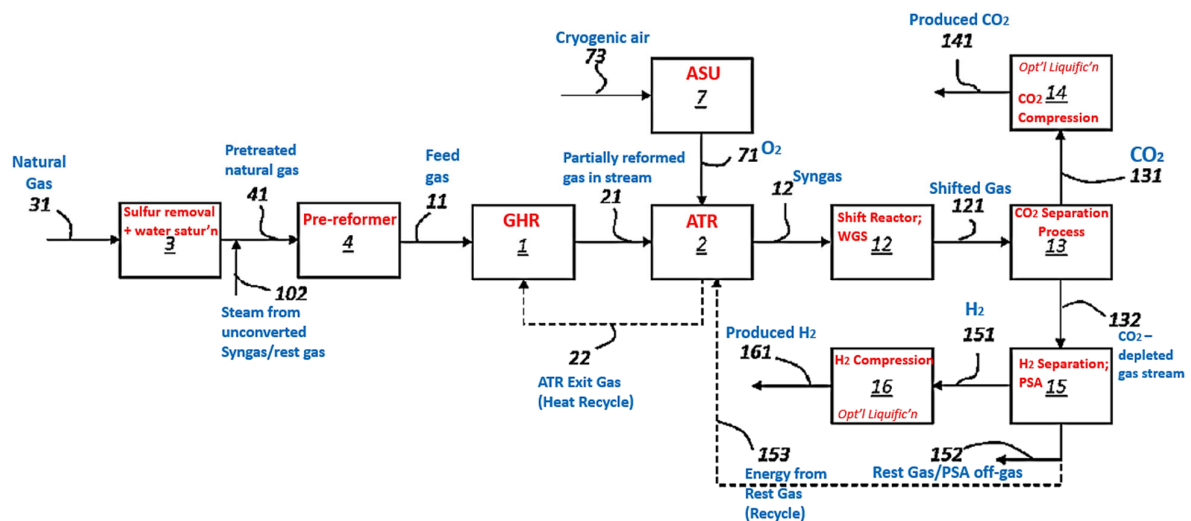


Figure 3 (annotated)

245. Rytter describes “Figure 3 shows production of hydrogen and CO₂ by combination of ATR **2** and GHR **1**.” EX1009, p.14, ln.32–33. Rytter further discloses “[s]eparation process **13** separates CO₂ **131** from the shifted gas, and the CO₂ is compressed **14** and optionally liquified.” EX1009, p.15, ln.13–14.

246. Darde discloses hydrogen production by either an endothermic reforming process (SMR) or an ATR reforming process combined with hydrogen separation by PSA followed by cryogenic CO₂ capture using a CO₂-purifying CPU unit. *See*, Section XI.B.2(a), *supra*. It is my opinion that Rytter alone or in view of Darde provides a process for preparing hydrogen by reforming hydrocarbons with steam, and for separation of carbon dioxide as ...recited in claim 1 of the '805 Patent.

(b) Element 1.1: providing a feed gas stream

247. Rytter discloses providing a feed gas stream. EX1009, p.20, Claim 1. For example, Rytter describes, “(a) method of producing hydrogen, the method comprising: receiving a feed gas comprising hydrocarbons ... ” EX1009, p.20, Claim 1. It is my opinion that Rytter alone or in view of Darde discloses providing a feed gas stream as recited in claim 1 of the '805 Patent.

(c) Element 1.1.1: hydrocarbon component and steam

248. Rytter discloses the feed gas stream comprises a hydrocarbon component and steam. EX1009, Claim 1, p.4, ln.26–27, p.9, ln.6–9, p.17, ln.17–18, p.20, ln.30. Rytter explicitly discloses a hydrocarbon component in the feed gas stream, such as provided in claim 1 (“receiving a feed gas comprising

hydrocarbons”). Rytter further discloses the inclusion of steam in the feed gas stream. Rytter notes that steam is introduced to the feed gas stream for pre-treatment. EX1009, p.9, ln.8–9 (“Pre-treatment may comprise pre-reforming whereby higher hydrocarbons, such as ethane, are converted by steam to methane and CO₂.”). Rytter further discloses “generating steam 102 that is added before the pre-reformer and before the WGS reactor 12.” EX1009, p.17, ln.17–18.

249. Additionally, Rytter discloses that the feed gas comprises water. For instance, Rytter describes a process of “optionally saturating the received feed gas with at least water.” EX1009, p.4, ln.26–27; p.20, ln.30. When the water-saturated feed gas is heated, as disclosed by Rytter, a POSA would have understood that steam is necessarily produced and is thus present within the feed gas stream. Rytter makes this understanding explicit by, for example, referring to the chemical reaction “CH₄ + H₂O $\rightleftharpoons$ CO + 3H₂” of the reforming process as “Steam reforming.” EX1009, p.9, ln.15. It is my opinion that Rytter alone or in view of Darde discloses a feed gas stream comprising a hydrocarbon and steam and therefore this element of claim 1.

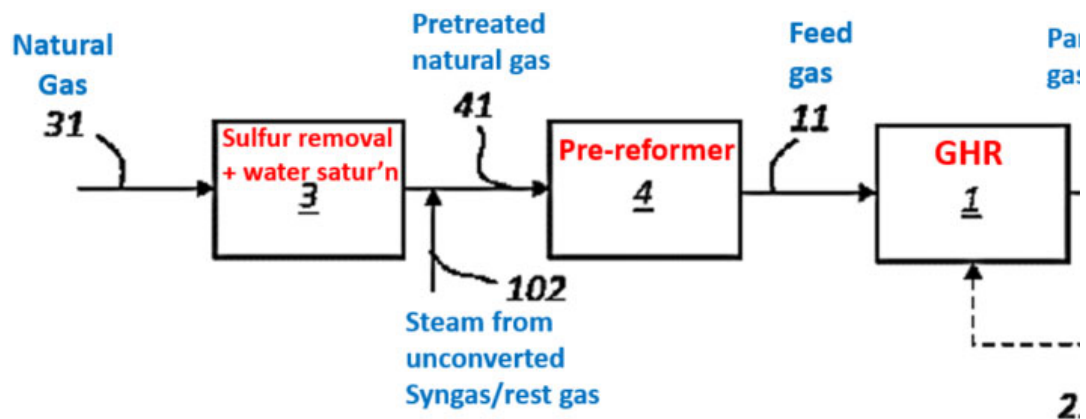


Figure 3, excerpt, annotated

(d) Element 1.1.2: methane

250. Rytter discloses a feed gas stream with a hydrocarbon component containing methane. EX1009, p.15, ln.24–26 (“Process simulations of the flow-sheet in Figure 3 using the program HYSYS is based on natural gas with molar composition 88.8% methane, 5.6% ethane, 2.0% propane, 1.6% higher hydrocarbons, 1.5% CO₂ and 0.6% nitrogen.”), p.9, ln.6–7 (“In a preferred embodiment, cleaning and pre-treatment of natural gas is performed so that the gas feed mainly comprises methane after these processes.”). It is my opinion that Rytter alone or in view of Darde discloses a feed gas stream with a hydrocarbon component comprising at least methane and therefore this element of claim 1.

(e) Element 1.2: endothermic reforming step

251. Rytter discloses reforming at least a portion of the feed gas stream in an endothermic reforming step over a reforming catalyst thereby producing a

first synthesis gas stream. EX1009, p.9, ln.15–19, p.10, ln.30–33. Rytter describes the “steam reforming” chemical reaction “ $\text{CH}_4 + \text{H}_2\text{O} \rightleftharpoons \text{CO} + 3\text{H}_2$ ” of the feed gas stream reforming process as being “strongly endothermic.”

EX1009, p.9, ln.15–19. Rytter further discloses that this endothermic reforming step is performed in a gas heated reformer (GHR) using a catalyst. EX1009, p.10, ln.30–33 (“...instead of burning fuel gas to provide the heat for the reforming reactions, the hot, autothermally reformed gas 22 is used to heat the catalyst tubes in a GHR 1. The feed gas 11 first passes through the catalyst in the GHR 1, then the ATR 2 by stream 21 and finally the heating side of the GHR to provide the heat for the initial reaction.”).

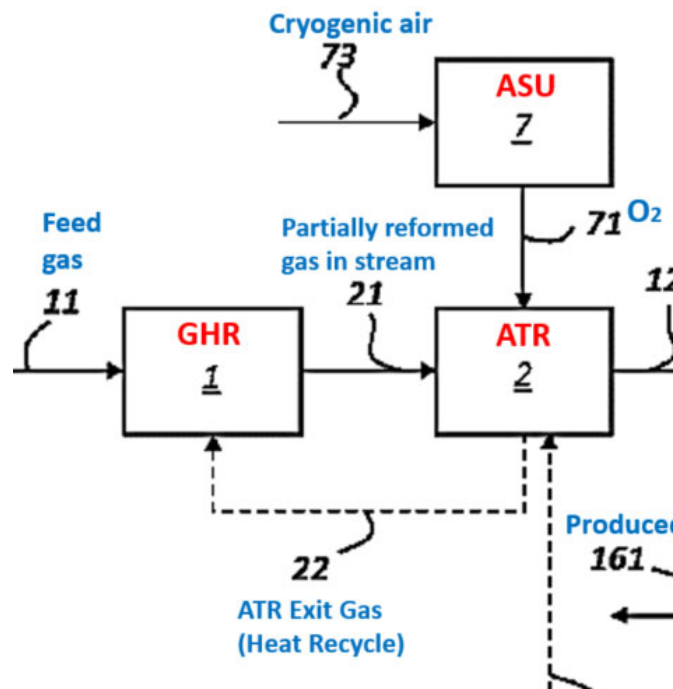
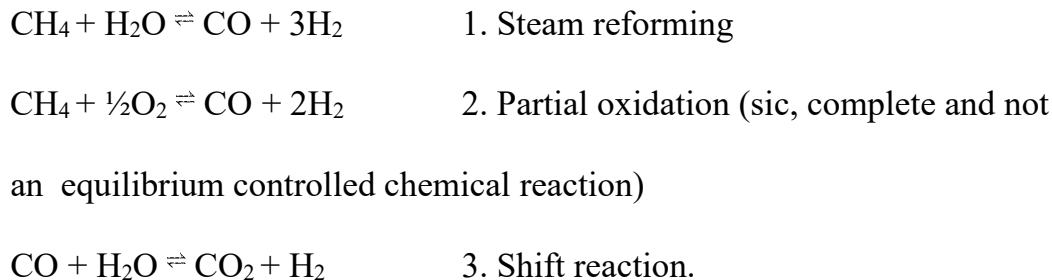


Figure 3, excerpt, annotated

252. Moreover, in my opinion, a POSA would have understood that a GHR process comprises an endothermic reforming step. *See*, Section IX.A, ¶64, *supra*. It is my opinion that Rytter alone or in view of Darde discloses reforming at least a portion of the feed gas stream in an endothermic reforming step over a reforming catalyst thereby producing a first synthesis gas stream and therefore this element of claim 1.

(f) Element 1.2.1: first synthesis gas

253. Rytter discloses, at least inherently, a first synthesis gas stream comprising hydrogen, carbon monoxide, carbon dioxide, and unreacted methane. EX1009, p.9, ln.15–19, p.11, ln.2–3. Rytter describes, “[t]he gas mixture from the reformer reactor contains mainly the gas components CO, H₂, H₂O, CO₂ and some CH₄.” EX1009, p.11, ln.2–3. Moreover, Rytter describes three reversible chemical reactions that occur during the production of synthesis gas (EX1009, p.9, ln.15–19):



254. The gas heated reformer of Rytter thus produces an output containing at least carbon monoxide (CO) and hydrogen (H₂). In my opinion, Rytter’s GHR would also naturally result in the production of unreacted methane and carbon dioxide. As the steam reforming reaction is in equilibrium,

both methane (CH₄) and steam (H₂O) are present in the output of the reaction. Thus, to the extent Rytter may not expressly disclose the composition of the first synthesis gas stream as including the gases enumerated in the claim, Rytter nevertheless discloses such composition of gases at least inherently. *See*, Sections IX.A and XI.A.5, *supra*.

255. Additionally, to the extent Rytter may not explicitly disclose such a composition of a synthesis gas stream, Darde discloses reforming processes for producing synthesis gases comprising hydrogen, carbon monoxide, carbon dioxide, and unreacted methane. *See*, Section XI.B.2(f), *supra*.

256. Darde discloses reforming processes for producing synthesis gases comprising hydrogen, carbon monoxide, carbon dioxide, and unreacted methane. *See*, Section XI.B.2(f), *supra*. It is my opinion that Rytter alone or in view of Darde discloses a first synthesis gas stream comprising hydrogen, carbon monoxide, carbon dioxide, and unreacted methane and therefore this element of claim 1.

(g) *Element 1.3: reforming the first synthesis gas*

257. Rytter discloses reforming the first synthesis gas stream in an autothermal reforming step thereby producing a third synthesis gas stream. EX1009, p.10, ln.30–33, p.14, ln.6–8, p.15, ln.1–2. Rytter discloses a series arrangement of passing a reformed gas (“stream 21”) through an autothermal reformer (“ATR 2”) to produce a new synthesis gas stream (“syngas 12”). EX1009, p.10, ln.30–33 (“(a)ccording to embodiments, instead of burning fuel

gas to provide the heat for the reforming reactions, the hot, autothermally reformed gas **22** is used to heat the catalyst tubes in a GHR 1. The feed gas **11** first passes through the catalyst in the GHR 1, then the ATR 2 by stream 21 and finally the heating side of the GHR to provide the heat for the initial reaction.”); EX1009, p.14, ln.6–8 (“After partial conversion of methane, the gas **52** is fed to an autothermal reformer **2** for further conversion to the syngas 22 according to eqs. 1 and 3.”); EX1009, p.15, ln.1–2 (“The syngas production, by the ATR 2 and GHR 1, is as described in Figure 1 with units with corresponding reference signs.”). It is my opinion that Rytter alone or in view of Darde discloses reforming the first synthesis gas stream in an autothermal reforming step thereby producing a third synthesis gas stream, and therefore this element of claim 1.

(h) Element 1.3.1: exothermic partial oxidation and endothermic reforming

258. Rytter discloses the autothermal reforming step comprises exothermic partial oxidation and endothermic reforming with steam over a reforming catalyst. EX1009, p.9, ln.1–5. Rytter discloses the use of an ATR, which by its nature operates in this manner. EX1009, p.9, ln.3–p.10, ln.5 (“... in an ATR, natural gas (methane) is fed together with oxygen or air into a combustion chamber. The energy which is required to operate the endothermic steam reforming reactions is provided by the exothermic reactions between hydrocarbons and/or hydrogen and oxygen. The temperature in the combustion

chamber can reach more than 2000° C. After the combustion chamber the reactions may be driven to equilibrium over a catalyst bed before the synthesis gas leaves the reactor at approximately 1000-1050° C.”). It is my opinion that Rytter alone or in combination with Darde discloses the autothermal reforming step comprising exothermic partial oxidation and endothermic reforming with steam over a reforming catalyst, and therefore this element of claim 1.

(i) Element 1.3.2: second and third synthesis gas

259. Rytter discloses the third synthesis gas stream comprises hydrogen, carbon monoxide, carbon dioxide, and unreacted methane. EX1009, p.11, ln.1–3, p.14, ln.22–25. Rytter discloses the third synthesis gas stream (syngas **12** of Figure 3), which feeds into the shift reactor (**12** of FIG. 3), comprises hydrogen, carbon monoxide, carbon dioxide, and unreacted methane. EX1009, p.11, ln.1–3 (“After reforming of the natural gas and cooling, the gas mixture is shifted. The gas mixture from the reformer reactor contains mainly the gas components CO, H₂, H₂O, CO₂ and some CH₄.”); EX1009, p.14, ln.22–25 (“The production of methanol and production of hydrogen have similarities in that both processes rely on reforming natural gas to a syngas mixture comprising major amounts of hydrogen and CO, with some CO₂ and residual methane and water.”).

260. Darde discloses reforming processes for producing synthesis gases comprising hydrogen, carbon monoxide, carbon dioxide, and unreacted methane. *See*, Section XI.B.2(i), *supra*.

261. Darde discloses a shift reaction processes for producing a residual gas composition comprising carbon dioxide, carbon monoxide unconverted, hydrogen not separated off, and unreacted methane; for example “a synthesis gas enriched with H₂ and CO₂ and depleted of CO compared with the gas **5** produced by reforming, and also containing additional impurities generated during the conversion of about 65% to 85% of H₂, 11 % to 22% of CO₂, 0.5% to 6% of unconverted CO and 3% to 10% of CH₄”; EX1008, p.15, ln.33–36. For the combination of ATR and GHR with downstream WGS, the hydrogen content is at the lower given range whereas the CO₂ content is at the upper given range. Due to the high outlet temperatures of ATR, the content of unreacted CH₄ is lower than the range given in Darde. Here, the series configuration results in lower content of unreacted CH₄ than the parallel configuration. It is my opinion that Rytter alone or in view of Darde discloses the third synthesis gas stream comprising hydrogen, carbon monoxide, carbon dioxide, and unreacted methane, and therefore this element of claim 1.

(j) Element 1.3.3: heat from autothermal reforming step

262. Rytter discloses the heat generated by the autothermal reforming step is utilized for heating in the endothermic reforming step of step (b). EX1009, p.10, ln.30–33, p.15, ln.2–3. Rytter discloses heat (via recycled exit gas **22** from the ATR) “... is used to heat the catalyst tubes in a GHR 1. ... The feed gas 11 first passes through the catalyst in the GHR, then the ATR 2 by stream 21 and finally the heating side of the GHR to provide the heat for the

initial reaction.” EX1009, p.10, ln.30–33. “The heat recycle **22** is the exit gas from the ATR used to heat the GHR.” EX1009, p.15, ln.2-3. It is my opinion that Rytter discloses that heat generated by the autothermal reforming step is utilized for heating in the endothermic reforming step of step (b), and therefore discloses this element of claim 1.

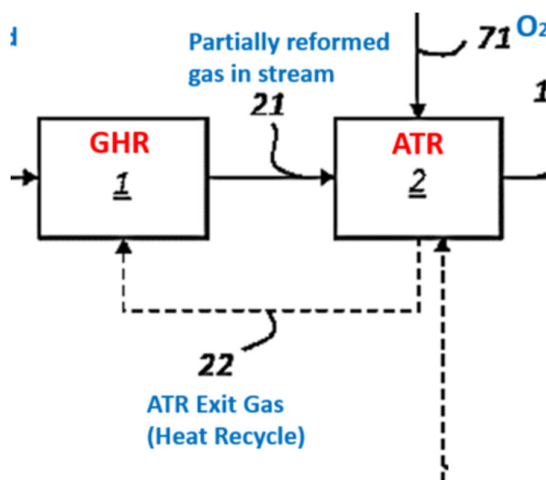


Figure 3, excerpt, annotated

(k) Element 1.4: converting the carbon monoxide

263. Rytter discloses converting the carbon monoxide present in the third synthesis gas stream with steam thereby producing hydrogen and carbon dioxide thereby producing a fourth synthesis gas stream. EX1009, p.9, ln.15–16, p.15, ln.9–12. Rytter describes using a water-gas shift (12 of FIG. 3) to convert carbon monoxide in the third gas stream using steam. EX1009, p.15, ln.9–12 (“The produced syngas **12** is shifted to increase the content of hydrogen and CO₂ in one or more shift reactors 12, i.e. water-gas-shift reactor(s) 12, to produce the shifted gas 121. Steam may be added to the gas mixture before the

gas mixture is input into the shift reactor(s) 12. The addition of steam increases the efficiency of the shift reaction.”). Rytter discloses that the result of this shift produces hydrogen and carbon dioxide as a fourth synthesis gas stream (“shifted gas 121” of Figure 3). EX1009, p.9, ln.15–16 (“ $\text{CO} + \text{H}_2\text{O} \rightleftharpoons \text{CO}_2 + \text{H}_2$ 3. Shift reaction”). It is my opinion that Rytter alone or in combination with Darde discloses converting the carbon monoxide present in a third synthesis gas stream with steam thereby producing hydrogen and carbon dioxide thereby producing a fourth synthesis gas stream, and therefore this element of claim 1.

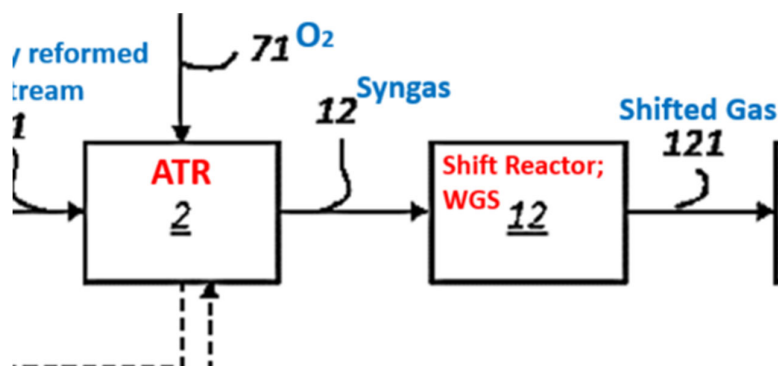


Figure 3, excerpt, annotated

(l) Element 1.4.1: fourth synthesis gas

264. Rytter discloses the fourth synthesis gas stream comprises hydrogen, carbon dioxide, unreacted methane, and carbon monoxide unconverted in step (d). EX1009, p.15, ln.9–14. Rytter discloses, “[t]he produced syngas 12 is shifted to increase the content of hydrogen and CO₂ in one or more shift reactors 12, i.e. water-gas-shift reactor(s) 12, to produce the shifted gas 121. ... The shifted gas 121 may be subsequently cooled to remove its water content. Separation process 13 separates CO₂ 131 from the shifted gas,

and the CO₂ is compressed 14 and optionally liquified.”). EX1009, p.15, ln. 9-14. In my opinion, unreacted methane and carbon monoxide unconverted in step (d) are also necessarily present in the shifted gas. *See* Ground 1, (¶¶ 140-145) *supra*.

265. Darde discloses reforming processes for producing synthesis gases comprising hydrogen, carbon monoxide, carbon dioxide, and unreacted methane. *See*, Section XI.B.2(l), *supra*. It is my opinion that Rytter alone or in view of Darde discloses a fourth synthesis gas stream comprising hydrogen, carbon dioxide, unreacted methane, and carbon monoxide unconverted in step (d) and therefore this element of claim 1.

(m) *Element 1.5: separating hydrogen*

266. Rytter discloses separating hydrogen from the fourth synthesis gas stream by pressure swing adsorption, thereby producing a hydrogen-rich stream and a residual gas stream. EX1009, p.20, ln.7–11, p.15, ln.16–17. In Figure 3, Rytter discloses a process with the hydrogen being separated “by PSA...from the gas 132 that has already been depleted of CO₂ by the process 13.”. EX1009, p.15, ln.16–17. However, Rytter further describes processes where the hydrogen is “separated from the gas mixture before the carbon dioxide is separated from the gas mixture.” EX1009, p.20, ln.7–11. Therefore, a POSA would have viewed the PSA hydrogen separation process of Rytter as capable of generating the first residual gas stream comprising CO₂, CO unconverted in step (d),

hydrogen not separated off in step (e), and unreacted methane as in the '805 patent, for example when provided upstream of cryogenic CO₂ separation.

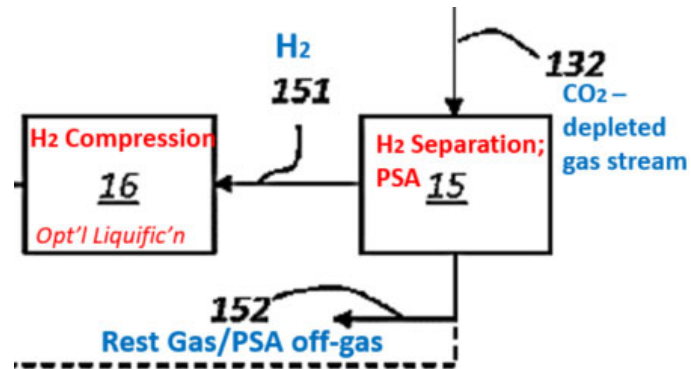
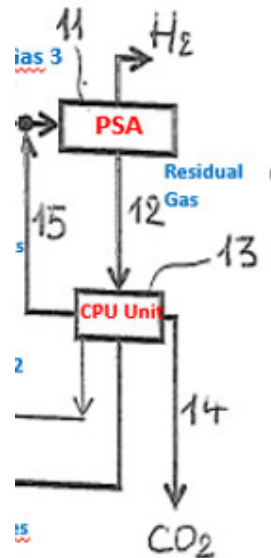


Figure 3, excerpt, annotated

267. Darde clearly discloses PSA for hydrogen separation provided upstream of cryogenic CO₂ separation, see Figure annotated excerpt below. In the upstream PSA configuration in Darde, the synthesis gas **9** is passed to a PSA unit **11** to produce a stream of highly purified hydrogen (shown exiting PSA **11**) and a residual gas **12** containing all of the carbon dioxide. EX1008, p.16, ln.6–10. The residual gas **12** is then sent to a CPU unit **13** for separation of carbon dioxide **14**.



Figure, excerpt, annotated

268. The PSA process of Darde, therefore, produces a hydrogen-rich stream and a first residual gas stream, where the first residual gas stream comprises carbon dioxide, unconverted carbon monoxide, hydrogen not separated off, and unreacted methane; for example the PSA process “produces at least one gaseous stream of highly pure (greater than 99%) hydrogen, and also a residual gas **12** which for its part contains all the carbon dioxide, the vast majority of the unconverted methane and of the unconverted carbon monoxide, and a large part of the nitrogen and hydrogen, the quality of which depends on the yield from the H₂ PSA.” EX1008, p.16, ln.6–10.

269. In my opinion, a POSA at the critical date, would have readily seen the advantages of placement of the hydrogen separation process, for example by PSA, upstream of the cryogenic CO₂ separation. For example, upstream placement of a PSA, as shown in Darde, would be viewed as providing an energy

efficient arrangement given that removal of hydrogen prior to cryogenic separation would decrease the overall energy required for cooling and compression used in a subsequent cryogenic process to separate CO₂. Therefore, a POSA would have been motivated to implement PSA upstream of the cryogenic separation as shown in Darde in the process in Rytter to achieve a subsequent cryogenic CO₂ separation at a lower overall energy. It is my opinion that Rytter alone or in combination with Darde, and for example in view of the additional disclosure in Darde regarding the sequence of PSA H₂ separation before cryogenic CO₂ separation, discloses separating hydrogen from a fourth synthesis gas stream by pressure swing adsorption, thereby producing a first hydrogen-rich stream and a first residual gas stream, and therefore this element of claim 1.

(n) *Element 1.5.1: first residual gas*

270. Rytter at least inherently discloses the first residual gas stream comprises carbon dioxide, carbon monoxide unconverted in step (d), hydrogen not separated off in step (e), and unreacted methane.

271. In my opinion, to the extent Rytter may not explicitly disclose this element, Darde discloses a PSA process for producing a residual gas composition comprising carbon dioxide, carbon monoxide unconverted in step (d), hydrogen not separated off in step (e), and unreacted methane. EX1008, p.15, ln.30–36. It is my opinion that Rytter alone or in view of Darde discloses a first residual gas stream comprising carbon dioxide, carbon monoxide

unconverted in step (d), hydrogen not separated off in step (e), and unreacted methane, and therefore this element of claim 1.

(o) Element 1.6: separating carbon dioxide

272. Rytter discloses separating carbon dioxide from the first residual gas stream obtained in step (e) by cryogenic carbon dioxide separation, thereby producing a first carbon dioxide-rich stream and a second residual gas stream. EX1009, p.15, ln.13–15, p.23, ln.5–6. Rytter discloses the separation of CO₂ via, e.g., a cryogenic process. EX1009, p.6, ln.21–24, p.23, ln.5–6 (“carbon dioxide is separated by cryogenic processes and/or a solvent that is optionally methanol.”) and Figure 3, excerpt below. In my opinion, a PSOA would have understood that if cryogenic processes are used to separate carbon dioxide in combination with PSA H₂ separation, then the PSA H₂ separation process should be positioned upstream of the cryogenic CO₂ separation, e.g., as shown in Darde, to achieve a subsequent cryogenic CO₂ separation at a lower overall energy. *See*, Section XI.B.2(o), *supra*. In my opinion, a POSA would have understood that making this modification to Rytter would result in a second residual gas stream as recited in claim 1 of the '805 Patent.

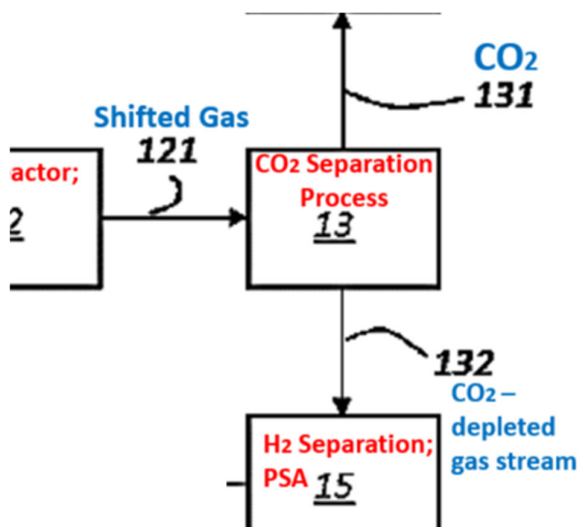


Figure 3, excerpt, annotated

273. Darde discloses cryogenic CO₂ capture using a CO₂-purifying CPU unit for producing a first carbon dioxide-rich stream and a second residual gas stream, where the second residual gas stream comprises carbon monoxide unconverted, hydrogen not separated off, carbon dioxide not separated off, and unreacted methane; for example, “The residual gas **12** is sent at **13** to a CPU unit for separation of the CO₂, the CPU unit producing at least one stream **14** of CO₂, a gaseous stream **15** rich in H₂ which is recycled so as to feed the hydrogen PSA in order to recover the hydrogen contained and to thus improve the overall hydrogen yield of the facility, a stream **16** of non-condensables comprising methane, hydrogen, carbon monoxide, non-condensed carbon dioxide, nitrogen and water, constituting the CPU offgas which is recycled as reforming fuel to the reforming module **4**”. EX1008, p.16, ln.11–16. It is my opinion that Rytter in view of Darde discloses separating carbon dioxide from a

first residual gas stream obtained in step (e) by cryogenic carbon dioxide separation, thereby producing a first carbon dioxide-rich stream and a second residual gas stream RG2, and therefore this element of claim 1.

(p) Element 1.6.1: second residual gas

274. Rytter discloses the second residual gas stream comprises carbon monoxide unconverted in step (d), hydrogen not separated off in step (e), carbon dioxide not separated off in step (f), and unreacted methane. EX1009, p.15, ln.18–20 (“The rest gas **152** contains remnants of CO and CH₄ together with unseparated CO₂ and hydrogen.”); Figure 3, excerpt below.

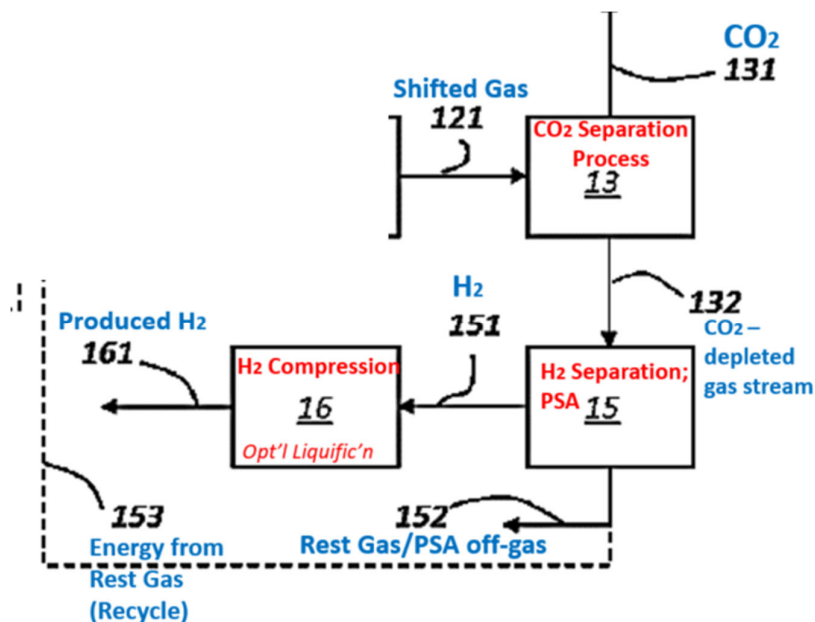


Figure 3, excerpt, annotated

275. In my opinion, to the extent that Rytter does not disclose this element, Darde discloses the second residual gas stream comprises carbon monoxide unconverted, hydrogen not separated off, carbon dioxide not

separated off, and unreacted methane. *See*, Section XI.B.2(p), *supra*. It is my opinion that Rytter in view of Darde discloses a second residual gas stream comprising carbon monoxide unconverted in step (d), hydrogen not separated off in step (e), carbon dioxide not separated off in step (f), and unreacted methane, and therefore this element of claim 1.

(q) *Element 6: first carbon dioxide-rich stream is subjected to a thermal separation process for separation of methane from carbon dioxide*

276. Element 6 recites a limitation of “(t)he process according to claim 1, where the first carbon dioxide-rich stream comprises unreacted methane, and the first carbon dioxide-rich stream is subjected to a thermal separation process for separation of methane, thereby producing a second carbon dioxide-rich stream.” It is my opinion that Darde discloses the first carbon dioxide-rich stream comprises unreacted methane, and the first carbon dioxide-rich stream is subjected to a thermal separation process for separation of methane, thereby producing a second carbon dioxide-rich stream.

277. Darde recites the liquid CO₂ generated by cryogenic CO₂ separation can be distilled to achieve CO₂ purities greater than 99%. *See*, Section XI.B.2(q), *supra*. As known in the art by a POSA as of the critical date and as described in the '805 Patent, distillation is a thermal separation process for separation of methane. It is my opinion that Rytter in view of Darde discloses the first carbon dioxide-rich stream is subjected to a thermal

separation process for separation of methane, thereby producing a second carbon dioxide-rich stream, and therefore this element of claim 6.

(r) *Element 11: compression step and cooling step*

278. I emphasize that cryogenic CO₂ separation was known in the art long before the critical date to include compression and cooling steps. See ¶40, *supra*; EX1018, EX1020; EX1021. In my opinion, a POSA at the critical dated would have understood that the cryogenic CO₂ separation process of Rytter, as in that of Reinertsen, necessarily includes the at least one compression step and at least one cooling step of claim 11 the '805 patent, ¶159, *supra*. In addition, the terms “cryogenic distillation” and “cryogenic process”, as used by Rytter, would have been readily understood by a POSA as examples of a process including cooling and condensation steps of a gas stream containing CO₂ in a manner necessarily producing a CO₂ rich stream and residual gas stream. Furthermore, Darde discloses that compression and cooling steps, as recited in claim 11, are steps in cryogenic CO₂ separation which is applied to residual gas from PSA separation. EX1008, p.8, ln.12–21. Based on this teaching in Darde, the teachings in Rytter and the general skill and knowledge in the art, a POSA would have understood to implement cryogenic CO₂ separation following PSA for hydrogen separation using at least one compression step and at least one cooling step to produce a carbon dioxide-rich stream at least partly in the form of a condensed carbon dioxide stream. In my opinion, Rytter in combination

with Darde, in view of what was known in the art, teaches all of the elements of claim 11 of the '805 Patent.

279. Darde explicitly discloses this feature in connection with cryogenic carbon dioxide separation from a first residual gas from PSA separation of hydrogen from shifted synthesis gas. See, ¶¶266-273, *supra*. For example, Darde discloses that compression and cooling steps, as recited in claim 11, are steps in cryogenic CO₂ separation. EX1008, p.8, ln.12–21 (“(CPU) calls for the partial condensation and/or the distillation of the CO₂ contained in the CO₂-rich gaseous stream in a cryogenic purification unit (CPU) ... The gas, after drying and compression to a pressure between 20 and 100 bar(a), is cooled to a temperature close to that of the triple point of CO₂ (approximately -56 °C).”). In my opinion, therefore, the cryogenic separation processes described in Darde and Rytter, taken alone or in combination, includes a compression step and a cooling step as provided in claim 11 of the '805 Patent.

(s) *Element 12: a plant configured for performance of the process*

280. Element 12 recites “a plant configured for performance of the process according to claim 1.”

281. In my opinion, the combination of Rytter and Darde discloses all the elements of the process of claim 1. *See*, Section XI.B.2.(a)-(p), *supra*. Rytter further discloses a plant configured for performance of the hydrogen separation

process. (“Figure 3 shows a configuration of a hydrogen production plant according to an embodiment.”) EX1009, p.8, ln.14, Figure 3. In my opinion, the plant of claim 12 is defined by the process of claim 1. No additional elements of the plant are provided. In my opinion, therefore, the combination of Rytter and Darde teaches each and every element of claim 12 as recited in the ’805 Patent.

3. Reasonable Expectation of Success

282. In my opinion, a POSA would have been readily able to adapt the teachings of Rytter with Darde to arrive at the process of claims 1, 6, 11 and 12 of the ’805 Patent with a reasonable likelihood of success.

283. The processes in Rytter with Darde are overlapping and used in a similar manner to achieve the same goal of hydrogen production with reduced CO₂ emissions. Rytter describes a process for producing hydrogen from a hydrocarbon-rich feed gas with reduced CO₂ involving i) a reforming process including an endothermic reforming step using a GHR and an autothermal reforming step using an ATR, where GHR and ATR components are provided in a series configuration and where heated gas from the ATR is utilized for heating the GHR; (ii) downstream conversion of CO to CO₂ via water gas shift conversion; and (iii) separation processes including PSA for hydrogen separation and cryogenic CO₂ separation. Darde discloses a similar process using methane reforming (e.g., SMR or ATR) with downstream water shift

conversion, and subsequent PSA for hydrogen separation provided upstream of cryogenic CO₂ capture.

284. When combined with Rytter, which also provides for PSA as an alternative, the processes disclosed in Darde, provides sufficient teaching to allow a POSA to select and implement from the finite number of process options disclosed in Rytter (i.e., Pd-membrane and PSA) a configuration having PSA for hydrogen separation provided upstream of cryogenic CO₂ separation. The guidance provided in Darde, therefore, would have been sufficient to allow a POSA to select for selection of the sequence of PSA provided upstream of cryogenic CO₂ separation and successful integration into the process of Rytter to predictably arrive at the processes as claimed in the '805 Patent with a reasonable expectation of success, for example, to provide a hydrogen product stream at moderate cost with sufficient purity for applications including clean energy and manufacturing of high value chemicals such as ammonia. In my opinion, any modification or other adaptation needed to successfully combine Rytter with Darde in this manner would have been a matter of routine engineering and, thus readily within the skill of a POSA at the critical date.

285. A POSA would also expect that the reforming and separation processes of Rytter and Darde would be compatible with each other. Rytter and Darde include known reforming, water-gas shift and separation processes having similar and overlapping process conditions, such as, pressures, flow

rates and temperatures; similar process inputs such as feed stream; and similar process outputs such the synthesis gas streams, hydrogen-rich stream, residual streams and CO₂ rich stream. Therefore, a POSA would require no more than ordinary skill to adapt the teachings of Rytter and Darde to implement any necessary adaptation or modification using a practical selection of known processes and process conditions used in their ordinary manner. Given that these well-known processes are used in Rytter and Darde in the same manner to achieve the same technical solution of hydrogen generation with reduced CO₂ emissions, a POSA would have been readily able to adapt the teachings of these references to arrive at the claimed invention with a reasonable likelihood of success.

4. The Claims are Obvious over the Combination of Rytter and Darde

286. The combination of Rytter and Darde discloses all of the features of Claims 1, 6, 11 and 12. It is my opinion that these claims are obvious, and not patentable under 35 U.S.C. § 103.

D. Petition I Ground 4: Claims 2-5 are Obvious over Reinertsen in Combination with Darde and in Further View of Terrien

287. It is my opinion that claims 2-5 would have been obvious to a POSA based on the combination of Reinertsen (EX1005), with Darde (EX1007), and in further view of Terrien (EX1024).

1. Motivation to Combine, Reasonable Expectation of Success

288. In my opinion, the combination of Reinertsen, Darde and Terrien teaches or suggests each and every element of claims 2–5.

289. In my opinion, motivation to combine the teachings of Reinertsen and Darde in the manner claimed is provided above. *See*, Section XI.B.1, *supra*. Reinertsen and Darde each share a recognized goal of producing hydrogen via gas reforming and separating and capturing CO₂, thereby decreasing CO₂ emissions. This common goal, as would be recognized by a POSA at the critical date, is achieved in Reinertsen and Darde via an energy efficient cryogenic capture solution, which allows for sequestration of CO₂ and/or generation of a CO₂ coproduct, in both cases reducing or preventing CO₂ emissions; e.g., to the environment.

290. Similarly, Terrien relates to a process for more efficient recovery of hydrogen and CO₂, which similar to Reinertsen and Darde, is applied and exemplified by recovery of hydrogen and CO₂ from synthesis gas generated through reforming of hydrocarbons and shifting the synthesis gas to increase hydrogen in the synthesis gas. EX1024, ¶[0019]. Terrien teaches additional steps of generating and using hydrogen-rich gas streams as feed into a PSA unit or as fuel for heating in a reformer. Terrien's goal is at least the efficient recovery of hydrogen and carbon dioxide (EX1014, ¶[0016]) and, as applied to hydrogen plants, Terrien's goals include at least recovery of additional

hydrogen and carbon dioxide to provide “significant savings with regards to the size of reformer, natural gas consumption, carbon dioxide tax, etc.” EX1024, ¶[0006]. The goals of Terrien are, thus, the same or similar to those of Reinertsen and Darde for increased efficiency of hydrogen production and decreased CO₂ emissions to, for example, save on carbon tax. Terrien, like Reinertsen and Darde, employs a PSA process to separate a first purified hydrogen-rich product stream followed by cryogenic CO₂ separation to produce a CO₂-enriched product stream. Terrien FIG. 5 provides an exemplary process for improved recovery of hydrogen and carbon dioxide from a process stream (1) obtained from a process unit (0). EX1024, ¶[0077]–[0078]. The process unit (0) is any unit which includes a purification step that results in a hydrogen rich fraction, e.g., stream 27 (EX1024, ¶[0018]) and is preferably a PSA unit that is used to recover and purify hydrogen. EX1024, ¶[0020]. Terrien describes that increased recovery of hydrogen and carbon dioxide is achieved by balancing the recycle of the hydrogen-rich permeate from hydrogen membrane separation units. EX1024, Abstract, ¶[0008]. Terrien specifically teaches steps of generating hydrogen-rich gas streams 5 and 12 using hydrogen selective membranes (4 and 11). EX1024, FIG.5. and FIG. 6; ¶[0025], [0041]. This is in addition to hydrogen-rich stream 27 which is separated in unit 0 for example using a PSA. I note that hydrogen-rich gas stream 12 is formed by passing carbon dioxide lean non-condensable stream 10 through hydrogen

selective membrane 11. EX1024, ¶[0077]. Terrien further discloses using the hydrogen-rich gas stream (27, 5 and 12) as feed into a PSA unit or as fuel for heating in a reformer. EX1024, ¶¶[0046], [0068], [0078], [0085]. In my opinion, a POSA would have been motivated to combine the teachings of Terrien with those of Reinertsen and Darde because each of the processes are directed to hydrogen production with low CO₂ emissions or improved capture of CO₂ and each of the processes use PSA hydrogen separation and cryogenic CO₂ separation for H₂ and CO₂ separation/capture. Each of the combined references addresses the same or similar problems, shares common goals and teaches the same or similar solutions to the problems. In Terrien, additional steps of hydrogen separation using hydrogen selective membranes are employed and hydrogen-rich gas streams are recycled to feed a PSA unit to enhance the effectiveness of hydrogen capture (i.e., recycled to the process unit 9). EX1024, ¶[0045]. Terrien also describes recycling of hydrogen-rich gas streams as fuel to a reforming unit to boost the percentage of carbon dioxide capture. EX1024, ¶[0078]. The goals of Reinertsen and Darde to produce hydrogen and lower emissions of CO₂ overlap with those of Terrien to increase recovery of hydrogen and carbon dioxide. In my opinion, a POSA would have been further motivated to combine the teachings of Terrien with those of Reinertsen and Darde to increase the efficiency of recovery of desirable products in the combined process and to increase at least the energy efficiency of the combined

process by recycling of product gases to feed and/or fuel the reformers in the process.

291. In my opinion, modifications of the combined process of Reinertsen and Darde with the additional purification and recycling steps as taught by Terrien would not change the principles of operation of the combined process of Reinertsen and Darde. The processes in Reinertsen and Darde employ the same feed gases in the same process steps (reforming, water-gas shift, PSA separation and cryogenic CO₂ separation), generate the same intermediates (syngas and shifted syngas) and the same products (H₂ and CO₂), and, therefore, a POSA would have recognized that modification using Terrien's additional methods would not change the principles of operation of a combined Reinertsen and Darde process and would further result in recognized benefits, such as enhanced recovery of hydrogen and carbon dioxide which at least improves process efficiency and economics.

292. In my opinion, the combination of the additional process steps of Terrien with those of Reinertsen and Darde are well within the ordinary skill of a POSA and represent the combination of known methods and process equipment to provide their known and intended function. In my opinion, the additional process steps of Terrien are compatible with the processes and process conditions in Reinertsen and Darde, such as conditions of temperature, pressure and flow rate. Because the additional steps of Terrien were well known

in the art at the critical date and their function and implementation were also then well-known and understood, in my opinion a POSA would have had a reasonable expectation that the additional process steps of Terrien could be successfully implemented in the combined process of Reinertsen and Darde and that the desired additional benefits of their implementation as taught by Terrien would be achieved. In my opinion, the combination of the additional process steps/process units of Terrien with those of the combined teaching Reinertsen and Darde were well within the ordinary skill of a POSA at the critical date and represent the combination of known methods and process equipment to provide their intended function.

293. With respect to claim 2, in my opinion, a POSA would have been motivated to apply any of the additional hydrogen separation steps as taught by Terrien to the combined method of Reinertsen and Darde to increase the yield of the desired hydrogen product by separating at least an additional hydrogen-rich stream, would have known in view of what was known in the art and the teachings of these references how to implement the separations and would have reasonably expected success in that implementation.

294. With respect to claim 3, in my opinion, a POSA would have been motivated to employ any of the additional specific membrane hydrogen separation steps as taught by Terrien to the combined method of Reinertsen and Darde again to increase the yield of the desired hydrogen product by separating

an additional hydrogen-rich stream. The use of a hydrogen selective membrane to separate hydrogen from synthesis gas product and residual gas streams was well known in the art at least by the critical date. In my opinion, a POSA would have known how to implement the membrane separation at least in view of what was known in the art, and the teachings of the references and would have reasonably expected success in that implementation.

295. With respect to claim 4, Terrien teaches separation of a second hydrogen-rich stream using membrane separation and recycling of that stream to a pressure swing adsorption unit. EX1024, ¶[0045]. Hydrogen separation/purification by use of membranes and pressure swing absorption processes was well known in the art at least at the critical date. In my opinion, a POSA would have been motivated to employ the hydrogen recycling taught by Terrien to the combined method of Reinertsen and Darde to increase the yield of the desired hydrogen product, but also to generally increase the efficiency of the entire hydrogen generation process. In my opinion, a POSA would have known how to implement the hydrogen recycling at least in view of what was known in the art and the teachings of the references and would have reasonably expected success in that implementation.

296. With respect to claim 5, Terrien teaches the use of membrane separated hydrogen rich gas streams as fuel gases for heating a reformer, specifically exemplified as a steam methane reformer (EX1024, ¶[0078]), but

where an ATR (autothermal reformer) is taught to be a suitable alternative reformer (EX1024, ¶¶[0019], [0072]). In my opinion, a POSA would have been motivated to employ the recycling of hydrogen-rich permeate of Terrien to the ATR of the combined method of Reinertsen and Darde in view of the teachings of the references and what was known in the art at the time for heating of feed gas to autothermal reformers to at least improve capture of carbon dioxide. It was known in the art long before the critical date that it was beneficial to provide a fired heating unit to heat feed gas entering the autothermal reformer. I note that Reinertsen acknowledges the use of the energy of rest gas generated from CO₂ separation for fuel in fired heaters for preheating of feed gases natural gas and water/steam. EX1005, ¶[0103]. Thus, in my opinion, a POSA would have recognized that in the combined Reinertsen and Darde process employing an endothermic reforming step (GHR) and an autothermal reforming step that recycling of hydrogen-rich permeate to fuel a reformer as taught by Terrien would have provided increased efficiency of carbon capture. In my opinion, a POSA would have known how to implement the hydrogen recycling to fuel at least to a feed gas heating unit or for preheating feed gases for a gas reformer, as described in Reinertsen, at least in view of what was known in the art and the teachings of Terrien and would have reasonably expected success in that implementation again in view of the similarities in method steps, process units, intermediates and final products.

2. Reinertsen and Darde in View of Terrien Disclose the Limitations of Claims 2-5

(a) *Claim 2: hydrogen is separated from the second residual gas stream, thereby producing a second hydrogen-rich stream and a third residual gas stream*

297. In my opinion, the combination of Reinertsen and Darde teaches each and every element of claim 1 from which claim 2 depends. *See*, Sections XI.A.1–15 and XI.B.2(a)–(p), *supra*.

298. In the '805 Patent RG2 is residual gas exiting the cryogenic CO₂ separation unit **205** and is described as being introduced into a membrane unit **206** to separate H₂ and generate HG2 enriched in hydrogen and a third residual gas stream RG3. EX1001, col.12, ln.20–29, Figs. 2–3.

299. The combination of Reinersten and Darde discloses generating a hydrogen-rich stream and residual gas stream. *See*, Section XI.B.2(q), *supra*.

300. Terrien FIG. 5 illustrates CO₂ lean non-condensable stream **10** from a CO₂ separation unit **8**, which in view of the compression/liquefaction and distillation steps and components described/illustrated contains a cryogenic CO₂ separation unit. EX1024, ¶¶[0037]–[0038]. Stream **10** from cryogenic CO₂ separation unit **8** clearly contains hydrogen because it is passed to hydrogen selective membrane **11** for additional separation of hydrogen to generate hydrogen-rich permeate **12** and a second hydrogen lean residual stream **13**. EX1024, ¶¶[0041]–[0042].

301. Terrien in FIG. 5 describes a carbon dioxide lean non-condensable stream **10** from a CO₂ separation unit **8**. The CO₂ separation unit **8** is described. EX1024, ¶[0038]. In view of the description of compression/liquefaction and distillation steps and the components described (EX1024, ¶[0038]) and the components and process flow illustrated (EX1024, FIG. 4), the CO₂ separation unit **8** is in my opinion a cryogenic CO₂ separation unit. The carbon dioxide lean non-condensable stream **10** from this cryogenic CO₂ separation unit clearly contains hydrogen because Terrien states that it is passed to hydrogen selective membrane **11** for additional separation of hydrogen to generate hydrogen-rich permeate **12** (EX1024, ¶[0041]) and a second hydrogen lean residual stream **13** (EX1024, ¶[0042]).

302. In my opinion:

(1) carbon dioxide lean non-condensable stream **10** of Terrien is a highly carbon dioxide depleted residual gas stream of cryogenic CO₂ separation, the same as the second residual gas RG**2** of the '805 Patent;

(2) hydrogen rich permeate **12** of Terrien is a hydrogen-rich stream enriched with hydrogen compared to **10** by passage through membrane **11**, the same as the second hydrogen rich gas HG**2** of the '805 Patent; and

(3) second hydrogen lean residual stream **13** of Terrien is a residual gas stream depleted of hydrogen compared to carbon dioxide lean non-condensable stream **10** since preferably hydrogen permeates through membrane **11**, the same

as the third residual gas RG3 of the '805 Patent. Thus, in my opinion the combination of Reinersten and Darde in view of Terrien teaches producing a second hydrogen-rich stream and a third residual gas stream as recited in claim 2 of the '805 Patent.

(b) Claim 3: hydrogen is separated from the second residual gas stream (RG2) by membrane separation.

303. In my opinion, the combination of Reinersten and Darde in view of Terrien teaches all of the elements of '805 Patent claim 2 from which claim 3 depends. *See*, Sections XI.A.1–15, XI.B.2(a)–(q), and XI.D.2(a), *supra*.

304. In the '805 Patent, RG2 exits cryogenic CO₂ separation unit **205** and is described as being introduced into a membrane unit **206** to generate HG2 and RG3. EX1001, col.12, ln.20–29.

305. The combination of Reinersten and Darde discloses generating a hydrogen-rich stream and residual gas stream. *See*, Section XI.B.2(q), *supra*.

306. In my opinion, hydrogen rich permeate **12** of Terrien is a second hydrogen-rich gas stream HG2 as described in the '805 Patent and second hydrogen lean residual stream **13** of Terrien is a third residual gas stream RG3 as described in the '805 Patent. In FIG. 5 of Terrien stream **10** is passed to hydrogen selective membrane **11** to generate hydrogen-rich permeate **12**. In my opinion, the hydrogen selective membrane **11** of Terrien is the same as the membrane unit **206** of the '805 Patent. Thus, in my opinion the combination of Reinersten and Darde in view of Terrien teaches producing a second hydrogen-

rich stream and a third residual gas stream using membrane separation as claimed in claim 3 of the '805 Patent.

(c) *Claim 4: the second hydrogen-rich stream is supplied to the fourth synthesis gas stream for separation of hydrogen by pressure swing adsorption in step (e).*

307. In my opinion, the combination of Reinersten and Darde in view of Terrien teaches all of the elements of claim 2 from which claim 4 depends. *See*, Sections XI.A.1–15, XI.B.2(a)–(q), and XI.D.2(a), *supra*.

308. In the '805 Patent, HG2 exits membrane unit **206** (Figures 2 and 3) and is optionally recycled to the synthesis gas stream SG4 which enters PSA unit **204** to increase overall hydrogen yield. EX1001, col.12, ln.29–31.

309. Terrien describes carbon dioxide lean non-condensable stream **10** which contains hydrogen from a cryogenic CO₂ separation unit **8**. EX1024, FIG. 5; ¶[0077]. Stream **10** is introduced to hydrogen selective membrane **11** to generate hydrogen rich permeate **12**. EX1024, ¶¶[0041], [0077]. The hydrogen-rich permeate **12** is recycled to feed gas stream **19** which can be synthesis gas (EX1024, FIG. 5). Feed gas **19** enters process unit **0** which contains a hydrogen separation unit, specifically a PSA unit. EX1024, ¶[0020]. In my opinion, hydrogen-rich permeate **12** is the same as a second hydrogen-rich gas stream HG2 as described in the '805 Patent, stream **19** is the same as a fourth synthesis gas stream SG4 as described in the '805 Patent. Process unit **0** contains a PSA unit which is the same as PSA unit **204** of the '805 Patent. Thus, the combination of Reinersten and Darde in view of Terrien discloses supplying the

second hydrogen-rich stream HG2 to the fourth synthesis gas stream SG4 for separation of hydrogen by pressure swing adsorption in step (e) as claimed in claim 4 of the '805 Patent.

(d) Claim 5: gases present in the second hydrogen-rich stream are utilized as fuel gases for heating in the autothermal reforming step of step (c).

310. In my opinion, the combination of Reinersten and Darde in view of Terrien teaches all of the elements of claim 2 from which claim 5 depends. *See*, Sections XI.A.1–15, XI.B.2(a)–(q), and XI.D.2(a), *supra*.

311. The '805 Patent states that “gases present in the hydrogen-rich stream HG2 are utilized as fuel gases for heating in the autothermal reforming step of step (c). EX1001, col.6, ln.9–11. It is explained in the '805 Patent that for the autothermal reforming step, the feed gas has to be heated which can be done using a fired heating unit. EX1001, col.6, ln.9–13. The fired heating unit of the autothermal reformer may be heated at least partly with the aid of gases present in HG2 that have high calorific value (CO, methane and hydrogen). EX1001, col.6, ln.11–18. Calorific value, as is known in the art, is the amount of heat energy present in a fuel determined by the complete combustion of a specified quantity of fuel at constant pressure and in normal conditions.

312. Reinertsen and Darde disclose generating a hydrogen-rich stream and residual gas stream, and processes using a hydrogen rich stream as fuel for a fire heater. *See*, Sections XI.B.2(q)–(t), *supra*.

313. Terrien teaches the recycling of hydrogen-rich gases and residual gases as feed or fuel to improve efficiency of hydrogen and CO₂ recovery. EX1024, Abstract. In my opinion, Terrien teaches recycling of hydrogen-rich streams **27**, **5** and **12** as feed and fuel, particularly in hydrogen generation plants. EX1024, ¶[0077]. For example, in Terrien FIG. 5, hydrogen-rich permeate **12** is shown as optionally recycled to SMR **23** as feed entering feed stream **22** or directly into the reformer unit as fuel. Terrien explains that when the method is employed in a hydrogen generation plant as shown in FIG. 5, hydrogen-rich permeate stream **12** is preferably used as a fuel to SMR **23** resulting in a boost in CO₂ capture (EX1024, ¶[0078]) and to eliminate or minimize natural gas fuel (EX1024, ¶[0078]). In my opinion, while Terrien specifically exemplifies the use of a SMR in the processes of FIGs. 5 and 6, Terrien also teaches that an ATR, among others, can be used to generate feed gas for the purification process. EX1024, ¶[0016]. In my opinion, the teaching of Terrien to employ hydrogen-rich permeate **12** as fuel in SMR and the related teaching of Terrien that reforming can be accomplished with an ATR would provide a strong teaching and motivation to a POSA to employ hydrogen-rich permeate **12** as fuel in an ATR, for example, to enhance overall energy efficiency of the process.

314. In my opinion, a POSA would have recognized that hydrogen-rich streams generated from hydrocarbon reforming as in the Reinertsen/Darde

combination would be useful for fuel to heat feed entering the ATR and would result in increased efficiency of hydrogen production and carbon dioxide recovery. In my opinion, a POSA would have been specifically motivated in view of Terrien's teachings to employ a hydrogen-rich gas stream, generated by removing hydrogen from a residual gas stream from a cryogenic CO₂ separation step, as fuel to heat the ATR in the Reinertsen/Darde process to achieve the benefits taught by Terrien. Thus, the combination of Reinertsen/Darde, in view of Terrien, provides sufficient teaching and guidance to utilize gases present in a second hydrogen-rich stream as fuel gases for heating in the autothermal reforming step as recited in claim 5 of the '805 Patent.

E. Petition II Ground 1: Claims 1, 6, 11 and 12 are Obvious over Martin and Rafati

315. It is my opinion that claims 1, 6, 11 and 12 would have been obvious to a POSA under 35 U.S.C. § 103 in view of Martin and Rafati.

1. Martin and Rafati—Motivation to Combine

316. In my opinion, Martin alone discloses processes within the scope of claims 1, 6, and 11 detailing all the steps of hydrogen production, CO₂ separation, and CO₂ purification. Because Martin fully discloses the process of claim 1, the reference also fully discloses a plant configured for performance of this process as claimed in claim 12.

317. In my opinion, to the extent Martin does not disclose the specific details of each of the claimed elements, there are several reasons why a POSA

would have been motivated to supplement Martin with the details disclosed in Rafati.

318. Martin describes all the process steps and their sequence for the parallel configuration of claim 1 of the '805 Patent. As a first matter, Martin discloses a combination reforming process, which includes an endothermic reforming step using a GHR and an autothermal reforming step using an ATR provided in a parallel configuration. To enhance efficiency, Martin describes that the high-temperature gas from the ATR provides the heat necessary for endothermic steam methane reforming in the GHR. Martin further describes that this process uses water gas shift conversion to convert CO to CO₂, and separation processes including a hydrogen separation via PSA provided upstream of cryogenic CO₂ separation. EX1006, p.11–12.

319. Rafati similarly describes all the process steps, in the same sequence, as the process of claim 1 of the '805 Patent and Martin. While both references are directed to the same methods and overall purpose, Rafati provides more detailed descriptions than Martin. Having reviewed both references, it is clear that the teachings of Rafati could be seamlessly integrated into Martin's process if additional guidance was needed. Thus, a POSA employing Martin's method and requiring clarification would have been motivated and encouraged to look to Rafati's descriptions.

320. This encouragement stems from Martin's and Rafati's shared purpose of efficiently producing hydrogen and separating CO₂ coproduct while reducing CO₂ emissions. Indeed, Rafati provides detailed discussion on how a dual reforming system advances this purpose. For example, Rafati describes, "An advantage of the two reactor configuration is that the yield of H₂+CO from hydrocarbon feed is maximized, and all CO₂ formed in the reactions is contained within the high-pressure system." EX1015, ¶[0115]. Martin similarly explains that downstream separation of CO₂ is "more facile" when a combination of partial oxidation and steam methane reforming reactors are used as compared to a single SMR process: "[A] two-stage high pressure syngas generation system utilizing a combination of partial oxidation and steam methane reforming reactors [i]n this way, all CO₂ generated due to the reforming reactions [to be] contained within the high pressure syngas loop which makes the downstream separation of CO₂ more facile." EX1006, p.11.

321. Additionally, in my opinion, the benefits of the use of a parallel dual reformer configuration were well known to a POSA prior to the critical date. For example, Patwardhan (EX1011) shows that a combination of a GHR with an ATR arranged in parallel with heat transfer from the ATR to the GHR was known to provide the benefits of saving on natural gas as fuel at the expense of steam production. Specifically, Patwardhan acknowledges "[n]ew process schemes such as the autothermal reformer and the gas-heated reformer

in series or parallel combination with SMR or ATR are gaining prominence, as they offer potential in reducing energy consumption and flue gas emissions.”

EX1011, p.6.

322. Martin and Rafati also describe the same downstream separation steps (PSA + cryogenic separation), in the same order, to efficiently produce hydrogen and reduce CO₂ emissions. Rafati explains the benefits of the downstream separation steps in stating: “The systems and methods of the present disclosure can be utilized to capture substantially 100% of the CO₂ from hydrogen generating processes by recovering CO₂ from a pressure swing absorber (PSA) waste stream. Separation of CO₂ from PSA off-gas increases the hydrogen concentration in CO₂ separation train waste gas to at least about 60 mol % which would make it suitable and economic for additional H₂ recovery within a second PSA.” EX1015, ¶[0111]. Martin similarly discusses the advantages of positioning a PSA upstream of cryogenic CO₂ separator, “the PSA waste gas contains about 76% CO₂ (mole basis) which is suitable for processing in a low temperature removal system.” EX1006, p.12. Therefore, in my opinion, there is sufficient teaching and guidance for a POSA to fill in any perceived gaps in Martin’s downstream separation processes with the detail provided in Rafati.

323. In my opinion, the benefits of combining a dual reformer with downstream PSA/cryogenic separation steps in enhancing efficiency were well

known in the art prior to the critical date. For example, Patwardhan (EX1011) acknowledges these benefits by explaining “[n]ew process schemes such as the autothermal reformer and the gas-heated reformer in series or parallel combination with SMR or ATR are gaining prominence, as they offer potential in reducing energy consumption and flue gas emissions.” *See*, EX1011, p.6. Patwardhan further reports numerical simulations showing significant reductions in “relative flue gas emission, CO₂ %” of approximately 5% are achievable via a GHR and ATR reformer configuration with heat exchange with downstream H₂ purification using PSA. *See*, EX1011, Table 3, p.5–6. In my opinion, a POSA would have been motivated to combine the teachings of Martin and Rafati to achieve the benefits acknowledged in Patwardhan of reduced energy consumption and flue gas emission.

324. In my opinion, the specific recitations of stream compositions and details of process steps of the '805 Patent are inherent in the process of Martin, because these are obvious extensions of well-known techniques for the parallel configuration of H₂ production and CO₂ separation having downstream PSA and cryogenic separation steps. *See, e.g.*, Section V., ¶¶28–31, 37–40, *supra*. However, such detailed descriptions are expressly provided in Rafati. Therefore, the combination of Martin with Rafati discloses all elements of the process steps and stream compositions described in claim 1 of the '805 Patent.

325. Moreover, in my opinion, a POSA would have had the requisite knowledge and experience to implement the process of Martin to achieve the gas and product stream compositions explicitly disclosed in Rafati. Given that Martin uses the same combination of methane reforming as Rafati, as well as the same downstream conversion and separation processes in the same order, introducing a methane feed gas to Martin's process would necessarily produce the same gas and product stream compositions described in Rafati. Thus, use of the techniques in Martin and Rafati would predictably achieve the gas stream compositions in the same way and with predictable results as provided in claim 1 of the '805 Patent.

2. Martin and Rafati Combine to Disclose the Limitations of the Claims

326. The combination of Martin and Rafati provides all the elements of claims 1, 6, 11 and 12 of the '805 Patent.

(a) Element 1.pre: Preamble

327. Martin discloses a process for preparing hydrogen by reforming hydrocarbons with steam, and for separation of carbon dioxide. *See, generally, Figure 7, as annotated below:*

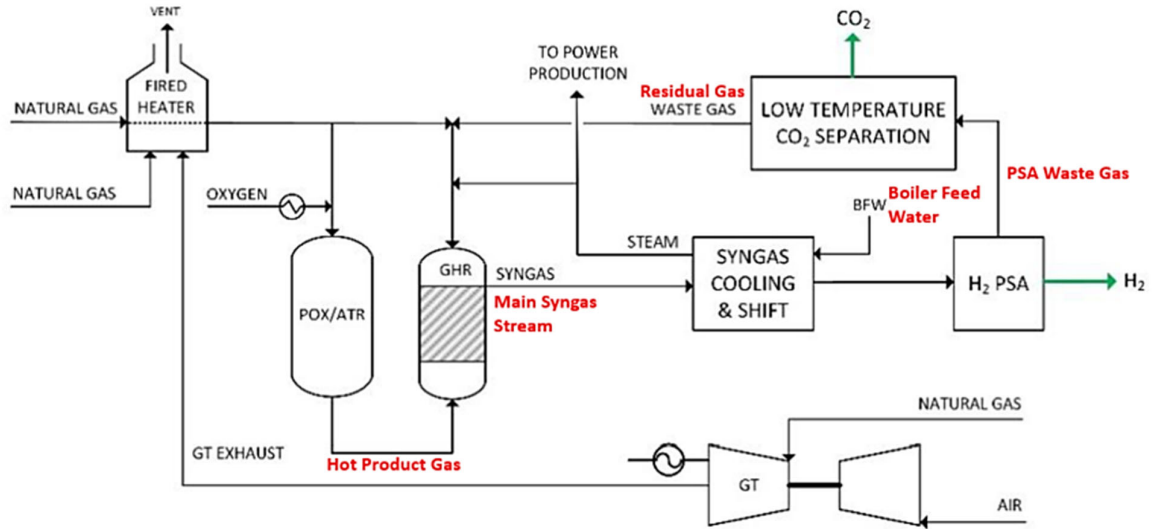


Figure 7, annotated

328. Martin describes Figure 7 as a “Simplified block flow diagram of 8RH2 process for hydrogen generation with carbon capture.” EX1006, p.12. Figure 7 shows the reforming of hydrocarbons (natural gas) using steam and “LOW TEMPERATURE CO₂ SEPARATION.” *Id.* Martin further describes another steam methane reforming process for reforming of hydrocarbons (natural gas) using steam in order to produce hydrogen, as shown in Figure 6 below:

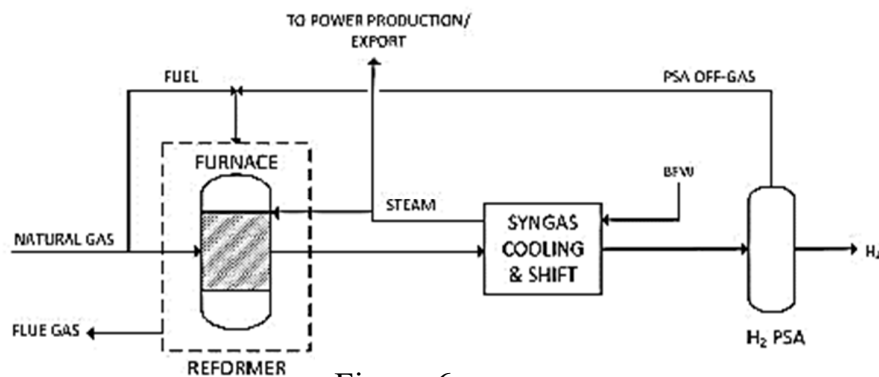


Figure 6

(b) Element 1.1: providing a feed gas stream

329. Martin discloses providing a feed gas stream. EX1006, p.12. For example, Martin describes, “[i]n the combined reforming concept, natural gas is partially oxidized in an oxygen deficient environment,” and depicts the natural gas first going through a fired heater, thereby generating a feed gas stream, as shown in the excerpt from Figure 7 below. EX1006, p.12.

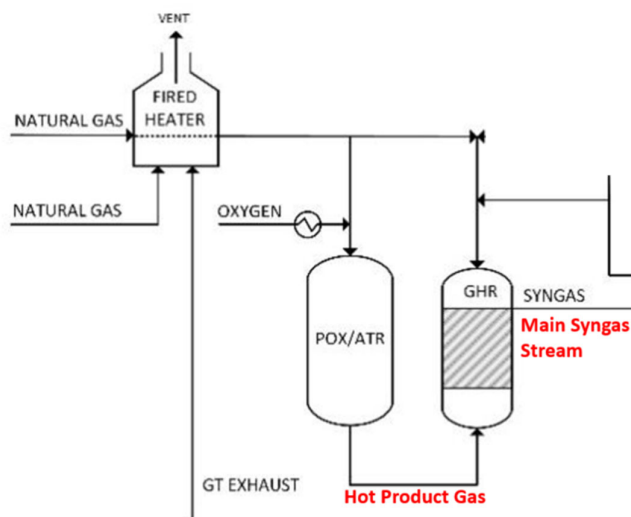


Figure 7, excerpt, annotated

(c) Element 1.1.1: hydrocarbon component and steam

330. Martin discloses the feed gas stream comprises a hydrocarbon component and steam. EX1006, p.12. Martin explicitly discloses natural gas in the feed gas stream, which a POSA would have recognized as containing a hydrocarbon component.

331. Martin further discloses the inclusion of steam in the feed gas stream. Martin expressly notes that “[t]he hot product gas at 1050-1400 C (depending on the type of partial oxidation reactor used) drives the steam methane reforming reactions within the GHR tubes.” EX1006, p.12. Moreover, in my opinion, a POSA would have understood that a steam methane reforming process in a GHR reformer necessarily requires that steam be present in the input stream.

332. In my opinion, a POSA at the critical date would have recognized that the process of Martin necessarily includes steam as a component of the feed gas stream given the GHR and ATR processes disclosed in this reference. *See*, Section V., ¶¶28, 35–36 *supra*. In addition, Rafati provides a process where a “steam stream **319** passes through the heat exchanger **412** before combining with methane stream **303** for entry into the GHR **403**” as shown in annotated FIG. 2, below. EX1015, ¶[0128].

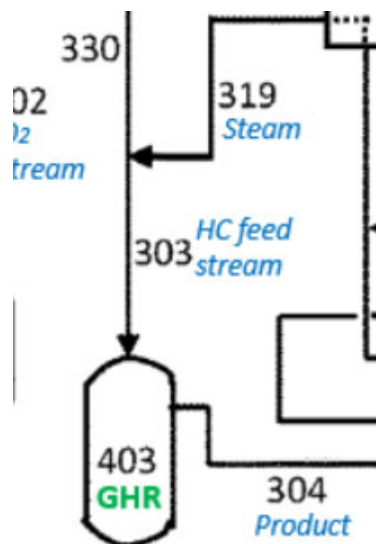


FIG. 2, excerpt, annotated

In my opinion, Rafati also describes a feed gas stream containing steam provided to an ATR for an autothermal reforming process, such as an ATR provided in a dual reformer configuration (e.g., parallel or series). (“Alternatively H_2+CO can be produced by the partial oxidation of a gaseous or liquid or solid hydrocarbon or carbonaceous fuel using pure oxygen (PDX) or from a catalytic auto-thermal reactor (ATR) using a gaseous or liquid hydrocarbon fuel with O_2 plus steam feed. In some preferred embodiments, the present disclosure further can relate to systems and methods for generation of H_2+CO in a PDX or ATR reactor followed by the use of a gas heated reformer (GHR) in either a series or parallel mode to the PDX or ATR reactor to produce additional H_2 and CO (i.e., synthesis gas) by utilizing the exhaust sensible heat in the PDX and/or ATR reactor system to provide the heat for endothermic catalytic steam plus hydrocarbon reforming reactions taking place in the GHR.” EX1015, ¶[0016].

(d) Element 1.1.2: methane

333. Martin discloses a feed gas stream with a hydrocarbon component containing methane. EX1006, p.12. Martin explicitly discloses natural gas in the feed gas stream, which necessarily contains methane.

334. In my opinion, a POSA as of the critical date would recognize that natural gas, as disclosed in the feed gas stream of Martin, necessarily includes methane as a primary component. Section V., ¶28. Rafati expressly recites the

hydrocarbon may be methane. EX1015, ¶[0124] (“The example embodiment of FIG. 2 is described in relation to the use of methane as the hydrocarbon.”).

(e) Element 1.2: endothermic reforming step

335. Martin discloses reforming at least a portion of the feed gas stream in an endothermic reforming step over a reforming catalyst thereby producing a first synthesis gas stream. EX1006, p.12. Martin describes that at least a portion of the natural gas is reformed in an endothermic reforming step and passed through a catalyst bed (in the gas heated reformer (GHR), *see* Figure 7). EX1006, p.12. Moreover, a POSA would have understood that a typical GHR process comprises an endothermic reforming step and a catalyst bed. *See*, Section V., ¶36.

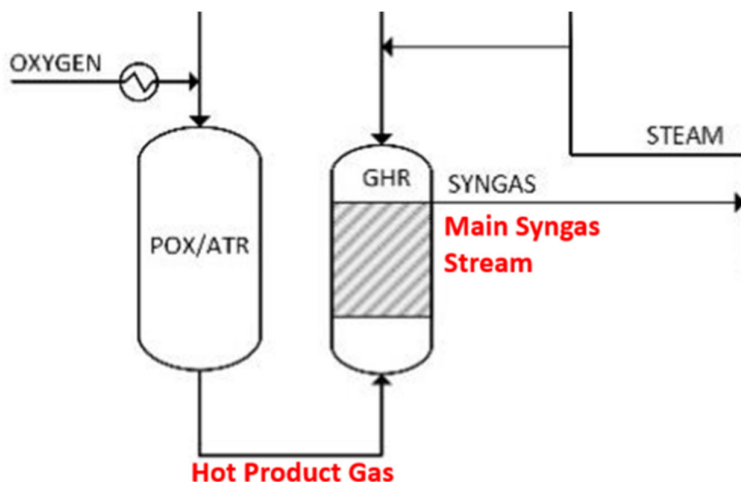


Figure 7, excerpt, annotated

(f) Element 1.2.1: first synthesis gas

336. Martin discloses at least inherently, a first synthesis gas stream comprising hydrogen, carbon monoxide, carbon dioxide, and unreacted methane

as these components are the natural result of the gas reforming process in Martin.

337. In my opinion, to the extent Martin may not explicitly disclose such a composition of a synthesis gas stream, Rafati discloses a process where a product stream having hydrogen and carbon monoxide exits the GHR. EX1015, ¶[0114] (“A favorable configuration uses a vertical gas heated reformer (GHR) with catalyst filled open ended tubes hanging from a single tube sheet at the top of the vessel, with the product H₂+CO leaving the reformer tubes and mixing with the product gas from a PDX reactor or an ATR in the base of the GHR, and the total product H₂+CO stream passing through the shell side and cooling typically from about 1050° C. to 550° C. to 800° C.”).

338. In my opinion, both Martin’s and Rafati’s GHRs would also naturally result in the production of unreacted methane and carbon dioxide. As the steam reforming reaction is in equilibrium, both methane (CH₄) and steam (H₂O) are present in the output of the reaction, as well as the H₂, CO and CO₂ product gases. See, Section V., ¶¶28-30, *supra*.

339. In my opinion, to the extent Martin and Rafati may not expressly disclose the composition of the first synthesis gas stream as including the gases enumerated in the claim, both references nevertheless disclose such composition of gases at least inherently. In my opinion, the output of the GHR disclosed in

Martin and Rafati would necessarily include hydrogen, carbon monoxide, carbon dioxide, and unreacted methane.

(g) *Element 1.3: reforming in an autothermal reforming step*

340. Martin discloses reforming a portion of the feed gas stream in an autothermal reforming step thereby producing a second synthesis gas stream, and combining the first synthesis gas stream and the second synthesis gas stream thereby producing a third synthesis gas stream. EX1006, p.12. Martin discloses an autothermal reforming step in, e.g., a POX/ATR reformer. EX1006, Figure 7. Martin shows the output of the POX/ATR—a second synthesis gas stream as a hot product gas—feeding the GHR in Figure 7. EX1006, p.12 (“(t)he hot product gas at 1050-1400 C (depending on the type of partial oxidation reactor used) drives the steam methane reforming reactions within the GHR tubes”). Martin then describes “eventually both syngas streams combines (sic, combine) to form the main syngas stream.” EX1006, p.12.

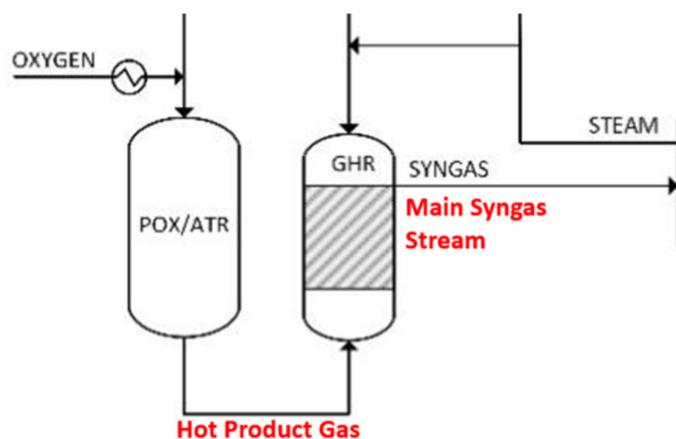


Figure 7, excerpt, annotated

(h) Element 1.3.1: exothermic partial oxidation and endothermic reforming

341. Martin discloses the autothermal reforming step comprises exothermic partial oxidation and endothermic reforming with steam over a reforming catalyst. EX1006, p.12.

342. Martin discloses the use of an ATR, which by its nature operates in this manner. EX1006, p.12 (“In the combined reforming concept, natural gas is partially oxidized in an oxygen deficient environment. In catalytic variation of partial oxidation, also known as auto-thermal reforming (ATR), the product gas from the partial oxidation section of the reactor is passed through a catalyst bed to enhance syngas generation reactions.”).

(i) Element 1.3.2: second and third synthesis gas

343. In my opinion, Martin discloses at least inherently, the second and third synthesis gas streams comprise hydrogen, carbon monoxide, carbon dioxide, and unreacted methane as these components are the natural result of the gas reforming process in Martin. *See*, Section V., ¶¶28–30, 34, *supra*.

344. In my opinion, to the extent Martin may not explicitly disclose such compositions of a second and third synthesis gas streams, Rafati expressly discloses reforming processes for producing synthesis gases comprising hydrogen, carbon monoxide, and unreacted methane. For example, Rafati provides “[t]he steam stream **319** fed to the GHR reactor **403** provides a steam to carbon ratio (carbon combined with hydrogen in the GHR reactor feed) of 6:1

in this case. This high ratio allows 80 bar H_2+CO production pressure with a low quantity of unconverted methane in the total product H_2+CO stream 304.”

EX1015, ¶[0129], FIG. 2 below. Thus, Rafati discloses that the product stream from the dual reforming reaction comprises at least hydrogen, carbon monoxide, and unreacted methane.

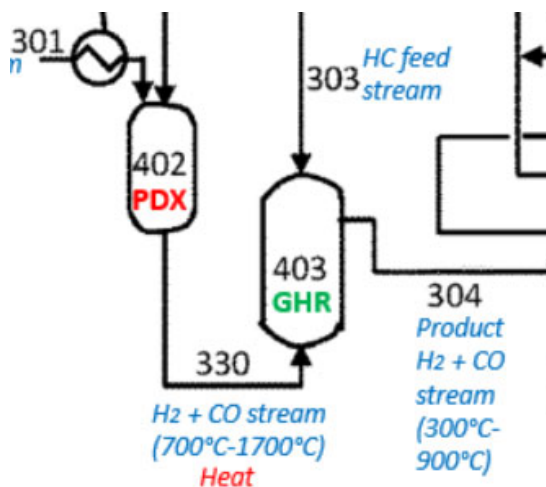


FIG. 2, excerpt, annotated

(j) Element 1.3.3: heat from autothermal reforming step

345. Martin discloses the heat generated by the autothermal reforming step is utilized for heating in the endothermic reforming step of step (b).

EX1006, p.12. Martin discloses heat (the hot product gas exiting the ATR) flows to the GHR to provide heat for the initial reaction. EX1006, p.12 (“The hot product gas at 1050-1400 C (depending on the type of partial oxidation reactor used) drives the steam methane reforming reactions within the GHR tubes.”).

(k) Element 1.4: converting the carbon monoxide

346. Martin discloses converting the carbon monoxide present in the third synthesis gas stream with steam thereby producing hydrogen and carbon dioxide thereby producing a fourth synthesis gas stream. EX1006, p.12. Martin describes using a “SYNGAS COOLING & SHIFT” (Figure 7) to convert carbon monoxide in the third gas stream using steam. EX1006, p.12. The result of this shift necessarily produces hydrogen and carbon dioxide in a fourth synthesis gas stream. See, Section V., ¶30, *supra*.

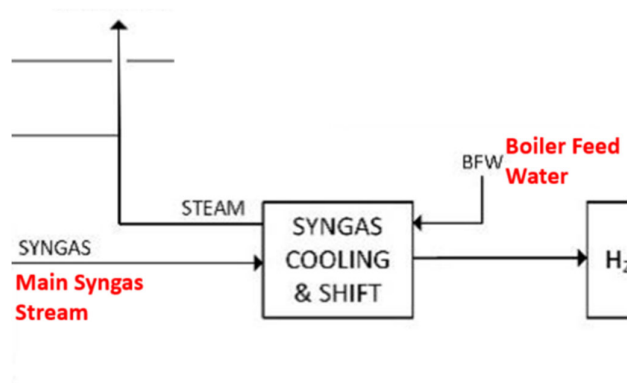


Figure 7, excerpt, annotated

347. In my opinion, to the extent Martin may not explicitly disclose that the CO conversion step necessarily produces H_2 and CO_2 , such products of a CO conversion step are expressly recited in Rafati. EX1015, ¶[0126] (“The product stream comprising H_2+CO is then reacted in at least one reactor to form a stream comprising H_2+CO_2 . As illustrated in FIG. 2, the total product $CO+H_2$ stream **305** passes through a first catalyst filled CO shift reactor **405** and a second catalyst filled CO shift reactor **406** in series with respective outlet streams **306** and **308**.”); FIG. 2.

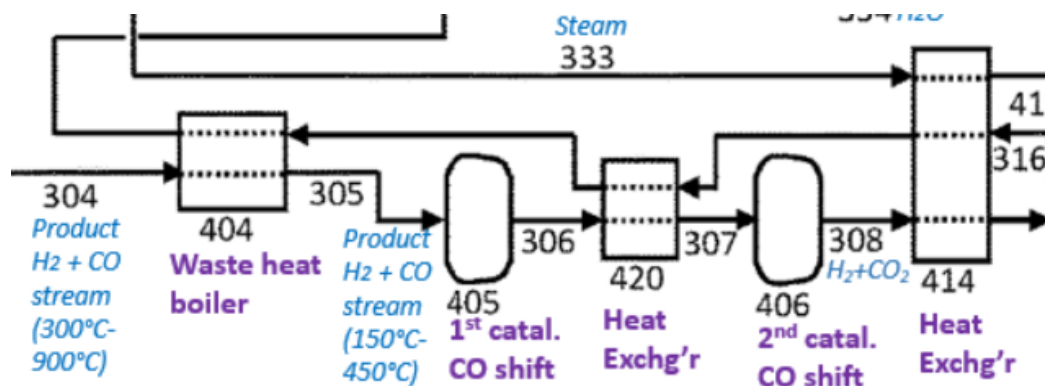


FIG. 2, excerpt, annotated excerpt

(l) *Element 1.4.1: fourth synthesis gas*

348. In my opinion, Martin discloses inherently the fourth synthesis gas stream comprises hydrogen, carbon dioxide, unreacted methane, and carbon monoxide unconverted in step (d). EX1006, p.12; *see*, Section V., ¶30, *supra*. Martin discloses “syngas shifting and cooling steps” that necessarily generates a shifted gas comprising hydrogen and carbon dioxide. *See*, Section XI.E.(k), *supra*. Unreacted methane and carbon monoxide unconverted in step (d) are also necessarily present in the shifted gas in the process of Martin. EX1006, p.12

349. In my opinion, to the extent Martin may not explicitly disclose such compositions of a fourth synthesis gas stream, Rafati expressly discloses that a H_2+CO stream which undergoes a CO shift reaction step results in a stream comprising H_2 and CO_2 , as well as CO and one or more carbon-containing materials. EX1015, ¶[0127] (“The stream **308** comprises H_2+CO_2 , but it is understood that any stream described herein as comprising H_2+CO_2

only defines the minimal composition of the stream, and further materials may be present in said stream, such as carbon monoxide and one or more carbon-containing materials.”).

350. In addition, Rafati discloses that the crude H_2+CO_2 stream (**331** of FIG. 2, below) EX1015, ¶[0127] “preferably can contain substantially all of the CO_2 derived from combustion of carbon in the hydrocarbon feed together with water vapor and minor amounts of CO , CH_4 , N_2 and Ar .” EX1015, ¶[0127]. Therefore, even if the composition of the fourth synthesis gas stream SG4 of the ’805 Patent is not considered inherent, Rafati expressly recites each of the components of the gas stream mixture.

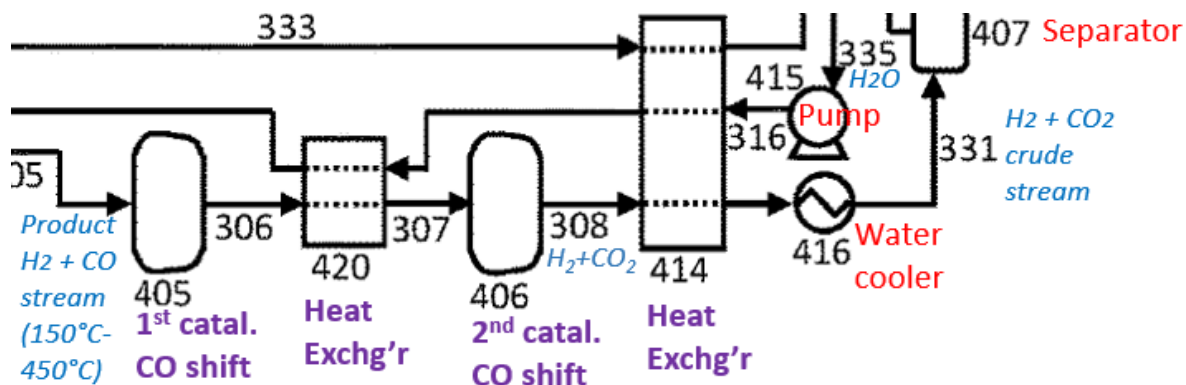


FIG. 2, excerpt, annotated

(m) Element 1.5: separating hydrogen

351. Martin discloses separating hydrogen from the fourth synthesis gas stream by pressure swing adsorption, thereby producing a first hydrogen-rich stream and a first residual gas stream. EX1006, p.12. Martin exemplifies this step schematically in, e.g., Figure 7, showing that hydrogen is separated from

the synthesis gas stream by “H₂ PSA,” producing a “PSA waste gas” as well as a hydrogen stream. EX1006, p.12, Figure 7.

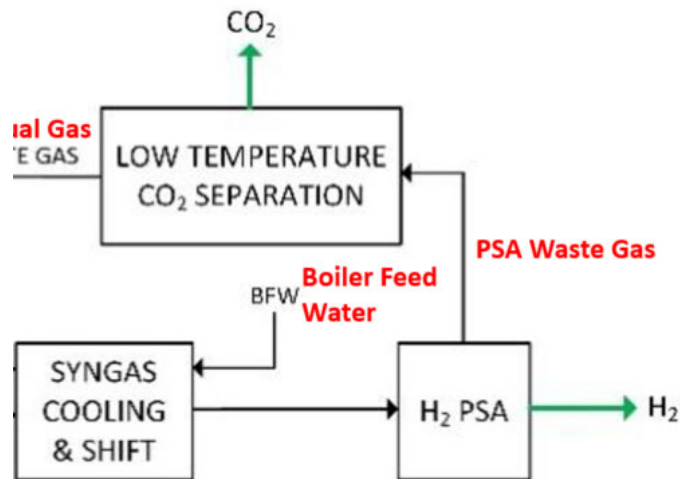


Figure 7, excerpt, annotated

(n) Element 1.5.1: first residual gas

352. Martin discloses the first residual gas stream comprises carbon dioxide, carbon monoxide unconverted in step (d), hydrogen not separated off in step (e), and unreacted methane. EX1006, p.12. Martin describes that, “[a]fter syngas shifting and cooling steps and hydrogen separation, the PSA waste gas contains about 76% CO₂ (mole basis) which is suitable for processing in a low temperature removal system.” EX1006, p.12. Further, in my opinion, carbon dioxide, carbon monoxide unconverted, hydrogen not separated, and unreacted methane, are the natural result of PSA separation of hydrogen in the process of Martin. *See*, Section V., ¶39, *supra*.

353. In my opinion, to the extent Martin may not explicitly disclose this element, Rafati discloses PSA for producing a substantially pure hydrogen

stream (310 of FIG. 2) and a waste gas (311 of FIG. 2), where the waste gas “preferably contains all the CO₂ plus CO, H₂, CH₄, Argon, N₂, and traces of water vapor previously in stream 309.” EX1015, ¶[0131].

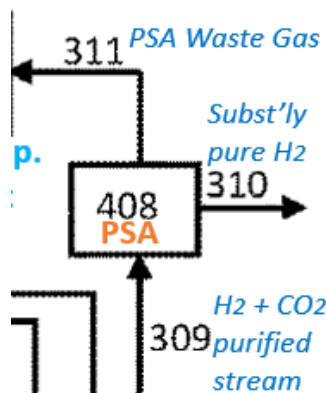


FIG. 2, excerpt, annotated

(o) Element 1.6: separating carbon dioxide

354. Martin discloses separating carbon dioxide from the first residual gas stream obtained in step (e) by cryogenic carbon dioxide separation, thereby producing a first carbon dioxide-rich stream and a second residual gas stream. EX1006, p.12. Martin discloses “[a]fter syngas shifting and cooling steps and hydrogen separation, the PSA waste gas contains about 76% CO₂ (mole basis) which is suitable for processing in a low temperature removal system. Such a system uses the auto-refrigeration principle to cool down the process stream near CO₂ triple point, and separates and purifies CO₂ product in a distillation column.” EX1006, p.12. In my opinion, a POSA would have understood this description to be a cryogenic carbon dioxide separation process. *See*, Section V., ¶40, *supra*. A result of the CO₂ separation is a CO₂-rich stream, as shown by

the green arrow directed outward from the low temperature CO₂ separation portion of Figure 7. EX1006, p.12. Martin also discloses that a second residual gas is produced from the cryogenic separation process. EX1006, p.12 (“WASTE GAS”).

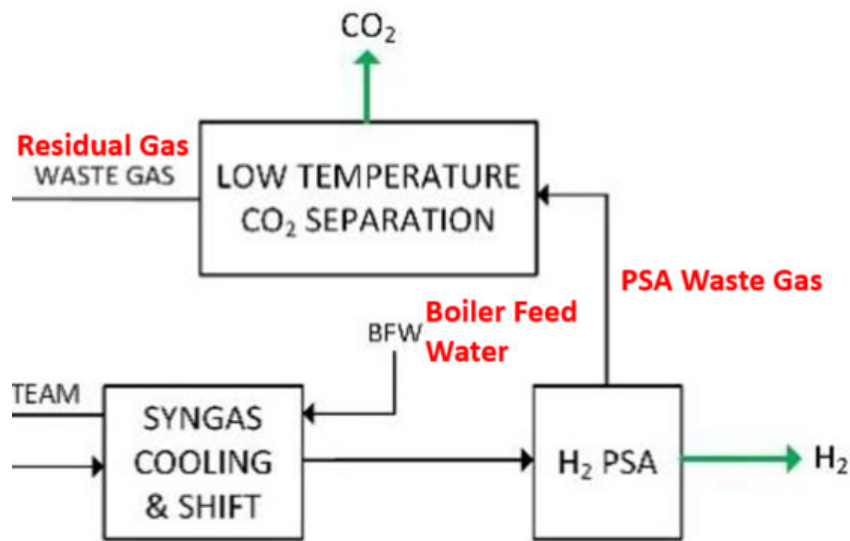


Figure 7, excerpt, annotated

355. In my opinion, to the extent Martin may not explicitly disclose this element, Rafati expressly discloses that a low temperature separation unit may be a cryogenic separation unit. EX1015, ¶[0131] (“The waste gas stream **311** is then processed in a low temperature separation unit **409** (e.g., a cryogenic separation unit) as otherwise described herein to form a liquid CO₂ product stream.”).

(p) Element 1.6.1: second residual gas

356. In my opinion, Martin discloses at least inherently the second residual gas stream comprises carbon monoxide unconverted in step (d),

hydrogen not separated off in step (e), carbon dioxide not separated off in step (f), and unreacted methane, as these gases are the natural result of cryogenic CO₂ separation in the process of Martin. *See*, Section V., ¶40, *supra*.

357. In my opinion, to the extent Martin may not explicitly disclose this element, Rafati discloses a process comprising a cryogenic CO₂ separation unit for producing a liquid CO₂ product stream (312 of FIG. 2) and a vapor phase stream (313 of FIG. 2) comprising the CO₂ remaining after cryogenic CO₂ separation as well as the other components of the PSA waste gas, including CO, H₂, CH₄, Argon, N₂, and traces of water vapor. EX1015, ¶[0131] (describing that the cryogenic separation step is “preferably carried out such that at least 50 mol % of the CO₂ in the waste gas stream 311 is separated into the liquid CO₂ product stream 312. The remaining vapor phase materials exit the low temperature separation unit 409 in vapor phase stream 313.”) See annotated excerpt of Fig. 2 below.

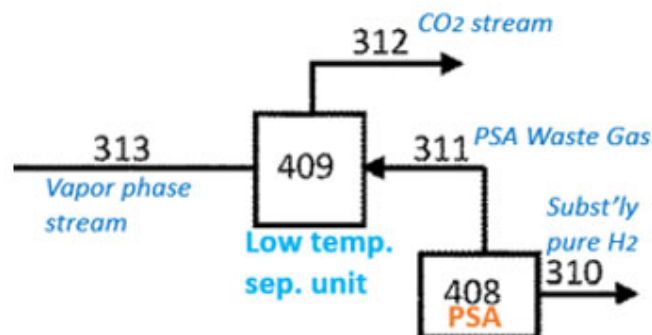


FIG. 2 (annotated excerpt)

(q) Element 6: first carbon dioxide-rich stream is subjected to a thermal separation process for separation of methane from carbon dioxide

358. In my opinion, claim 6 recites nothing more than applying methods known in the prior art. *See*, Section V., ¶41, *supra*. Martin, alone or in combination with Rafati, discloses all elements of claim 6.

359. As discussed in the description of the '805 Patent, distillation is described as an example of a thermal process that may be used to separate methane from carbon dioxide. *See*, Section IX., ¶91, *supra*. Martin discloses a process using “LOW TEMPERATURE CO₂ SEPARATION”, which is described as applying “the auto-refrigeration principle to cool down the process stream near CO₂ triple point, and separates and purifies CO₂ product in a distillation column.” EX1006, p.12.

360. In my opinion, to the extent Martin may not explicitly disclose this element, Rafati discloses, “[o]ptionally the liquid CO₂ can be treated in a stripping distillation column to remove dissolved H₂+CO+CH₄ which will be transferred to the vapor phase.” EX1015, ¶[0022].

(r) Element 11: compression step and cooling step

361. I emphasize that cryogenic CO₂ separation was known in the art long before the critical date to include compression and cooling steps. *See*, Section V., ¶40, *supra*; EX1018, EX1020; EX1021. In my opinion, claim 11 recites nothing more than applying methods known in the prior art to be

included in cryogenic CO₂ separation. Martin, alone or in combination with Rafati, discloses all elements of claim 11.

362. Martin discloses a process using “LOW TEMPERATURE CO₂ SEPARATION”, which is described as applying “the auto-refrigeration principle to cool down the process stream near CO₂ triple point, and separates and purifies CO₂ product in a distillation column.” EX1006, p.12. As understood by a POSA, cooling close to the CO₂ triple point necessarily results in condensation of CO₂ in a manner which necessarily produces a CO₂ rich stream at least partly in the form of a condensed CO₂ stream.

363. In my opinion, a POSA at the critical date would have understood that the cryogenic CO₂ separation process of Martin necessarily includes the at least one compression step and at least one cooling step of claim 11 the '805 patent. In addition, the terms “LOW TEMPERATURE CO₂ SEPARATION” and “low temperature CO₂ removal system”, as used in Martin, was readily understood by a POSA as examples of a process including cooling and condensation steps of a gas stream containing CO₂ in a manner necessarily producing a CO₂ rich stream and residual gas stream. In addition, Rafati expressly discloses that condensation and cooling steps, as recited in claim 11, may be steps in cryogenic CO₂ separation. EX1016, ¶[0094] (“The forming of the liquid CO₂ product stream in the cryogenic separation unit can comprise ... cooling the waste gas stream comprising CO₂ in at least one heat exchanger to

provide the waste gas stream comprising CO₂ as a two phase stream”), ¶[0086] (“The cryogenic separation unit further can comprise: a compressor configured for compressing the waste gas stream comprising CO₂”). Based on the teachings in Martin and Rafati and the general skill and knowledge in the art, a POSA would have understood to implement cryogenic CO₂ separation following PSA for hydrogen separation using at least one compression step and at least one cooling step to produce a carbon dioxide-rich stream at least partly in the form of a condensed carbon dioxide stream,

(s) *Element 12: a plant configured for performance of the process*

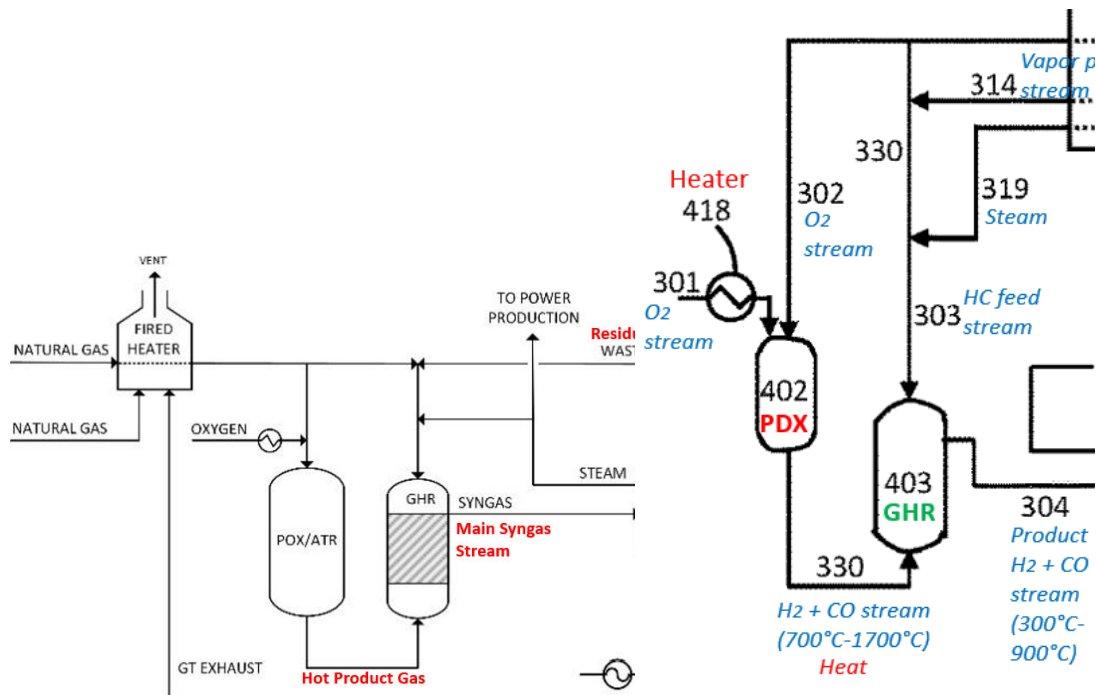
364. Element 12 recites “a plant configured for performance of the process according to claim 1.”

365. In my opinion, the combination of Martin and Rafati discloses all the elements of the process of claim 1. *See*, Section XI.C.2.(a)-(p)., *supra*. Further, each of Martin and Rafati disclose a plant configured for performance of the hydrogen separation process. EX1006, Figure 7; EX1015, ¶[0113] (“the present disclosure may refer to a hydrogen plant, and it is understood that such hydrogen plant refers to the combination of elements necessary to form the hydrogen production system utilized herein.”). In my opinion, the plant of claim 12 is defined by the process of claim 1. No additional elements of the plant are provided. In my opinion, therefore, the combination of Martin and Rafati teaches each and every element of claim 12 as recited in the ’805 Patent.

3. Martin and Rafati – Reasonable Expectation of Success

366. In my opinion, a POSA combining the teachings of Martin and Rafati would have expected to successfully practice the method of claims 1, 6, 11 and 12. Martin discloses all the process steps, in the same sequence, as the process of claim 1 of the '805 Patent. However, it may be argued that the teachings of Martin are described at a high level, and a POSA could not rely on the teachings of Martin alone to arrive at the claimed invention. Rafati, which addresses the same technical problem of producing hydrogen and separating carbon dioxide as Martin and claim 1, provides more detailed descriptions that clarify Martin's teachings. In my opinion, therefore, a POSA reviewing both references would have recognized that the details of Rafati could be integrated into the framework of Martin to yield predictable results.

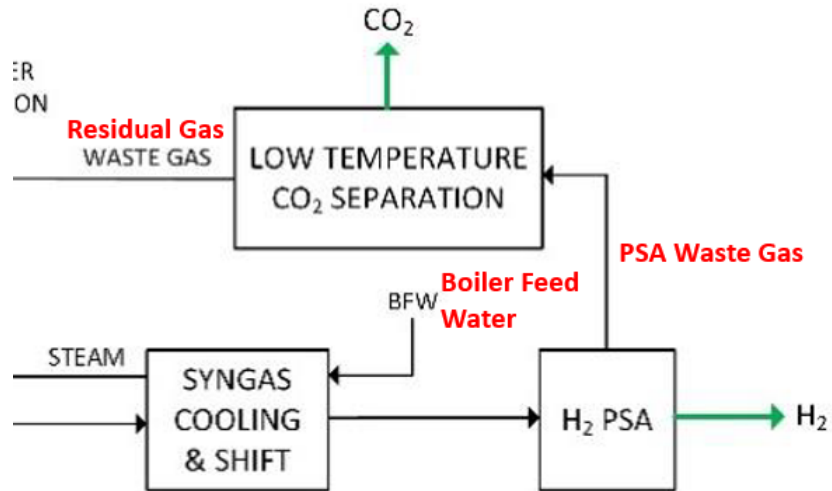
367. In my opinion, each of the individual steps recited in Martin and Rafati could be combined with no material change in their respective functions. For example, the parallel configuration of the reforming systems of Martin and Rafati both function to enhance the generation of syngas with the goal of improving efficiency by using the heat from the PDX/ATR process to drive the endothermic reforming reactions of the GHR, as shown below.



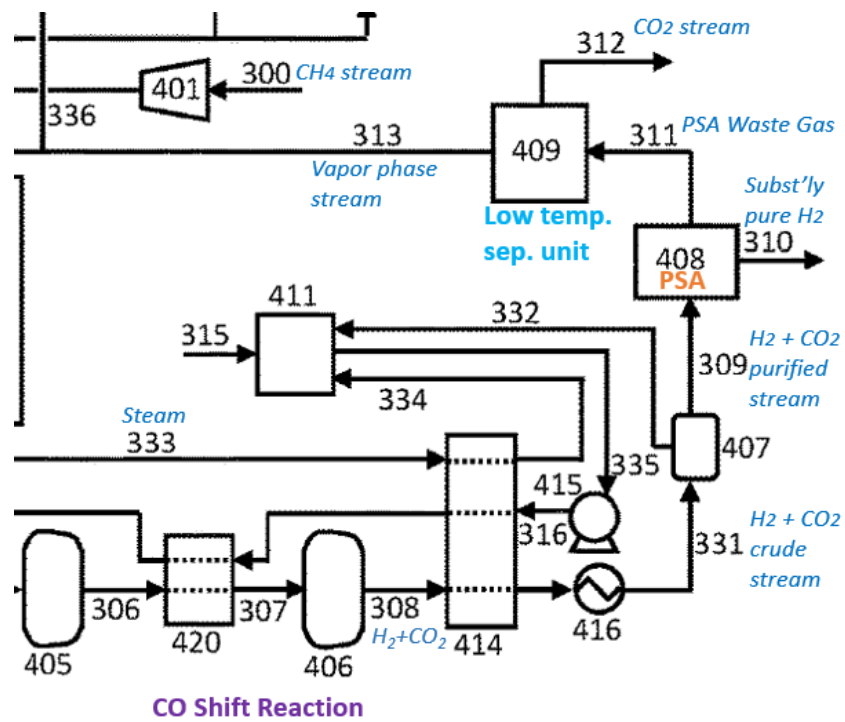
Martin, Figure 7, excerpt annotated

Rafati, FIG. 2, excerpt, annotated

368. In addition, both Martin and Rafati describe the same combination of devices, in the same order, for downstream CO conversion (CO Shift Reactor) as well as H₂/CO₂ separation (H₂ PSA unit positioned upstream of a low temperature CO₂ separation unit), as shown below. Therefore, a POSA would have a reasonable expectation of combining the teaching of Martin and Rafati to predictably achieve an operable hydrogen production process using a parallel dual reformer configuration and cryogenic carbon dioxide separation.



EX1006, Figure 7, excerpt, annotated



EX1015, FIG. 2, excerpt, annotated

F. Petition II Ground 2: Claims 2-4 and 6 are Obvious over Martin in Combination with Rafati and in Further View of Gauthier

369. It is my opinion that claims 2-4 and 6 would have been obvious to a POSA based on the combination of Martin (EX1006), with Rafati (EX1015) and in further view of Gauthier (EX1014).

1. Motivation to Combine, Reasonable Expectation of Success

370. In my opinion, the combination of Martin and Rafati in view of Gauthier teaches each and every limitation of claims 2-4 and 6 of the '805 Patent. The motivation to combine the teachings of Martin and Rafati in the manner claimed is provided above. *See*, Sections XI.E.1, XI.E.3, *supra*. Martin and Rafati share a common, recognized goal of efficiently producing hydrogen and separating CO₂ coproduct while reducing CO₂ emissions. Martin and Rafati both achieve this goal employing a similar solution by producing hydrogen through reforming of hydrocarbons, including natural gas/methane, and shifting (water-gas shift) the synthesis gas to increase hydrogen in the synthesis gas, followed by separation of hydrogen from the shifted synthesis gas using a PSA and separation of CO₂ through cryogenic CO₂ separation.

371. Similarly, Gauthier relates to a process for combined production of hydrogen and carbon dioxide, again, from synthesis gas generated through reforming of hydrocarbons, including natural gas/methane and shifting the synthesis gas to increase hydrogen in the synthesis gas. *See*, Section IX.F.,

¶¶89-92, *supra*; Ex1014, ¶¶[0001], [0026], [0142]. Gauthier's goal is an "economically viable method for producing hydrogen" and for "producing carbon dioxide, while ensuring the utilization of the combustible gases included in the offgas issuing from the PSA". EX1014, ¶[0017]. Gauthier's method is particularly advantageous to "optimize hydrogen production and to jointly produce food grade carbon dioxide". EX1014, ¶[0047]. The method involves capturing and purifying CO₂ to provide a food grade product. EX1014, ¶[0014]. A POSA would have recognized that such CO₂ capturing steps decrease the emission of CO₂ from the process. Gauthier, like Martin and Rafati, uses a PSA process **14** to separate a purified hydrogen-rich product stream (H₂ arrow) and a residual gas stream **15** followed by cryogenic CO₂ separation of **15** to produce a CO₂-enriched product stream **23**. EX1014, Figures 1 and 2, ¶¶[0143]-[0146]. Gauthier, however, further teaches additional steps of generating an additional hydrogen-rich gas stream **31**, particularly using a permeation module **29** (hydrogen membrane), recycling of this hydrogen-rich product stream back to the PSA **14** and/or employing this hydrogen rich product stream as fuel for heating in a reformer. EX1014, ¶[0151]. Gauthier additionally teaches further purification of the CO₂-enriched product stream **23** by distillation to remove light impurities in a distillation step separate from the cryogenic CO₂ separation and providing food grade CO₂. EX1014, ¶[0147]. Gauthier also includes

additional steps recycling residual/waste gases for further capture of H₂ or CO₂ and for use as feed and/or fuel in other parts of the process system.

372. In my opinion, a POSA would have been motivated to combine the teachings of Gauthier with those of Martin and Rafati because each of the processes are directed to hydrogen production with low CO₂ emissions and each of the processes use PSA hydrogen separation and cryogenic CO₂ separation. Gauthier, Martin, and Rafati each address the same or similar problems recognized in the art prior to the critical date, share common goals and teach the same or similar solutions to the problems. In Gauthier, separated CO₂ is purified and captured as a value-added product of the process (food grade CO₂). The shared goal of Martin and Rafati to produce hydrogen and lower emissions of CO₂ would be viewed by a POSA as overlapping with those of Gauthier. Thus, in my opinion, a POSA would have been motivated to combine the teachings of Gauthier with those of Martin and Rafati to increase the purity and value of products of the combined process and to increase at least the energy efficiency of the combined process by recycling of product gases to feed and/or fuel the reformers in the process.

373. In my opinion, modifications of the combination of Martin and Rafati with the additional purification and recycling steps as taught in Gauthier would not change the principles of operation of the combined process of Martin and Rafati. Because the processes employ the same feed gases in the same

process steps (reforming, water-gas shift, PSA separation and cryogenic CO₂ separation), generate the same intermediates (syngas and shifted syngas) and the same products (H₂ and CO₂), a POSA would have recognized that modification using Gauthier's additional methods would not change the principles of operation of a Martin/Rafati process and would further result in recognized benefits, such as a higher degree of energy efficiency and higher value products.

374. The combination of the additional process steps of Gauthier with those of Martin and Rafati were well within the ordinary skill in the art of a POSA at the critical date and represent the combination of known methods and process equipment to provide their intended function. The additional process steps of Gauthier are compatible with the processes and process conditions in Martin and Rafati, such as temperature, pressure and flow rate. Additionally, because the additional steps of Gauthier were well known in the art and their function and implementation were also well known and understood in the art, a POSA would have had a reasonable expectation that the additional process steps of Gauthier could be successfully implemented in the combined process of Martin/Rafati and that the desired additional benefits of their implementation would be achieved.

375. Specifically with respect to claim 2, in my opinion, a POSA would have been motivated to apply the additional hydrogen separation step as taught by Gauthier to the combined method of Martin/Rafati to increase the yield of

the desired hydrogen product by separating an additional hydrogen-rich stream and would have known in view of what was known in the art and the teachings of these references how to implement that separation and would have reasonably expected success in that implementation.

376. Specifically with respect to claim 3, in my opinion, a POSA would have been motivated to employ the specific membrane hydrogen separation step as taught by Gauthier with the combined method of Martin/Rafati, again, to increase the yield of the desired hydrogen product by separating an additional hydrogen-rich stream. The use of a hydrogen selective membrane to separate hydrogen from synthesis gas product streams was well known in the art at least by the critical date. A POSA would have known how to implement the membrane separation at least in view of what was known in the art and the teachings of the references and would have reasonably expected success in that implementation.

377. Specifically with respect to claim 4, in my opinion, Gauthier teaches separation of second hydrogen-rich stream using membrane separation and recycling of that stream to a pressure swing adsorption unit. Hydrogen separation/purification by use of membranes and pressure swing absorption processes was well known in the art at least at the critical date. A POSA would have been motivated to employ the hydrogen recycling taught by Gauthier with the combined method of Martin/Rafati to increase the yield of the desired

hydrogen product, but also to generally increase the efficiency of the entire hydrogen generation process. A POSA would have known how to implement the hydrogen recycling at least in view of what was known in the art and the teachings of the references and would have reasonably expected success in that implementation.

378. Specifically with respect to claim 6, in my opinion Gauthier teaches the further purification of CO₂ that is cryogenically separated from synthesis gas to provide higher value CO₂ product (food grade CO₂). The further purification is removal of light impurities in the CO₂ by distillation. As described in the '805 Patent, distillation is a thermal separation process for separation of methane. A POSA would have been motivated to employ the additional purification of CO₂ taught by Gauthier to the combined method of Martin/Rafati to increase the yield and value of the CO₂ product of the process. Increasing the value of the CO₂ product would generally increase the cost effectiveness of the combined process. More specifically, a POSA would have known that cryogenically separated CO₂ as produced in the Martin/Rafati process would contain unreacted or residual methane as a light impurity in view of the feed employed and the processes employed to generate and shift synthesis gas. A POSA would also have known that the well-known distillation method described by Gauthier could be employed to remove methane from CO₂, would have known how to implement that distillation method at least in view of what

was known in the art and the teachings of the references and would have reasonably expected success in implementation of the distillation to remove methane from CO₂ and generate a value added product.

2. Martin and Rafati in View of Gauthier Disclose the Limitations of Claims 2-4 and 6.

(a) Claim 2: hydrogen is separated from the second residual gas stream (RG2), thereby producing a second hydrogen-rich stream (HG2) and a third residual gas stream (RG3).

379. In my opinion, as discussed in section X1.E.2. *supra*, the combination of Martin and Rafati teaches each and every element of claim 1 from which claim 2 depends. *See*, Sections XI.E.2(a)–(p), *supra*.

380. In the '805 Patent RG2 is residual gas exiting the cryogenic CO₂ separation unit **205** and is described as being introduced into a membrane unit **206** to separate H₂ and generate HG2 enriched in hydrogen and a third residual gas Stream (RG3). EX1001, col.5, ln.53–63.

381. Both Martin and Rafati generate hydrogen-containing residual gas (from a cryogenic CO₂ separation unit) “WASTE GAS” (EX1006, Figure 7) and vapor phase stream **313** (EX1015, FIG. 2). “WASTE GAS” (Figure 7) of Martin is recycled to the reforming reactors to maximize the process efficiency and CO₂ capture. EX1006, p.12. In Rafati stream **313** can be recycled for a variety of uses, and specifically to hydrocarbon feed streams **303**. EX1015, ¶[0132]. Stream **313** derives from cryogenic CO₂ removal from stream **311** and contains CO₂ plus CO, H₂, CH₄, Argon, N, and traces of water. EX1015,

¶[0131]. “WASTE GAS” (Figure 7) is not expressly described in Martin as containing hydrogen, but in my opinion, in view of the feed gas and process steps employed, the similarities in the Martin and Rafati processes, and the fact that “WASTE GAS” and stream **313** both are residual gas from cryogenic CO₂ separation, a POSA would have recognized that the “WASTE GAS” of Martin Figure 7 would contain hydrogen.

382. Gauthier in Figure 1 describes a gas mixture **24** from a cryogenic CO₂ separation unit **22** which contains hydrogen. This purge gas is introduced to permeation module **29** to generate a permeate **31** enriched in hydrogen and a purge waste **30** enriched with methane and CO. EX1014, ¶[0150]. First, I note that based on the description in Gauthier, the permeation module is a hydrogen selective separation membrane. I further note that purge waste **30** is enriched in methane and CO, as described by Gauthier, but is also depleted in H₂. Purge waste **30** is residual gas from the H₂ membrane separation and is depleted in H₂ because hydrogen largely passes through the membrane to permeate **31**.

383. In my opinion, WASTE GAS/stream **313**/purge gas **24** of the combination of Martin/Rafati and Gauthier is the same as the second residual gas stream RG**2** of the '805 Patent. In my opinion, permeate **31** of Gauthier is the same as the second hydrogen-rich stream HG**2** of the '805 Patent. In my opinion, purge gas **30** is the same as the third residual gas stream RG**3** of the '805 Patent. Thus, the combination of Martin and Rafati in view of Gauthier

teaches all of the elements of claim 2, that is, it teaches producing a second hydrogen-rich stream and a third residual gas stream as claimed in claim 2.

(b) Claim 3: hydrogen is separated from the second residual gas stream (RG2) by membrane separation

384. In my opinion, the combination of Martin and Rafati in view of Gauthier teaches all of the elements of claim 2 from which claim 3 depends. See, Section XI.F.2.(a), *supra*.

385. In Patent '805, RG2 exits cryogenic CO₂ separation unit **205** and is described as being introduced into a membrane unit **206** to generate HG2 and RG3. EX1001, col.10, ln.26–38.

386. Gauthier in Figure 1 describes a gas mixture **24** from a cryogenic CO₂ separation unit **22** which contains hydrogen. In my opinion, WASTE GAS/stream **313**/purge gas **24** of the combination of Martin/Rafati and Gauthier is the same as the second residual gas stream RG2 of the '805 Patent. This purge/waste gas is introduced to permeation module **29** to generate a permeate **31** enriched in hydrogen and a purge waste **30** enriched with methane and CO. EX1014, ¶[1050]. Such a permeation module for generating a hydrogen-enriched stream and a purge gas enriched with methane and CO as described in Gauthier is therefore a membrane for hydrogen separation. Thus, in my opinion, the permeation module **29** is equivalent to the membrane **206** of the '805 Patent. Thus, the combination of Martin and Rafati in view of Gauthier

teaches producing a second hydrogen-rich stream and a third residual gas stream using membrane separation as claimed in claim 3.

(c) ***Claim 4: the second hydrogen-rich stream is supplied to the fourth synthesis gas stream for separation of hydrogen by pressure swing adsorption in step (e).***

387. In my opinion, the combination of Martin and Rafati in view of Gauthier teaches each and every element of claim 2 from which claim 4 depends. *See*, Section XI.F.2(a), *supra*.

388. In the '805 patent, HG2 exits membrane unit **206** (Figures 2 and 3) and is optionally recycled to the synthesis gas stream SG4 which enters PSA unit **204** to increase overall hydrogen yield. EX1001, col.12, ln.29–31.

389. Gauthier in Figure 1 describes a purge gas **24** from a cryogenic CO₂ separation unit **22** which contains hydrogen. In my opinion, WASTE GAS/stream **313**/purge gas **24** of the combination of Martin/Rafati and Gauthier is the same as the second residual gas stream SG2 of the '805 Patent. *See*, Sections XI.F.2(a)–(b), *supra*. This purge/waste gas is introduced to permeation module **29** to generate a permeate **31** enriched in hydrogen. *See*, Section XI.F.2(b), *supra*. The hydrogen-rich permeate **31** is recycled to the synthesis gas stream **13** which enters the hydrogen purification unit **14** which is referred to as a PSA type unit. EX1014, Figure 1, ¶¶[0151], [0143]. Therefore, the hydrogen-rich permeate **31** is equivalent to the second hydrogen-rich stream HG2 of claim 4 of the '805 Patent, stream **13** is equivalent to the fourth synthesis gas stream SG4 of claim 4 of the '805 Patent and hydrogen

purification unit **14** is equivalent to the PSA **204** of the '805 Patent. Thus, the combination of Martin and Rafati in view of Gauthier teaches supplying the second hydrogen-rich stream to the fourth synthesis gas stream for separation of hydrogen by pressure swing adsorption in step (e) as claimed in claim 4.

(d) Claim 6: the first carbon dioxide-rich stream comprises unreacted methane, and the first carbon dioxide-rich stream is subjected to a thermal separation process for separation of methane, thereby producing a second carbon dioxide-rich stream.

390. In my opinion, the combination of Martin and Rafati teaches each and every element of claim 1 from which claim 6 depends. *See*, Sections XI.E.2(a)–(p), *supra*.

391. The '805 patent states that in a preferred embodiment of the process the carbon dioxide-rich stream CG1 comprises unreacted methane, and the carbon dioxide-rich stream CG1 is subjected to a thermal separation process for separation of methane, giving a second carbon dioxide rich stream CG2 from which methane is removed. EX1001, col.6, ln.19–24 (no figure is provided in the '805 Patent showing the process configuration with the second carbon dioxide rich stream CG2). The patent states that CG1 contains significant residual amounts of methane that can optionally be removed by distillation of the carbon dioxide-rich stream CG1 (not shown in any figure). EX1001, col.12, ln.11–14. Further, the '805 Patent also states that the thermal separation process is preferably a distillation. EX1001, col.6, ln.30–31. Thus, in the '805 Patent

distillation of methane from CG1 is a thermal process for separation of methane from CG1 to generate a second carbon dioxide stream CG2.

392. In Gauthier, Figure 1, stream **23** which exits the cryogenic CO₂ separation unit **22** is described as a condensate essentially containing CO₂. EX1014, ¶[0146]. In Martin, the CO₂ stream exiting cryogenic separation is shown as a green arrow in Figure 7, and in Rafati it is shown as stream **312** in FIG. 2. CO₂-containing stream **23** of Gauthier is purified by distillation in order to strip it of the lighter impurities (e.g., CH₄) entrained in the liquid phase. EX1014, ¶[0147]. In my opinion, in view of the feed streams and processes of Martin, Rafati and Gauthier, the CO₂ separated by cryogenic separation in these processes (e.g., condensate **23**) will contain some residual level of methane. I also note that methane, when present in CO₂, would be understood by a POSA to be a lighter boiling component (i.e., lower molecular mass) than CO₂. The condensate **23** is expanded to 23 bar before being fed to the distillation column **25** and a liquid phase **26** consisting of food grade CO₂ is recovered at the bottom of the column. EX1014, ¶[0147]. In my opinion, in view of the similarity of feed gas, process steps and purifications steps in the '805 Patent and Martin, Rafati and Gauthier, a POSA would have recognized that the stream exiting cryogenic separation unit (e.g., stream **23**) would contain methane and would have recognized that methane is a light impurity, when present in a CO₂ stream, that can be removed by distillation as described in Gauthier.

393. Thus, stream **23** of Gauthier is the same as the first carbon dioxide-rich stream **CG1** comprising unreacted methane as claimed in claim 6 of the '805 Patent, and stream **26** of Gauthier is the same as the second carbon dioxide stream of increased purity as claimed in the '805 Patent. The distillation column **25** of Gauthier performs a distillation to remove light impurities (e.g., methane) from CO₂. EX1014, ¶[0147]. Distillation is exemplified in the '805 Patent as “The thermal separation process is preferably a distillation.” EX1001, col.6, ln.30–31. The distillation of Gauthier (implemented in column **25**) is thus a thermal separation process for separation of methane. Thus, the combination of Martin and Rafati in view of Gauthier teaches subjecting a first CO₂-rich stream comprising unreacted methane to a thermal separation process for separation of methane to produce a second CO₂-rich stream as claimed in claim 6 of the '805 Patent.

G. Petition II Ground 3: Claims 2-5 are Obvious over Martin in Combination with Rafati and in Further View of Terrien

394. It is my opinion that claims 2-5 would have been obvious to a POSA based on the combination of Martin (EX1006), with Rafati, (EX1015), and in further view of Terrien (EX1024).

1. Motivation to Combine, Reasonable Expectation of Success

395. In my opinion, the combination of Martin, Rafati and Terrien teaches or suggests each and every element of claims 2-5. In my opinion,

motivation to combine the teachings of Martin and Rafati in the manner claimed is provided above. *See*, Section XI.E.1, *supra*.

396. Martin and Rafati share a common, recognized goal of efficiently producing hydrogen and separating CO₂ coproduct while reducing CO₂ emissions. Martin and Rafati both achieve this goal employing a similar, if not the same, solution by producing hydrogen through reforming of hydrocarbons, including natural gas/methane, and shifting (water-gas shift) the synthesis gas to increase hydrogen in the synthesis gas, followed by separation of hydrogen from the shifted synthesis gas using a PSA and separation of CO₂ through cryogenic CO₂ separation.

397. Similarly, Terrien relates to a process for more efficient recovery of hydrogen and carbon dioxide, which as in Martin and Rafati, is applied and exemplified by recovery of hydrogen and carbon dioxide from synthesis gas generated through reforming of hydrocarbons and shifting the synthesis gas to increase hydrogen in the synthesis gas. EX1024, ¶¶[0016], [0019]. With respect to application to hydrogen plants, Terrien's goals include recovery of additional hydrogen and carbon dioxide to provide "significant savings with regards to the size of reformer, natural gas consumption, carbon dioxide tax, etc." EX1024, ¶[0006]. The goals of Terrien are, thus, the same or similar to those of Martin and Rafati for increased efficiency of hydrogen production and decreased CO₂ emissions to, for example, save on carbon tax. Terrien, like Martin and Rafati,

employs a PSA process to separate a first purified hydrogen-rich product stream followed by cryogenic CO₂ separation to produce a CO₂-enriched product stream. Terrien FIG. 5 provides an exemplary process for improved recovery of hydrogen and carbon dioxide from a process stream **1** obtained from a process unit **0**. EX1024, ¶¶[0077]–[0078]. The process unit **0** is any unit which includes a purification step that results in a hydrogen rich fraction, e.g., stream **27** (EX1024, ¶[0018]) and is preferably a PSA unit that is used to recover and purify hydrogen. EX1024, ¶[0020]. Terrien describes that increased recovery of hydrogen and carbon dioxide is achieved by balancing the recycle of the hydrogen-rich permeate from hydrogen membrane separation units. EX1024, Abstract, ¶[0008]. Terrien specifically teaches steps of generating hydrogen-rich gas streams **5** and **12** using hydrogen selective membranes (**4** and **11**). EX1024, FIG.5, FIG. 6; ¶¶[0025], [0041]. This is in addition to hydrogen-rich stream **27** which is separated in unit **0** for example using a PSA. I note that hydrogen-rich gas stream **12** is formed by passing carbon dioxide lean non-condensable stream **10** through hydrogen selective membrane **11**. EX1024, ¶[0077]. Terrien further discloses using the hydrogen-rich gas stream (**27**, **5** and/or **12**) as feed into a PSA unit or as fuel for heating in a reformer. EX1024, ¶¶[0046], [0078], [0068], [0085]. In my opinion, a POSA would have been motivated to combine the teachings of Terrien with those of Martin/Rafati because each of the processes are directed to hydrogen production with low CO₂ emissions or

improved capture of CO₂ and each of the processes use PSA hydrogen separation and cryogenic CO₂ separation for H₂ and CO₂ separation/capture. The references address the same or similar problems, share common goals and teach the same or similar solutions to the problems. In Terrien, additional steps of hydrogen separation using hydrogen selective membranes are employed and hydrogen-rich gas streams are recycled to feed a PSA unit to enhance the effectiveness of hydrogen capture (i.e., recycled to the process unit 9). EX1024, ¶[0045]. Terrien also describes recycling of hydrogen-rich gas streams as fuel to a reforming unit to boost the percentage of carbon dioxide capture. EX1024, ¶[0078]. The goals of Martin and Rafati to produce hydrogen and lower emissions of CO₂ overlap with those of Terrien to increase recovery of hydrogen and carbon dioxide. In my opinion, a POSA would have been further motivated to combine the teachings of Terrien with those of Martin and Rafati to increase the efficiency of recovery of desirable products in the combined process and to increase at least the energy efficiency of the combined process by recycling of product gases to feed and/or fuel the reformers in the process.

398. In my opinion, modifications of the combined process of Martin and Rafati with the additional purification and recycling steps as taught by Terrien would not change the principles of operation of the combined process of Martin and Rafati. Because the processes employ the same feed gases in the same process steps (reforming, water-gas shift, PSA hydrogen separation and

cryogenic CO₂ separation), generate the same intermediates (syngas and shifted syngas) and the same products (H₂ and CO₂), a POSA would have recognized that modification using Terrien's additional methods would not change the principles of operation of a Martin/Rafati process and would further result in recognized benefits, such as enhanced recovery of hydrogen and carbon dioxide which at least improves process efficiency and economics.

399. In my opinion, the combination of the additional process steps of Terrien with those of Martin and Rafati was well within the ordinary skill of a POSA and represent the combination of known methods and process equipment to provide their known and intended function. The additional process steps of Terrien are compatible with the processes and process conditions in Martin and Rafati, such as conditions of temperature, pressure and flow rate. Because the additional steps of Terrien were well known in the art at the critical date and their function and implementation were also then well-known and understood, in my opinion, a POSA would have had a reasonable expectation that the additional process steps of Terrien could be successfully implemented in the combined process of Martin/Rafati and that the desired additional benefits of their implementation as taught by Terrien would be achieved.

400. With respect to claim 2, in my opinion, a POSA would have been motivated to apply any of the additional hydrogen separation steps as taught by Terrien to the combined method of Martin/Rafati to increase the yield of the

desired hydrogen product by separating at least an additional hydrogen-rich stream, would have known in view of what was known in the art and the teachings of these references how to implement the separations and would have reasonably expected success in that implementation.

401. With respect to claim 3, in my opinion, a POSA would have been motivated to employ any of the additional specific membrane hydrogen separation steps as taught by Terrien to the combined method of Martin/Rafati again to increase the yield of the desired hydrogen product by separating an additional hydrogen-rich stream. The use of a hydrogen selective membrane to separate hydrogen from synthesis gas product streams was well known in the art at least by the critical date. In my opinion, a POSA would have known how to implement the membrane separation at least in view of what was known in the art, and the teachings of the references and would have reasonably expected success in that implementation.

402. With respect to claim 4, Terrien teaches separation of a second hydrogen-rich stream using membrane separation and recycling of that stream to a pressure swing adsorption unit. EX1024, ¶[0045]. Hydrogen separation/purification by use of selective membranes and pressure swing absorption processes was well known in the art at least at the critical date. In my opinion, a POSA would have been motivated to employ the hydrogen recycling taught by Terrien to the combined method of Martin/Rafati to increase the yield

of the desired hydrogen product, but also to generally increase the efficiency of the entire hydrogen generation process. In my opinion, a POSA would have known how to implement the hydrogen recycling at least in view of what was known in the art and the teachings of the references and would have reasonably expected success in that implementation.

403. With respect to claim 5, Terrien teaches the use of membrane separated hydrogen rich gas streams as fuel gases for heating a reformer, specifically exemplified as a steam methane reformer. EX1024, ¶[0078]. Terrien also discloses an ATR (autothermal reformer) as a suitable alternative for a steam methane reformer in a hydrogen production process. EX1024, ¶¶[0019], [0072]. In my opinion, therefore, a POSA would have been motivated to employ the recycling of hydrogen-rich permeate of Terrien as fuel for preheating the feed of the ATR of the combined method of Martin/Rafati in view of the teachings of the references and what was known in the art at the time about heating of feed gas to autothermal reformers to at least improve capture of carbon dioxide. It was known in the art long before the critical date that it was beneficial to provide a fired heating unit to heat feed gas entering the autothermal reformer. I note that Martin shows the use of a fired heater to heat feed entering the parallel reformer configuration of Figure 7 and that Rafati at least discusses the benefits of preheating feed to reformers. EX1015, ¶[0019]. Thus, in my opinion, a POSA would have recognized that in the combined

Martin/Rafati process employing an endothermic reforming step (GHR) and an autothermal reforming step that recycling of hydrogen-rich permeate to fuel a reformer as taught by Terrien would have provided increased efficiency of carbon capture. In my opinion, a POSA would have known how to implement the hydrogen recycling to fuel at least to a fired heater which is used as a feed gas heating unit of an ATR, as is shown in Martin Figure 7. At least in view of what was known in the art and the teachings of Terrien and would have reasonably expected success in that implementation again in view of the similarities in method steps, process units, intermediates and final products.

2. Martin and Rafati in View of Terrien Disclose the Limitations of Claims 2-5.

(a) Claim 2: hydrogen is separated from the second residual gas stream, thereby producing a second hydrogen-rich stream and a third residual gas stream.

404. In my opinion, the combination of Martin and Rafati teaches each and every element of claim 1 from which claim 2 depends. *See*, Sections XI.E.2.(a)–(p), *supra*.

405. In the '805 Patent RG2 is residual gas exiting the cryogenic CO₂ separation unit **205** and is described as being introduced into a membrane unit **206** to separate H₂ and generate HG2 enriched in hydrogen and a third residual gas stream RG3. EX1001, col.12, ln.20–29, Figs. 2–3.

406. The combination of Martin and Rafati generates hydrogen-containing residual gas (from a cryogenic CO₂ separation unit) “WASTE GAS”

(Figure 7) and vapor phase stream **313** (FIG. 2), respectively. Residual gas from CO₂ separation (“Waste Gas”, Figure 7) is recycled “back to the reforming reactors to maximize the process efficiency and CO₂ capture.” EX1006, p.12. Stream **313** derives from cryogenic CO₂ removal from stream **311** (“contains all the CO₂ plus CO, H₂, CH₄, Argon, N, and traces of water”) and thus contains hydrogen. EX1015, ¶[0131]. Stream **313** can be recycled for a variety of uses, and specifically to hydrocarbon feed stream **303** EX1015, ¶[0132]. “WASTE GAS” (Figure 7) is not expressly described as containing hydrogen, but in view of the similarities in feed gas and process steps, in my opinion, a POSA would have recognized that the “WASTE GAS” of Martin Figure 7 contains hydrogen.

407. Terrien in FIG. 5 describes a carbon dioxide lean non-condensable stream **10** from a CO₂ separation unit **8**. In view of the description of compression/liquefaction and distillation steps and the components described (EX1024, ¶[0038]) and the components and process flow illustrated (EX1024, FIG. 4), the CO₂ separation unit **8** is in my opinion a cryogenic CO₂ separation unit. The carbon dioxide lean non-condensable stream **10** from this cryogenic CO₂ separation unit clearly contains hydrogen because Terrien states that it is passed to hydrogen selective membrane **11** for additional separation of hydrogen to generate hydrogen-rich permeate **12** (EX1024, ¶[0041]) and a second hydrogen lean residual stream **13** (EX1024, ¶[0042]).

408. In my opinion:

(1) carbon dioxide lean non-condensable stream **10** of Terrien is a highly carbon dioxide depleted residual gas stream of cryogenic CO₂ separation, and is the same as the second residual gas RG**2** of the '805 Patent;

(2) hydrogen rich permeate **12** of Terrien is a hydrogen-rich stream enriched with hydrogen compared to **10** by passage through membrane **11**, and is the same as the second hydrogen rich gas HG**2** of the '805 Patent ; and

(3) second hydrogen lean residual stream **13** of Terrien is a residual gas stream depleted of hydrogen compared to carbon dioxide lean non-condensable stream **10** by its failure to pass through membrane **11**, and is the same as the third residual gas RG**3** of the '805 Patent. Thus, the combination of Martin and Rafati in view of Terrien teaches producing the second hydrogen-rich stream and the third residual gas stream as claimed in claim 2 of the '805 Patent.

(b) Claim 3: hydrogen is separated from the second residual gas stream (RG2) by membrane separation.

409. In my opinion, the combination of Martin and Rafati in view of Terrien teaches all of the elements of '805 Patent claim 2 from which claim 3 depends. *See, Section XI.G.2(a), supra.*

410. In the '805 Patent, RG**2** exits cryogenic CO₂ separation unit **205** and is described as being introduced into a membrane unit **206** to generate HG**2** and RG**3**. EX1001, col.12, ln.20–29.

411. As discussed in Section XI.G.2(a), *supra*, it is my opinion that “WASTE GAS” (Figure 7) of Martin, vapor phase stream **313** (FIG. 2) of Rafati, and carbon dioxide lean-condensable stream **10** of Terrien are each equivalent to the second residual gas stream RG2 as described in the ’805 Patent. In my opinion, hydrogen rich permeate **12** of Terrien is the same as the second hydrogen-rich gas stream HG2 as described in the ’805 Patent and second hydrogen lean residual stream **13** of Terrien is the same as the third residual gas stream RG3 as described in the ’805 Patent. In FIG. 5 of Terrien, stream **10** is passed to a hydrogen selective membrane **11** to generate hydrogen-rich permeate **12**. In my opinion, the hydrogen selective membrane **11** of Terrien is the same as the membrane unit **206** of the ’805 Patent. Thus, the combination of Martin and Rafati in view of Terrien teaches producing a second hydrogen-rich stream and a third residual gas stream using membrane separation as claimed in claim 3 of the ’805 Patent.

(c) *Claim 4: the second hydrogen-rich stream is supplied to the fourth synthesis gas stream for separation of hydrogen by pressure swing adsorption in step (e).*

412. In my opinion, the combination of Martin and Rafati in view of Terrien teaches all of the elements of claim 2 from which claim 4 depends. *See*, Section XI.G.2(a), *supra*.

413. In the ’805 Patent, HG2 exits membrane unit **206** (Figures 2 and 3) and is optionally recycled to the synthesis gas stream (SG4) which enters PSA unit **204** to increase overall hydrogen yield. EX1001, col.12, ln.29–31.

414. Terrien describes carbon dioxide lean non-condensable stream **10** which contains hydrogen from a cryogenic CO₂ separation unit **8**. EX1024, FIG. 5, ¶[0077]. Stream **10** is introduced to hydrogen selective membrane **11** to generate hydrogen rich permeate **12**. EX1024, ¶[0041], [0077]. The hydrogen-rich permeate **12** is recycled to feed gas stream **19** which can be synthesis gas. EX1024, FIG. 5. Feed gas **19** enters process unit **0**, which contains a hydrogen separation unit, specifically a PSA unit. EX1024, ¶[0020]. In my opinion, hydrogen-rich permeate **12** is the same as a second hydrogen-rich gas stream HG2 as described in the '805 Patent, stream **19** is the same as a fourth synthesis gas stream SG4 as described in the '805 Patent. Process unit **0** contains a PSA unit which is the same as PSA unit **204** of the '805 Patent. Thus, the combination of Martin and Rafati in view of Terrien discloses supplying the second hydrogen-rich stream to the fourth synthesis gas stream for separation of hydrogen by pressure swing adsorption in step (e) as claimed in claim 4 of the '805 Patent.

(d) Claim 5: gases present in the second hydrogen-rich stream are utilized as fuel gases for heating in the autothermal reforming step of step (c).

415. In my opinion, the combination of Martin and Rafati in view of Terrien teaches all of the elements of claim 2 from which claim 5 depends. *See*, Section XI.G.2(a), *supra*.

416. The '805 Patent states that “gases present in the hydrogen-rich stream HG2 are utilized as fuel gases for heating in the autothermal reforming

step of step (c)”. EX1001, col.6, ln.9–11. It is explained in the ’805 Patent that for the autothermal reforming step, the feed gas has to be heated which can be done using a fired heating unit. EX1001, col.6, ln.9–13. The fired heating unit of the autothermal reformer may be heated at least partly with the aid of gases present in HG2 that have high calorific value (CO, methane and hydrogen). EX1001, col.6, ln.11–18. Calorific value, as is known in the art, is the amount of heat present in a fuel determined by the complete combustion of a specified quantity of fuel at constant pressure and in normal conditions.

417. Terrien generally teaches the recycling of hydrogen-rich gases from hydrogen membrane separation units (EX1024, Abstract; ¶[0008]) as feed or fuel, particularly in hydrogen generation plants, to improve efficiency of hydrogen and carbon dioxide recovery (EX1024, ¶[0008]). More specifically, Terrien teaches recycling of hydrogen-rich streams **27**, **5** and **12** as feed and fuel, particularly in hydrogen generation plants. In Terrien (EX1024, FIG. 5), hydrogen-rich permeate **12** is shown as optionally recycled to steam reformer **23** as feed entering the steam reformer together with feed stream **22**. Alternatively, hydrogen-rich permeate **12** can be utilized as fuel gas for the reformer **23**. Terrien states that when the method is employed in a hydrogen generation plant as shown in FIG. 5, hydrogen rich permeate stream **12** is preferably used as a fuel to steam methane reformer **23** resulting in a boost in carbon dioxide capture and to eliminate or minimize natural gas fuel. EX1024,

¶¶[0078], [0085]. While Terrien specifically exemplifies the use of a steam reformer in the processes of FIGs. 5 and 6, Terrien also teaches that an autothermal reformer unit (ATR), among others, can be used to generate feed gas for the purification process. In my opinion, the teaching of Terrien to employ hydrogen-rich permeate **12** as fuel in a steam reformer and the teaching of Terrien that reforming can be accomplished with an ATR strongly suggest to a POSA to employ hydrogen-rich permeate **12** as fuel in an autothermal reforming step and more specifically as fuel in a fired heater to heat feed entering an autothermal reformer. It was known in the art before the critical date that autothermal reformers can employ fired heaters to preheat feed entering the ATR to improve process efficiency and decrease use of methane for combustion. In my opinion, the suggestion of Terrien is amplified by the teaching of Rafati to preheat feed streams into the reformers to improve efficiency of synthesis gas production. EX1015, ¶[0019]. Further, Martin shows such a fired heater used to preheat feed into a parallel (GHR with ATR) configuration. In my opinion, in view of the disclosure of Terrien and the combined disclosure of Martin and Rafati and what was known in the art, a POSA would have recognized at least at the critical date, that hydrogen-rich streams generated from hydrocarbon reforming as in the combination of Martin and Rafati would be useful for fuel to preheat feed entering an ATR and would result in the increased efficiency of hydrogen and carbon dioxide recovery. In

my opinion, the specific teaching of use of a fired heater with the parallel reformer configuration (GHR with ATR) of Martin in Figure 7, specifically teaches use of such a fired heater to preheat feed gases in such hydrogen generation systems. In my opinion, a POSA would have been specifically motivated in view of Terrien's teachings to employ a hydrogen-rich gas stream, generated by removing hydrogen from a residual gas stream from a cryogenic carbon dioxide separation step, as fuel to heat the ATR in the combined method of Martin and Rafati to achieve the benefits taught by Terrien.

418. In my opinion, hydrogen-rich permeate **12** is the same as the second hydrogen-rich stream HG2 as described in the '805 Patent, and Terrien teaches use of permeate **12** as fuel in an autothermal reformer. The combined process of Martin and Rafati provides a combination of an endothermic reforming step (a GHR) and an autothermal reforming step (an ATR) and shows heating of reformer feed with a fired heater. Providing a fired heater to preheat feed entering an ATR was well known at the critical date. Thus, the combination of Martin and Rafati in view of Terrien discloses utilizing gases present in the second hydrogen-rich stream as fuel gases for preheating in the autothermal reforming step of step (c) as claimed in claim 5 of the '805 Patent.

XII. ACKNOWLEDGMENTS

I, Harald Klein, declare under penalty of perjury under the laws of the United States of America that the foregoing is true and current.



Executed on 28.06.25