

UNITED STATES PATENT AND TRADEMARK OFFICE

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BEFORE THE PATENT TRIAL AND APPEAL BOARD

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SYMRISE AG and SYMRISE INC.,  
Petitioners,

v.

INOLEX INVESTMENT CORP.,  
Patent Owner.

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IPR2023-01035  
U.S. Patent No. 10,995,048

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**SECOND DECLARATION OF MICHAEL E. WRIGHT, Ph.D.**

I, Michael E. Wright, Ph.D, declare and state as follows:

1. I am the same Michael E. Wright who submitted a Declaration dated February 7, 2024 (hereinafter, the “First Declaration”), in this proceeding on behalf of Patent Owner, Inolex Investment Corp. (“Inolex” or “Patent Owner”) in support of Patent Owner’s Revised Non-Contingent Motion to amend the challenged claims of U.S. Patent No. 10,995,048 (“the ’048 patent”).

2. The opinions and conclusions I express in this declaration are based on my education, my extensive experience in the fields of heterogeneous catalysis, homogeneous catalysis, organic and organometallic synthesis, and my review of materials related to this matter.

3. My Qualifications set forth in my First Declaration (**Ex. 2001**) have not changed and are incorporated herein by reference.

4. I still have no financial interest in the outcome of this proceeding and am not being compensated for my time spent working on this matter.

## **I. MATERIALS REVIEWED**

5. In forming my opinions set forth herein, I reviewed, among other things, the ’048 patent (**Ex. 1001**), as well as the First and Second Declarations of Douglass F. Taber (**Ex. 1002** and **Ex. 1038**) and the materials referenced therein (**Ex. 1007-Ex. 1037** and **Ex. 1039-Ex. 1044**). I also reviewed my first Declaration (**Ex. 2001**) and the following materials:

| Materials Reviewed |                                                                                                                                                                                                                                                                                  |
|--------------------|----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| <b>Ex. 2002</b>    | J. Ross, “Heterogeneous Catalysis,” Elsevier B.V. (2012)                                                                                                                                                                                                                         |
| <b>Ex. 2003</b>    | Kim <i>et al.</i> , “Dual-bed catalytic system comprising Al <sub>2</sub> O <sub>3</sub> and Ba/Al <sub>2</sub> O <sub>3</sub> with enhanced 1-octene productivity in 1-octanol dehydration for linear $\alpha$ -olefin production,” <i>J. Ind. Eng. Chem.</i> 119:376-85 (2023) |
| <b>Ex. 2004</b>    | Experimental data generated by Inolex, Inc. for the conversion of 1-octanol to octenes                                                                                                                                                                                           |
| <b>Ex. 2005</b>    | Deposition Testimony of Douglass F. Taber, January 23, 2024                                                                                                                                                                                                                      |
| <b>Ex. 2007</b>    | Deposition Testimony of Douglass F. Taber, June 11, 2024                                                                                                                                                                                                                         |
| <b>Ex. 2008</b>    | Int’l Patent Pub. No. WO 2022/122139 A1 (“Bugdahn”)                                                                                                                                                                                                                              |
| <b>Ex. 2009</b>    | D.J. Ward <i>et al.</i> , “Sustainable routes to alkenes: applications of homogeneous catalysis to the dehydration of alcohols to alkenes,” <i>Catal. Sci. Technol.</i> 13:2638-2647 (2023)                                                                                      |

## II. BACKGROUND OF THE INVENTION

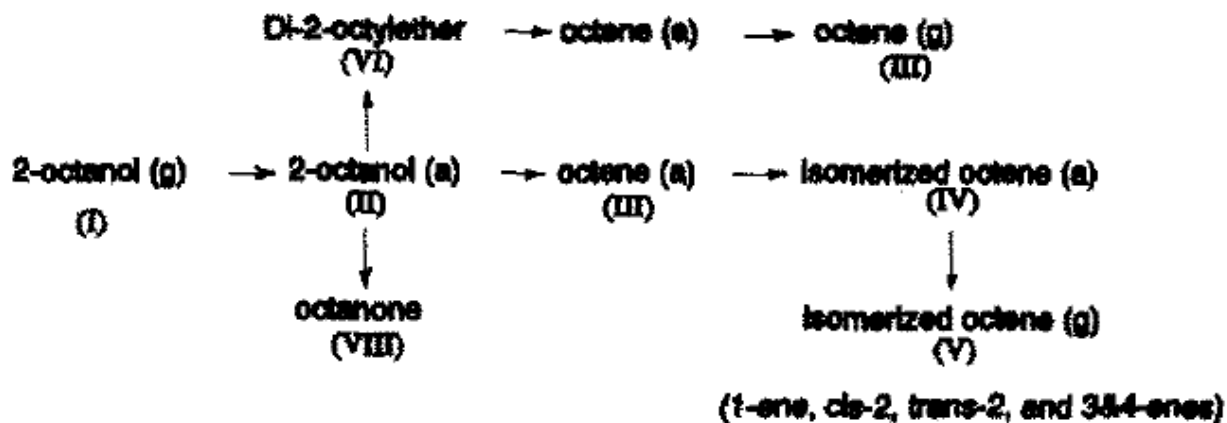
6. The preparation of 1-alkenes from 1-alcohols is an important industrial process for a wide range of chemical products. A good summary is provided in *Makgoba* (**Ex. 1027**), which describes the importance of catalysts capable of providing high 1-alkene regioselectivity for extended periods of time-on-stream (i.e., TOS). Generally speaking, there are significant commercial and societal advantages for catalysts that are capable of providing extended time-on-stream without a reduction in 1-alkene regioselectivity. Replacing catalyst materials and using a recycling stream(s) wastes energy, precious infrastructure

resources, time of employees, and in general counters the ultimate goal to replace petroleum products using bioalcohols in a way that creates a renewable and sustainable society. There continues to be a constant need and pressure to invent new catalysts that deliver unquestionable advantages in energy use and atom efficiency (i.e. high product yields of a target molecule/product). Inolex's '048 patent does just that by demonstrating and teaching (in examples) the art of using a novel catalyst preparation method and reaction methods/conditions to employ this superior and unexpected catalyst.

7. Before further discussing the background of the technology, it is also important to note that statements in Dr. Taber's First and Second Declarations and testimony on cross examination raise significant questions concerning his understanding of the process under which heterogeneous catalytic dehydration of 1-alkanols to 1-alkenes occurs. My conclusion, is although I certainly respect Dr. Taber's extensive research background and accomplishments, it is very evident that he does not have the background in heterogeneous catalysis to operate as POSITA, let alone be considered an expert, in the technology of the '048 patent. For example, Dr. Taber first referred to "calcined" as a "rather archaic term" that a POSITA "might operate without ever hearing the word" and could not predict what time and temperature would activate a catalyst; however, in his more recent statements "calcined" now seems to be a common process that is required for

virtually all  $\gamma$ -alumina catalysts and is completely predictable. Compare **Ex. 2005**, p. 69, l. 13 – p. 70, l. 3; p. 70, l. 16 – p. 71, l. 14 with **Ex. 2007**, p. 62, l. 14 – p. 65, l. 6; p. 83, l. 4 – p. 86, l. 6; p. 89, l. 13 – p. 91, l. 19; p. 97, l. 16 – p. 98, l. 6; p. 107, ll. 1-16; p. 115, l. 19 – p. 116, l. 9; p. 118, ll. 1-5, 14-17; p. 122, ll. 6-18;

8. Further, in his discussion of the technology of the '048 patent, Dr. Taber cites to a sophomore level organic chemistry textbook, *Wade* (**Ex. 1030**) that discusses the acid-catalyzed solution phase dehydration of alcohols that proceed through a carbocation intermediate followed by deprotonation. **Ex. 1002**, ¶¶35-41 (citing **Ex. 1030**, pp. 313-14). However, as any POSITA would understand, the catalytic dehydration of 1-alkanols using the  $\gamma$ -alumina catalysts at issue occurs by flowing the 1-alkanol over the catalyst in the gas phase, and molecules react in an adsorbed phase on active sites of the catalysts. A good summary of the possible reactions that can occur during the catalytic dehydration of 2-octanol is provided in *Chokkaram*:



**Scheme 1.**

**Ex. 1041**, p. 164, col. 2 (Scheme 1). Despite having reviewed numerous prior art references such as *Chokkaram*, Dr. Taber's Second Declaration cites references such as *Takamura* (**Ex. 1044**), which discusses the catalytic dehydration of 1-alcohols in liquid solution over a solid catalyst. A POSITA would not find references such as *Takamura* (**Ex. 1044**) or *Wade* (**Ex. 1030**) relevant the technology of the '048 patent. Furthermore, the calcining used in *Chokkaram* is the calcining from the aluminum hydroxides (plus other metals) to form the original "alumina" matrix and has nothing to do with calcining an already formed gamma-alumina. See **Ex. 1041**, pp. 158-164. A POSITA would be well aware that the change in surface area during catalyst formation (as in **Ex. 1041**) has no correlation to that of calcining an already formed  $\gamma$ -alumina catalyst that has been modified with a selected promoter.

9. Dr. Taber's lack of a basic understanding of how gas-phase catalysis is performed is further evident by his testimony that liquid reagents actually come into contact with the catalyst, and therefore lead to degradation. Specifically, Dr. Taber testified as follows:

Q. So just so we're clear, in all of these examples we're not adding liquid on to any of these reactors in Wu, in Wright '788, in Wright '451, or is it your testimony that literally wet liquid phase reagents are being added onto the catalyst?

A. Well, liquid phase starting materials are being added. At some point they vaporize at the temperatures of the reactor.

Q. So you're saying they're not preheated?

A. There may be a preheating in the reactor.

Q. Before the reagents get to the reactor your testimony is they enter the reactor as liquids and then hit the catalyst?

A. I don't see a reactor design outlined here. So I do not know.

Q. How about DeBruyn and Le, are they adding liquid reagents on to their reactors?

A. Nor do they have a reactor design. So I don't know.

Q. What is your understanding about how these processes are conducted under normal conditions in the art? Is it your testimony that under normal conditions liquid reagents are added directly into the reactor?

A. From an engineering point of view there would have to be containment of the substrate, of the starting material going in. And so in the world outside the reactor these starting materials are liquids. Somehow they get into the reactor, not necessarily the catalyst bed but into the reactor, they would have to go in as liquids. If you define

the reactor as only being the catalyst bed, then presumably the liquids are not coming into contact with the catalyst bed. But if you define the reactor as being the whole reactor from where you put the substrate in, then yes, they go in as liquids.

**Ex. 2007**, p. 143, l. 21 – 145, l. 14. However, a POSITA working in this field would know that liquid phase reactants never come into contact with the catalyst, and that the reactants are always in the gas phase or adsorbed onto the catalyst. The prior art need not spell out this basic concept in order for it to be understood, but in case there is any doubt, it is stated in *Chokkaram* (**Ex. 1041**) as noted above, and *Wright '451*, for example, explicitly states that “[a] preheater/vaporizer immediately upstream of the reactor was used to ensure that the nominally 100% 1-butanol feed (no diluent) was completely vaporized prior to delivery to the top of the upstream bed of glass beads.” **Ex. 1039**, ¶ [119].

10. A POSITA working in the field of alcohol dehydration would also know that alcohol dehydrations are endothermic and ether formation is exothermic. See, e.g., **Ex. 2009** (“*Ward*”). Specifically, *Ward* states: “[f]or ethanol, dehydration to ethene is favoured at high temperatures (starting at 300 °C) as this is an endothermic reaction, whereas the exothermic formation of diethyl ether is favoured at lower temperatures.” **Ex. 2009**, p. 2640.<sup>1</sup> This is just one more example

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<sup>1</sup> I am aware that *Ward* itself is not prior art to the '048 patent. However, it is reflective of the fact that a POSITA would have been well aware that dehydration is an endothermic process. Indeed, the references cited by *Ward* are from 2013 and

that Dr. Taber lacks the basic knowledge needed to evaluate what is normal, what is expected, and what is required to create a truly innovative and new heterogeneous catalyst/process, even at the level of a POSITA.

11. Dr. Taber also testified that he has no knowledge of “WHSV,” which is a basic concept in heterogeneous catalysis a POSITA would surely be aware of and is an acronym for “Weight Hour Space Velocity” and defines the weight of the reactant feed flowing per unit weight of catalyst per hour. **Ex. 2005**, p. 64, ll. 10-19. Optimizing and controlling the WHSV is one of only two fundamental process control parameters any POSITA would understand before undertaking the dehydration of a bioalcohol using a heterogeneous catalyst. The other being temperature. Again, the POSITA in this matter has a B.S. in chemistry with at 1-2 years of industrial experience, or a masters degree with commensurate experience. In the general area of heterogeneous catalysis, any POSITA would understand WHSV and how to adjust it for achieving optimum catalyst performance.

12. Further, Dr. Taber testified that differences in temperature such as 290 °C vs. 315 °C “may not be very different” because the dehydration is exothermic and factors such as “the air conditioner kicks on in the other corner of the lab all of a sudden heat loss is greater” would lead to temperature deviations in

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2015 (i.e., M. Zhang and Y. Yu, *Ind. Eng. Chem. Res.* 52:9505-9514 (2013) and T. K. Phung, L. Proietti Hernández, A. Lagazzo and G. Busca, *Appl. Catal., A* 493:77–89 (2015), respectively).

the reactor of plus or minus twenty degrees. See **Ex. 2007**, p. 161, p. 1 – p. 163, l. 9. However, in my experience the flow rate of vapor-phase reagents into the reactor is always tightly controlled. Regardless of the endothermic or exothermic nature of the chemical reactions, heterogeneous reactors are fact designed (i.e. sufficient heating and/or cooling) to allow the operators to maintain a safe and accurate temperature control over the entire reactor. As mentioned above, alcohol dehydrations are endothermic, so heating is the issue for reactor designs. And, air conditioning in a room having any effect on a commercial chemical process reaction temperature, is not a reasonable statement from a POSITA.

13. Dr. Taber also testified that a POSITA would “have experience in industrial chemistry or industrial processes” and “experience working at a chemical company” (**Ex. 2005** at p. 22, ll. 4-13), which he does not possess (**Ex. 2005**, p. 21, ll. 17-22). And, while Dr. Taber testified that he has consulted with chemical companies, he did not testify that he discussed this matter or the technology with those colleagues. Collectively in the two depositions and two declarations, it is evident that Dr. Taber lacks experience and the basic know-how of how to evaluate heterogeneous catalyst synthetic methods, catalyst performance, and ultimate value or utility in a commercial setting. Using such descriptions as “the POSITA would do as instructed” hardly suggests a working knowledge of heterogeneous catalysis, nor does assist a finder of fact in the current matter.

14. Despite these significant and large gaps in his knowledge of the heterogeneous catalysis processes at issue in this matter, Dr. Taber was keen to point to statements in prior art references that, in his opinion, would have led a POSITA to the inventions described in the '048 patent. For example, despite *Wu*'s silence on whether any of the catalysts used were calcined, Dr. Taber testified that a POSITA would understand that calcination always follows base treatment. **Ex. 2007**, p. 89, l. 13 - p. 91, l. 19; p. 118, ll. 1-17. Based on my experience, one cannot assume calcination has been carried out. As shown in *Wright '451* and the '048 patent, calcination, and how it was done, can have a significant impact and unexpected effect on catalyst performance, under both short- and long-term use (i.e., the latter referring to high time-on-stream). The bottom line is that a POSITA would not infer or assume that a calcination step has been done when none is reported.

15. Turning back to the mechanism of catalytic dehydration at issue in this case, Makgoba discusses the mechanism whereby “a proton is transferred from a surface hydroxyl group to the alcohol oxygen, facilitating the transfer of a proton on the neighboring carbon to a basic oxygen atom on the surface.” **Ex. 1027**, p. 145, col. 2. The olefinic product then desorbs “and the water molecule remains adsorbed on the surface and acts as a reaction inhibitor.” **Ex. 1027**, p. 145, col. 2 (citing *Catalytic Chemistry*, John Wiley and Sons Inc., New York, pp. 371-373

(1992)). Thus, the ability of a catalyst for regioselective dehydrogenation of 1-alcohols to 1-alkenes to maintain high conversion and regioselectivity for extended durations of time-on-stream was and is an important consideration, and very difficult to achieve.

16. I note that Makgoba discloses processes whereby the conversion of 1-hexanol to 1-hexene is performed at 280 °C to 325 °C, while the conversion of 1-octanol to 1-octene is performed at 280 °C to 330 °C. **Ex. 1027** at 146, col. 2.

17. Typically, high regioselectivity for alcohol dehydration to 1-alkenes is obtained at lower temperatures while higher temperatures provide higher conversion but lower regioselectivity. *See, e.g., Ex. 1041; see also* Int'l Patent Pub. No. WO2022/122139 A1 ("Bugdahn," **Ex. 2008**). According to Bugdahn and co-workers at Petitioner, Symrise AG, "[u]sually, the selectivity of a reaction can be increased by using doped or modified catalysts. However, due to a decrease in reactivity generally higher reaction temperatures are required." **Ex. 2008**, ¶[0056].

18. Typically, base treatments of alumina, such as treatment with sodium, potassium, calcium and magnesium promoters, increase regioselectivity but decrease activity for alcohol dehydration. As stated by *Le*, "a basic catalyst, for example a barium-doped  $\gamma$ -alumina, is used to further increase the selectivity of the dehydration [and] ... an acidic catalyst is used to improve the reaction yield." **Ex. 1007**, ¶ [0064]. Similarly, Berteau, which is a new prior art reference cited by

Petitioners and Dr. Taber, shows in Figure 4 that conversion of 1-butanol to butene increases with increasing temperature with untreated alumina. **Ex. 1043**, p. 130.

Figure 6 of Berteau indicates that to achieve high conversion of 1-butanol to butene at lower temperatures, acid treatment is preferred over base treatment, such as treatment with sodium or magnesium promoters. **Ex. 1043**, p. 131. Indeed, Berteau states, “[c]ations, which present a negative specific effect on acidity, have to be eliminated for application in alcohol dehydration.” **Ex. 1043**, p. 135. Thus, there is a challenge in achieving high conversions of 1-alcohols to 1-alkenes at reaction temperatures low enough to maintain high regioselectivity for the 1-alkene.

19. The challenge of achieving high regioselectivity can be approached by running reactors under conditions that provide the desired regioselectivity, however, they provide lower alcohol conversion and other products (e.g. ethers). Overall net conversion of alcohol for such a process can be improved by isolating the unreacted alcohol and ethers (i.e. a purification step(s)) and reintroducing (and heating) those materials back into the reactant stream. *See, e.g.*, **Ex. 1008**, ¶ [0053] (Example 3). However, this comes with the price of increased catalyst amounts, higher energy inputs, strong potential of decreased catalyst lifetime (due to impurities in recycle stream), increased equipment cost and added failure points in the process, and an overall more complicated process to monitor and stay on

stream. And even after recycling the unreacted starting materials, *De Bruyn* claims to achieve a 94% conversion of 1-hexanol to alkenes. **Ex. 1008**, ¶ [0053], although no experimental details to prove this claim nor discussion of how this was calculated are presented.

### III. U.S. PATENT NO. 10,995,048

20. While noted in passing in my First Declaration, it is important that the methods disclosed in the '048 patent for dehydrating bio-1-alcohols to provide corresponding bio-1-alkenes with high-regioselectivity and a high alcohol conversion do so in a single pass through an isothermal fixed bed (or similar) chemical reactor. For example, the '048 patent teaches that:

The reactor can be designed as a simple heated reaction tube packed with catalyst and a flow of the gaseous bio-1-alcohol over the catalyst, preferably in a single pass affording greater than about 95% conversion, with greater than about 98% a preferred embodiment, and in all cases with no recycle stream used whatsoever in the overall process flow.

**Ex. 1001**, col. 15, ll. 27-32 (emphasis added). In my opinion, this is particularly notable and entirely unexpected given the general disclosures in the prior art. Specifically, none of the prior art of record in this proceeding teaches or suggests a process capable of dehydrating a C<sub>6</sub> to C<sub>10</sub> bio-1-alcohol to provide a bio-1-alkene in a conversion of at least 95% and a bio-1-alkene regioselectivity of 92% to about 99% using a constant flow a single pass reactor design. Thus, avoiding use of

excess energy to reheat a recycle stream, eliminates the potential and reality of catalyst contamination from recycle impurities, and assures a simple and continuous production of for example bio-1-octene and greatly contributes to a favorable life cycle analysis (LCA) of energy and resource uses.

21. As the '048 patent teaches, this remarkable regioselectivity and conversion is provided by heating the bio-1-alcohol in a reactor with a  $\gamma$ -alumina catalyst treated with a calcium promoter and then calcined at a temperature of about 400 °C to about 480 °C. See **Ex. 1001** at pp. 22-23, col. 26, l. 10 – col. 27, l. 35 (Examples 1-5); *see also* **Ex. 1006** at pp. 578-59 of 719, ¶¶ [0121]-[0127]; **Ex. 1005** at pp. 38-40 of 236, ¶¶ [0121]-[0127]. Specifically, the '048 patent reports the use of a calcium promotor-treated and then calcined  $\gamma$ -alumina catalyst to dehydrate 1-hexanol and 1-octanol. In the case of 1-hexanol, a  $\gamma$ -alumina catalyst was first treated with a calcium acetate solution, followed by calcination in air at 440 °C for 12 hours. Dehydration reactions conducted at 330 °C and 350 °C provided conversion (to 1-, 2-, and 3-hexenes) of 96.3% and 96.9%, respectively, with a regioselectivity for the formation of 1-hexene of 95.8%<sup>2</sup> and 96.8% (as reported in Table 5, below).

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<sup>2</sup> I note that the calculation of 1-Hexene Selectivity for Example 11A in Table 5 appears to be incorrect and should be 96.4%.

TABLE 5

| Catalyst    | Reactor Temperature (° C.) | 1-Hexene (%) | 2-Hexenes & 3-Hexene (%) | 1-Hexanol (%) | 1-Hexene Selectivity (%) |
|-------------|----------------------------|--------------|--------------------------|---------------|--------------------------|
| EXAMPLE 11A | 330                        | 92.8         | 3.5                      | 1.8           | 95.8                     |
| EXAMPLE 11B | 350                        | 93.8         | 3.1                      | 1.7           | 96.8                     |
| EXAMPLE 11C | 370                        | 90.1         | 3.2                      | 1.6           | 96.6                     |

(**Ex. 1001**, col. 29, ll. 1-25 (Example 11); **Ex. 1006** at p. 62 of 719, ¶ [0133],

**Ex. 1005** at p. 43 of 236, ¶ [0133].)

22. Surprisingly, increasing the reactor temperature to 370 °C reduced conversion (to hexenes) to 93.3% (from 96.9% at 350 °C), while maintaining a regioselectivity of 96.6% (as shown in Table 5, above). This increase in leading to less conversion was unexpected, particularly in view of prior art calcium-promoted catalysts such as those disclosed in *Wright '451* that provide conversion (i.e. higher reactor temperature equals higher conversion) in excess of 95% at 380 °C. *See Ex. 1039*, Figs. 10-12.

23. In the case of 1-octanol,  $\gamma$ -alumina was first treated with a calcium acetate (i.e., promoter) solution, followed by calcination in air at 440 °C or 500 °C for 12 hours. Dehydration conducted at 315 °C provided conversion (to octenes) of 97.2% and 95.5%, respectively, with a regioselectivity of 97.4%<sup>3</sup> and 93.1% (per Table 2, below). Thus, the '048 patent discloses that treatment with a calcium

<sup>3</sup> I note that the calculation of 1-Octene Selectivity for Example 1 in Table 2 appears to be incorrect and should be 96.7%.

promoter followed by calcination was important to obtain the claimed conversion and bio-1-alkene regioselectivity. Further, while both catalysts far exceed any known catalyst performance, it is noteworthy that a higher calcination temperature led to a less selective catalyst. Thus, the '048 patent teaches that higher calcination temperatures can be detrimental to catalyst performance—a truly unexpected result.

TABLE 2

| Catalyst  | Calcination Temperature (° C.) | 1-Octene (%) | 2-Octene & 3-Octenes (%) | 1-Octanol (%) | 1-Octene Selectivity (%) |
|-----------|--------------------------------|--------------|--------------------------|---------------|--------------------------|
| EXAMPLE 1 | 440                            | 94.0         | 3.2                      | 1.6           | 97.4                     |
| EXAMPLE 2 | 500                            | 88.9         | 6.6                      | 3.5           | 93.1                     |

(**Ex. 1001**, col. 26, l. 10 – col. 27, l. 9 (Examples 1-3); **Ex. 1006** at pp. 57-58 of 719, ¶¶ [0121]-[0125]; **Ex. 1005** at pp. 38-39 of 236, ¶¶ [0121]-[0125].)

24. The catalyst prepared in the '048 patent was removed from the reactor and photographed. The catalyst showed only minor discoloration, and notably, after 1400 h (~58 days ) of TOS, conversion efficiency and 1-octene regioselectivity remained absolutely steady. (See **Ex. 1001**, col. 29, ll. 26-35 (Example 12), Fig. 7; **Ex. 1006** at p. 62 of 719, ¶ [0135], Example 12, p. 79 of 719, Fig. 7; **Ex. 1005** at p. 40 of 236, ¶ [0135], Example 12, p. 54 of 236, Fig. 7.) The bio-1-octanol was not dried nor were any precautions taken to remove silicon-containing compounds from the 1-octanol feedstock. (Compare with **Ex. 1027** at

pp. 149-50.) In my opinion, a person of ordinary skill in the art (“POSITA”) would view the TOS achieved by the catalyst prepared in the ’048 patent as being entirely unexpected.

#### **IV. LEGAL STANDARDS**

25. In forming my opinions set forth herein, I applied the legal standards set forth in my First Declaration, which are incorporated herein by reference, as well as the following legal standards that were explained to me.

##### **A. Obviousness of a Range**

26. I have been informed that a prior art disclosure of a range, such as for example 200 to 600 °C, is no more a disclosure of the end points of the range than is of each of the intermediate points. In such instances, the relevant inquiry for a POSITA would be to look at specific temperatures that were used, for instance in working examples, in order to determine values in a range that were specifically disclosed.

27. I have been informed and understand that a process or process step that requires a range of values—for example, a temperature range—is generally considered obvious where prior art discloses a broader range that encompasses the claimed values or a range that overlaps with a portion of the claimed values. I also understand that, in such instances, the relevant inquiry in assessing the obviousness of a claimed range is to ask whether a POSITA would have been motivated to

select the claimed range based on the prior art as a whole. I have been informed that, also relevant to this inquiry, it is necessary to assess whether there is evidence, for example, that: (1) the prior art taught away from the claimed range of values; or (2) the claimed range provides any new and unexpected results compared to the prior art.

28. I have been informed that a prior art reference is said to teach away when a POSITA, upon reading the reference, would be discouraged from following the path set out in the reference, or would be led in a direction divergent from the path that was taken by the inventors. I am also informed that a reference does not teach away if it merely expresses a general preference for an alternative invention but does not criticize, discredit, or otherwise discourage investigation into the range or invention that is claimed.

29. I have also been informed that results provided by a claimed range are considered “unexpected” (and therefore probative of non-obviousness), where there is a difference in kind, and not merely in degree, over the results provided by a prior art range.

## **B. Obvious to Try**

30. I have been informed and understand that a process or process step may be “obvious to try” when there is a design need or market pressure to solve a problem and there are a finite number of identified, predictable solutions, and a

POSITA has good reason to pursue those known options within his or her technical grasp. If this leads to the anticipated success, it is likely the product not of innovation but of ordinary skill and common sense.

### **C. Anticipation**

31. I have been informed and understand that a claimed process is anticipated where a single prior art reference describes the same invention as that which is claimed. That is, each and every element of the claim at issue must be disclosed within a single prior art reference in the same manner as recited in the claim at issue (without the necessity for a POSITA to pick and choose or combine various aspects of a prior art disclosure).

### **D. Claim Construction**

32. I understand that in its Preliminary Guidance (**Paper 16**), the PTO found that “base-treated and calcined  $\gamma$ -alumina catalyst” merely requires that the  $\gamma$ -alumina catalyst is both base-treated and calcined without requiring any particular order. **Paper 16**, at 7-8. I agree with this construction, but understand that Inolex now seeks to revise substitute claim 29 (and claims depending therefrom) to explicitly require “a calcium-promoted  $\gamma$ -alumina catalyst calcined at a temperature of about 420 °C to about 480 °C.” In my opinion a POSITA would readily understand the meaning of these terms and no construction is required. This

appears to comport with the PTO's finding concerning the term "calcined." **Paper 16**, at 8.

33. In my opinion, a POSITA reviewing the revised substitute claims would, in view of the specification of the '048 patent, understand the words used in those claims 29-44 to have their plain and ordinary meaning.

34. Nonetheless, in case there is any doubt, a POSITA would understand that the plain and ordinary meaning of "a calcium-promoted  $\gamma$ -alumina catalyst" is "a  $\gamma$ -alumina catalyst that has been modified by reacting with a  $\text{Ca}^{+2}$  compound." A POSITA would also understand that "a calcium-promoted  $\gamma$ -alumina catalyst" contains sufficient calcium as  $\text{Ca}^{+2}$  in order to enhance or "promote" the reactivity of the  $\gamma$ -alumina catalyst. As the '048 patent discloses, a preferable amount of calcium present in the calcium-promoted  $\gamma$ -alumina catalyst is about 1 wt-% to about 2 wt-% based on the weight of CaO determined after calcination. **Ex. 1001**, col. 8, ll. 43-45.

35. Further, a POSITA would understand that the plain and ordinary meaning of "from a single-pass reaction" is a single pass of the bio-1-alcohol through the specified continuous flow reactor at a temperature of 240 °C to 360 °C with a calcium-promoted  $\gamma$ -alumina catalyst calcined at a temperature of about 420 °C to about 480 °C, wherein the catalyst has not been silanized." A POSITA would further understand that "from a single-pass reaction" excludes any

affirmative steps of isolating unreacted bio-1-alcohol, ethers (and possible contaminants) from the effluent stream of the continuous flow reactor and recycling it in a second pass through the reactor in order to attempt an apparent improvement of the overall conversion of bio-1-alcohol to bio-1-alkene.

36. Lastly, the substitute claims also include the term “about” which is a term of approximation and should be given its plain and ordinary meaning of “approximately.” In the context of this technology, an approximate “temperature” for example, does not refer to a POSITA’s ability to monitor temperature (which can be done within a tenth of a degree) but to control the temperature of a reactor or a calcination process. In that context, based on my experience a POSITA would understand “about” to include deviations of at most plus or minus five degrees centigrade from a specified temperature.

**E. The Level of Skill In The Art and Predictability In the Art Inform A POSITA’s Understanding of The Prior Art**

37. As discussed in my First Declaration, for purposes of this matter the PTAB has found that a POSITA at the time of the invention would have had “a bachelor’s degree and two or more years of experience or a master’s degree in organic synthesis or chemical engineering, with education substituting for experience and vice versa.” **Paper 7** (Decision to Institute) at 6-7 (citing **Ex. 1002** at ¶ 16). In my opinion, this level of education and experience would not permit a POSITA to make inferences beyond the examples and most preferred

embodiments disclosed in a particular prior art reference—for example, as to whether experimental results concerning a specific catalyst’s conversion and regioselectivity could be analogized to other seemingly similar catalysts, or whether experimental results concerning a specific catalyst’s conversion and regioselectivity for a particular substrate could be extended to other similar compounds. See **Ex. 1040** at 144:11 – 146:19.

38. In addition to the level of ordinary skill in the relevant art, I understand that the level of predictability in the art can significantly impact an analysis of whether a particular claimed process is deemed non-obvious over the prior art. Here, the field of organic synthesis using heterogeneous catalysis has been an active area of research for more than 70 years. However, organic synthesis using heterogeneous catalysis remains a highly unpredictable field. As I testified at my deposition, there is no way to predict how the treatment of a catalyst with, for example, a particular base, promoter, or other chemical will affect its performance for conversion and regioselectivity of a given substrate without conducting experiments. See **Ex. 1040** at 43:18 – 45:16; 139:1-20.

## **V. THE SCOPE AND CONTENT OF THE PRIOR ART**

39. Petitioners and Dr. Taber cite several new prior art references: *Wright ’451* (**Ex. 1039**), *Chokkaram* (**Ex. 1041**), *Cortez* (**Ex. 1042**), *Berteau* (**Ex. 1043**), *Takamura* (**Ex. 1044**) in their Opposition (**Paper 14**) as well as Dr. Taber’s

Second Declaration (**Ex. 1038**). Further, Petitioners and Dr. Taber also introduce the new prior art combination of *Wu* (**Ex. 1025**) and *Chokkaram* (**Ex. 1041**).

**A. Wu (Ex. 1025)**

40. U.S. Patent No. 4,234,752 (“*Wu*” **Ex. 1025**) is titled “Dehydration of Alcohols,” and relates to research into the use of  $\gamma$ -alumina catalysts for the dehydration of alcohols that was conducted by Phillips Petroleum Co. *Wu* was published on November 18, 1980.

41. *Wu* discloses the concept that base-treated  $\gamma$ -alumina catalysts can provide increased 1-alkene regioselectivity (i.e., reduced double-bond isomerization) by neutralizing acidic sites on the alumina. **Ex. 1025**, col. 1:21-26. The “[a]cidic sites on the alumina surface may be neutralized by treatment of the alumina with aqueous potassium hydroxide or aqueous sodium hydroxide, followed by washing with water and subsequent drying, or otherwise altered.” **Ex. 1025**, col. 2:32-36. Thus, a POSITA would understand *Wu* as teaching base-treatment of  $\gamma$ -alumina catalysts followed by drying. *Wu* does not teach or suggest that calcination can have any effect on the performance of  $\gamma$ -alumina catalysts, as measured by conversion, regioselectivity, or time-on-stream.

42. For the conditions of the dehydration reaction, *Wu* teaches that “temperatures in the range of from about 200° to 500° C. are suitable for the

desired dehydration reaction to occur. It is currently preferable, however, to employ temperatures in the range of 300°-450° C.” **Ex. 1025**, col. 2:51-54.

43. *Wu* provides two examples, both of which show that dilution of the 1-alcohol starting material (3-methyl-1-butene) with an inert carrier gas can increase the conversion of 1-alcohol to 1-alkene (in *Wu*’s case, 3-methyl-1-butene).

44. As discussed below, *Wright ’451* teaches away from the use of alkali-base modification to  $\gamma$ -alumina catalysts as taught by *Wu*. See **Ex. 1039**, ¶¶ [42], [98], Table 2, Fig. 9. This is because the alkali-base modification does not maintain robust conversion as a function of time-on-stream. Instead, *Wright ’451* shows that conversion as a function of time-on-stream can be maintained by silanization and the use of a calcium-promoted  $\gamma$ -alumina catalyst.

#### **B. *Chokkaram* (Ex. 1041)**

45. *Chokkaram* (**Ex. 1041**) was published in 1997, and relates to work performed at the University of Kentucky’s Center for Applied Energy Research to characterize nickel-alumina and cobalt-alumina catalysts. These catalysts differ significantly from promoted or base-treated alumina in that nickel and cobalt are present in stoichiometric amounts throughout the catalyst structure. Further, these mixed metal aluminates are not  $\gamma$ -alumina, and do not have a catalytic surface that approaches that of  $\gamma$ -alumina. Rather, their reactivity, calcination, and performance have no bearing on the present matter that relates to whether a POSITA would

have a reasonable expectation of the catalytic performance of a calcium-promoted  $\gamma$ -alumina catalyst calcined at a specific temperature.

46. The catalysts were prepared by co-precipitation of Al/Co and Al/Ni from aqueous nitrate solution by adding ammonium carbonate (1 M) solution sufficient to produce a pH of about 8. **Ex. 1041**, at 158, col. 2. The authors assume that this co-precipitates  $\text{Al}(\text{OH})_3$  along with  $\text{Co}(\text{OH})_2$  and  $\text{Ni}(\text{OH})_2$ , respectively. See **Ex. 1041**, 160. Because the cobalt and nickel aluminates (as well as alumina) were prepared from precipitation reactions, calcination is required in order to form the crystalline aluminate materials, similar to the procedure described in *Komarewsky*. Compare **Ex. 1041**, p. 158, col. 2 with **Ex. 1029**, p. 557, col. 2. The co-precipitates were first dried at 100 °C for 20 hours, and then calcined in air for 4 hours at temperatures of 200 °C, 400 °C, 500 °C, 700 °C, and 900 °C. **Ex. 1041**, 158, col. 2; 160 (Tables 1-2). Alumina was also prepared in an analogous manner, followed by calcination at 400 °C or 900 °C. **Ex. 1041**, 158, col. 2. Despite Dr. Taber's testimony that calcination was performed on the catalysts of *Chokkaram* "after treatment," none of the catalysts in *Chokkaram* were chemically modified or treated. **Ex. 2007**, p. 61, l. 2 – p. 63, l. 11.

47. As discussed in *Ross*, the calcination of alumina and other aluminates is performed in order to dehydrate an aluminum hydroxide precipitate and form alumina, and it is well-known that the conditions for aging the precipitates, as well

as the conditions and times for drying and calcination can be crucial in determining the structure of the resultant alumina. See **Ex. 2002**, pp. 72-72. A POSITA reviewing *Chokkaram*, similar to *Ross*, would understand that a wide range of temperatures can be used to calcine precipitates in order to prepare alumina and aluminates. Compare **Ex. 1041**, p. 160 (disclosing calcination temperatures of 473 K, 673 K, 773 K, 973 K, and 1173 K) with **Ex. 2002**, p. 73 (disclosing calcination temperatures of 453 K, 503 K, 523 K, 723 K, 873 K, 1123 K, 1173 K, and 1473 K). Thus, a POSITA would not have looked to *Chokkaram* to develop an understanding nor expectations around calcining a calcium-promoted  $\gamma$ -alumina catalyst.

48. The effect of calcination temperature on the resulting crystallinity, surface area, pore volume, and catalytic activity for the conversion of 2-octanol to octenes using the alumina, nickel-aluminate, and cobalt-aluminate was examined. The data for alumina showed that alumina calcined at 400 °C and 900 °C provided a maximum conversion to octenes of 97.4 mol-% and 89.7 mol-%, respectively, at a reactor temperature of 280 °C. See **Ex. 1041**, 166 (Tables 3-4). *Chokkaram* teaches that:

The selectivity for dehydrogenation, dehydration, and isomerization depend on the type of catalyst, LHSV, catalyst calcination temperature and the reaction temperature. The selectivities for l-octene, cis-2-octene, and trans-2-octene among the octenes (Tables 3-6) depend

mostly on the type of catalyst, LHSV and reaction temperature.

**Ex. 1041**, p. 164, col. 2. Thus, *Chokkaram* specifically teaches that calcination temperature can affect the selectivity for the type of reaction that is catalyzed—i.e., dehydration vs. dehydrogenation vs. isomerization. Dr. Taber agreed that *Chokkaram* teaches that selectivity can actually improve with calcination temperature. **Ex. 1041**, p. 167, col. 2; **Ex. 2007**, p. 112, l. 10 – p. 113, l. 7. Further, *Chokkaram* states that the selectivity for dehydration over alumina is close to 100% regardless of the calcination temperature. **Ex. 1041**, p. 167, col. 2; **Ex. 2007**, p. 111, l. 13 – p. 112, l. 5.

49. Lastly, it is important to note that 2-octanol is so very much NOT 1-octanol, as its chemical reactivity, regioselectivity upon dehydration, and impurities formed in the dehydration for 2-octanol are unique, and have no relation to the dehydration of 1-octanol to 1-octene.

**C. *Le* (Ex. 1007) and *De Bruyn* (Ex. 1008)**

50. I discussed *Le* (**Ex. 1007**) and *De Bruyn* (**Ex. 1008**) at length in my First Declaration, and provide the following additional details.

51. As stated in my First Declaration, a POSITA reading *Le* and *De Bruyn* would not know whether any of the catalysts used by *Le* or *De Bruyn* had been chemically modified or calcined. **Ex. 2001**, ¶40, ¶55, ¶64; *see also* **Ex. 2005** at p. 97, l. 21 – p. 99, l. 20; p. 101, ll. 6-20; **Ex. 2007**, p. 33, ll. 7-11; p. 125, l. 2 – p.

127, l. 20. However, Dr. Taber more recently affirmatively stated—without any evidence—that a POSITA would know that the commercial catalyst Sasol uses is the product of treatment with calcium acetate followed by calcination. **Ex. 2007**, p. 32, ll. 14-19; p. 34, ll. 1-7.

52. However, *Le* merely discloses that catalysts for dehydration can be acidic or basic (**Ex. 1007** at ¶ [0063]), and that a barium-doped  $\gamma$ -alumina catalyst (an exemplary basic catalyst) “is used to further increase the selectivity of the dehydration” whereas “an acidic catalyst is used to improve the reaction yield” (**Ex. 1007** at ¶ [0064]). In my opinion, a POSITA would not read these portions of *Le* as specifically teaching the utilization of a barium-doped  $\gamma$ -alumina catalyst to obtain a 1-alkene regioselectivity of greater than 95%. Instead, a POSITA would view *Le* as specifically teaching the use of a commercially available undoped  $\gamma$ -alumina catalyst available from “Eurosupport under the tradename ESM110®.” *Le*, ¶¶ [0064], [0171]. In my opinion, a POSITA simply “listing” an optional chemical treatment as possible does not teach or enable a POSITA how to prepare a catalyst, let alone with any reasonable expectation about how it will perform and/or for how long. As is seen in prior art such as *Wu, Wright* ’788, *Wright* ’451, for example—the details in catalyst preparation matter particularly for producing high conversion, high regioselectivity, and time-on-stream.

53. Further, I disagree that ¶¶ [0012], [0021], or [0184] of *Le* (cited by Petitioners, Dr. Taber, and the PTAB) would inform a POSITA that a 1-alkene regioselectivity of greater than 95% had been achieved. To be clear, nothing in these sections of *Le* specifically discloses 1-alkene regioselectivity (or conversion). Nor would a POSITA reviewing these sections of *Le* reasonably infer that a 1-alkene regioselectivity of greater than 95% had been achieved. For example, *Le* ¶ [0012] merely states the well-known fact that separating alkene regioisomers (for example, by distillation) is difficult, and therefore, a process capable of producing “predominantly one olefin with a very limited number of isomers has great advantages.” Nothing in ¶ [0012] of *Le* states that a 1-alkene regioselectivity greater than 95% had actually been achieved, let alone the conversion provided by such process. Similarly, *Le* ¶ [0021] states that “the process for producing this polymer is facilitated in comparison with the conventional processes using olefins derived from oil products since the olefin (a) has fewer isomers,” but again plainly does not state that a 1-alkene regioselectivity of greater than 95% had actually been achieved, let alone the conversion provided by such process. Finally, a POSITA would view *Le*’s disclosure at ¶ [0184] of the molar amounts of 1-heptene used in Ziegler-Natta polymerization reactions as unconnected to the 1-heptene regioselectivity obtained by the earlier catalytic dehydration step because *Le* specifically discloses that following dehydration, “a step of purifying the 1-heptene

is performed before carrying out the polymerization step C of the process.” *Le*, ¶ [0173]. Thus, despite *Le*’s disclosure at ¶ [0012] that a process capable of producing predominantly one olefin would have great advantages (such as eliminating the need to purify the 1-olefin), a POSITA would understand that *Le*’s sole working example nonetheless includes an additional step to purify the 1-heptene prior to polymerization. Therefore, a POSITA reading *Le* would not understand that a 1-alkene regioselectivity of greater than 95% had been obtained by *Le*. **Ex. 2005**, p. 104, l. 14 – p. 105, l. 1; p. 106, l. 14 – p. 107, l. 9;

54. Turning to *De Bruyn*, Example 3 of *De Bruyn* discloses a catalytic process whereby 1-hexene is produced with a regioselectivity of greater than 92% and a conversion of at least 95%. See **Ex. 1038** at ¶ 47-48; **Paper 16** at 13. Based on my further review of *De Bruyn*’s Example 3, Example 4 (Step 5), and Example 5 (Step 4), along with the data disclosed in **Ex. 1027** (p. 147, Fig. 2), in my opinion a POSITA reviewing these documents would understand that *De Bruyn* discloses a 1-alcohol conversion of about 75% with a 1-alkene regioselectivity of about 95% resulting from a single pass of the relevant reactants over the catalyst. Indeed, Dr. Taber admits that *De Bruyn*’s Example 3 only obtains a conversion of 95% or greater by recycling the unreacted 1-alcohol and dihexyl ether (and impurities). **Ex. 1038** at ¶ 48 (citing **Ex. 1008** at [0053]); **Ex. 2007**, p. 127, l. 21 – p. 128, l. 13.

55. Further, a POSITA reviewing *De Bruyn* would understand that promoted  $\gamma$ -alumina catalysts were known—for example,  $\text{CaO} \cdot \text{Al}_2\text{O}_3$ ,  $\text{Ca}_2\text{O}_3 \cdot \text{Al}_2\text{O}_3$ —but nothing in *De Bruyn* teaches or suggests which catalyst is preferred or calcination at the specific temperature range of about 420 °C to about 480 °C (**Ex. 1008**, ¶ [0037].) Thus, *De Bruyn* provides no teaching or suggestion as to which catalyst is preferred, and whether the preferred catalyst would be chemically modified or how it was (or was not) calcined.

56. In sum, a POSITA would have understood *Le* and *De Bruyn* as disclosing processes to dehydrate  $\text{C}_6$  to  $\text{C}_8$  1-alkanols using commercially available  $\gamma$ -alumina catalysts with a reasonable expectation using a single pass through the reactor to obtain an alcohol conversion efficiency in the range of 59% to 77%, a 1-alkene regioselectivity in the range of 95% to 98%, and chemical yield of 1-alkene at 20% to 35%. (See, e.g., **Ex. 1008** at p. 11, Fig. 2 (describing percentage conversion, yield of octenes and purity versus TOS).) Importantly, the only discussion of catalyst activity as a function of TOS in *Le* and *De Bruyn* shows a catalyst that performed poorly at even a modest amount of TOS. (See **Ex. 1008**, ¶ [0081], Figure 2, Table 14.)

**D. Wright '788 (Ex. 1009)**

57. As noted in my First Declaration, I am the same Michael E. Wright who is the sole named inventor of *Wright '788* (**Ex. 1009**).

58. *Wright* '788 discloses methods to make 1-butene from 1-butanol with 1-butene regioselectivity of at least 92% using a  $\gamma$ -alumina catalyst that is 1) treated with an aqueous base solution, 2) washed with water, 3) dried, and then 3) reacted with an organosilane, 4) washed with a hydrocarbon solvent, and 5) air-dried. See **Ex. 1009**, ¶ [0020]. *Wright* '788 uses this specific  $\gamma$ -alumina catalyst to produce bio-1-butene from bio-1-butanol with a conversion of 96% to 98% and a 1-butene regioselectivity of 94% to 95%. See **Ex. 1009**, ¶ [0034], Table 1.

59. Dr. Taber argues that a POSITA reading *Wright* '788 would understand that the inventive process of *Wright* '788 does not require a silanized catalyst, as there are embodiments that do not undergo silanization. **Ex. 1038**, ¶55. Dr. Taber testified that he is referring to *Wright* '788, ¶ [002], but that *Wright* '788 provides no working examples of an unsilanized alumina catalyst. **Ex. 2007**, p. 135, l. 20 – p. 136, l. 20; **Ex. 1009**, ¶ [0034], Table 1. Indeed, *Wright* '788 states that “two runs were carried out according to the process of the invention” and both utilized a silanized alumina catalyst. **Ex. 1009**, ¶ [0034]. Thus, a POSITA would not view these results as extending generally to any base-treated  $\gamma$ -alumina catalyst, particularly one that had not been silanized. What is more, the lack of working examples showing conversion, regioselectivity, or time-on-stream for such unsilanized, base-treated  $\gamma$ -alumina catalyst would have led a POSITA

reviewing *Wright* '788 to pursue silanization as a strategy to improve these outcomes.

60. I also note that inventors and patent drafters regularly disclose embodiments that are in the prior art, and a POSITA would understand that such disclosures do not necessarily mean that an inventive process can be practiced using all of the embodiments. For example, *Wu* (**Ex. 1025**, discussed above) teaches the inventive concept that base-treatment of a  $\gamma$ -alumina catalyst can provide increased 1-alkene regioselectivity by neutralizing acidic sites on the alumina. **Ex. 1025**, col. 1:21-26. Nonetheless, *Wu* also discloses that:

In one of its concepts the invention provides a process for the dehydration of an alcohol [sic] in the presence of an inert carrier gas with gamma-alumina.

**Ex. 1025**, col. 1:18-21. A POSITA reading *Wu* would understand that an unmodified  $\gamma$ -alumina catalyst was not what *Wu* invented and would not assume that an unmodified  $\gamma$ -alumina catalyst could be used to obtain the same regioselectivity as a base-treated  $\gamma$ -alumina catalyst provided by *Wu*'s inventive process. Similarly, more than 30 years after *Wu*'s publication, *Le* disclosed that:

This dehydration step can be carried out in the presence of a dehydration catalyst, for example a  $\gamma$ -alumina-based catalyst, which may be acidic or basic, .... [and] According to a first version of the invention, a basic catalyst, for example a barium-doped  $\gamma$ -alumina, is used to further increase the selectivity of the dehydration.

**Ex. 1007, ¶ [0062]-[0063].** Here again, a POSITA would view this disclosure as reiterating the general understanding that base-treatment could increase alkene selectivity. Further, as discussed above, a POSITA would not understand that *Le* had actually used a barium-doped  $\gamma$ -alumina catalyst, as such experiments are not disclosed by *Le*.

61. Turning back to *Wright* '788, taken as a whole, a POSITA would understand *Wright* '788's general disclosure of "embodiments" using an alumina-based catalyst "specifically washed with base solutions," as re-stating the generally known concept in the art, as taught by *Wu* and restated by *Le*, that base-treatment was known to improve regioselectivity. However, similar to the disclosures in *Wu* and *Le*, a POSITA would not view *Wright* '788 as disclosing the use of a base-treated, but unsilanized  $\gamma$ -alumina catalyst to obtain the exceptional regioselectivity, conversion, and time-on-stream taught by *Wright* '788 using silanized  $\gamma$ -alumina catalyst.

62. Moreover, based on *Wright* '788 ¶ [0022], a POSITA would have no expectation that a calcium-promoted but unsilanized alumina catalyst could provide an alkene conversion of 95% or greater with a 1-alkene regioselectivity of 95% to about 99%. Rather, based on the unpredictability in this field, a POSITA reading *Wright* '788 would focus on its examples for guidance. In doing so, a POSITA would understand that *Wright* '788 relies upon the activity and

regioselectivity of a silanized alumina catalyst to achieve high alkene conversion and high 1-alkene regioselectivity for extended time-on-stream. For these reasons, I strongly disagree with Petitioners and Dr. Taber's argument that a POSITA reading *Wright* '788 would be motivated to prepare a catalyst without silanization and have any expectation that it would provide an alkene conversion of 95% or greater and a 1-alkene regioselectivity of 92% or greater after time-on-stream of more than 20 hours. This is because *Wright* '788 specifically targeted and solved the issue of short time-on-stream stability for  $\gamma$ -alumina catalysts dehydrating bio-1-butanol by using a silanization step in the catalyst preparation.

63. Petitioners and Dr. Taber also attempt to cabin the example in *Wright* '788 using a silanized catalyst as somehow limited to water-containing feedstocks. **Ex. 1038**, ¶55. I disagree. The dehydration of 1-alcohols to 1-alkenes necessarily involves the production of equimolar quantities of water, as each alcohol molecule reacts to form one molecule of water and one alkene. Thus, even anhydrous 1-hexanol or 1-octanol starting material will result in the generation of an effluent stream containing about 12 wt-% to 17 wt-% water. A POSITA would also understand that most bio-1-alcohols contain at least some amount of water. Further, a POSITA reading *Wright* '788 would understand that the only disclosure in *Wright* '788 that enables a POSITA to prepare 1-alkenes with the disclosed regioselectivity and high conversion for extended time-on-stream required a

silanized alumina catalyst. And, to the extent *Wright '788* would have motivated a POSITA to utilize an unsilanized  $\gamma$ -alumina catalyst, in doing so a POSITA would have looked to other prior art disclosures such as *Le* (**Ex. 1007**), *De Bruyn* (**Ex. 1008**) and/or *Makgoba* (**Ex. 1027**) for guidance, as well as for a reasonable expectation of the resulting conversion, 1-alkene regioselectivity, and time-on-stream.

64. Finally, *Wright '788* also discloses that the inventive silanized  $\gamma$ -alumina catalyst can be used for the dehydration of C<sub>5</sub> to C<sub>23</sub> 1-alkanols (**Ex. 1009**, ¶ [0017], ¶ [0022]), but the examples of *Wright '788* are limited to dehydration of 1-butanol to 1-butene.

**E. *Wright '451* (Ex. 1039)**

65. Int'l Patent Pub. No. WO 2015/130451 ("*Wright '451*," **Ex. 1039**) is titled "Regioselective Dehydration of Terminal Alcohols," and was published on September 3, 2015. I am the same Michael E. Wright who is one of the named inventors of *Wright '451*.

66. Similar to *Wright '788*, *Wright '451* discloses the methods of preparing bio-1-alkenes from bio-1-alcohols, and in particular bio-1-butene from bio-1-butanol, by catalytic dehydration with a 1-alkene regioselectivity of greater than about 90%, about 90% to about 99%, or greater than about 95% using a  $\gamma$ -

alumina catalyst that is 1) treated with a calcium promoter, 2) calcined, and 3) optionally reacted with an organosilane, washed, and dried. **Ex. 1039**, ¶[18], ¶[22].

67. *Wright '451* discloses the use of promoted  $\gamma$ -alumina catalysts that are prepared by modifying a  $\gamma$ -alumina catalyst with a  $\text{Ca}^{+2}$  compound. **Ex. 1039** at ¶[14]. “In some embodiments  $\text{Ca}^{+2}$  is the modifier and the  $\text{Ca}^{+2}$  compound is calcium acetate ( $\text{Ca}(\text{OAc})_2$ ).” **Ex. 1039** at ¶[14]. According to *Wright '451*, “Calcium imparts an increase in reaction rate when compared to Group 1 metals such as sodium and potassium when used at similar wt% loading values. Furthermore, calcium provides an increase in selectivity for the terminal alkene product over use of Group 1 metals at high and comparable terminal olefin conversion.” **Ex. 1039** at ¶[14].

68. *Wright '451* also discloses that:

Promoter (e.g. Group 2 metal) modified and heat-treated (e.g., calcined) catalysts can be further modified by treatment with at least one organosilane to create new catalysts that are especially useful for the dehydration of 1-alcohols mixed with high wt% of steam. The promoter-organosilane-modified alumina catalysts afford high regioselectivity in forming 1-alkene and can perform for extended periods of time on stream with little loss in regioselectivity.

**Ex. 1039** at ¶[15]. Thus, *Wright '451* teaches that silanized  $\gamma$ -alumina catalysts provided longer time-on-stream.

69. After modification with a  $\text{Ca}^{+2}$  compound, *Wright '451* discloses that the modified  $\gamma$ -alumina catalysts

[C]an be further heated ... to provide drying or calcination of the catalyst. The drying or calcination can be carried out in vessel **203** or **205** at temperatures of about 100 °C to about 900 °C, or about 200 °C to 600 °C for a time period of about 1 h to about 8 h, or about 2 h to 6 h.

**Ex. 1039** at ¶[67]. Further, *Wright '451* discloses Examples 1-26 in which drying and/or calcination of various catalysts is performed at 120 °C, 200 °C, 500 °C, 550 °C, and/or 600 °C for various amounts of time. **Ex. 1039** at pp. 24-33 (Examples 2-4, 8-24).

70. *Wright '451* discloses that the dehydration methods are generally applicable to 1-alcohols of general formula  $\text{R}^1\text{R}^2\text{CH}(\text{CH}_2)_2\text{OH}$ , where  $\text{R}^1$  and  $\text{R}^2$  are independently H or a  $\text{C}_1$  to  $\text{C}_{20}$  alkyl radical or contain one or more aromatic rings containing exclusively carbon or a mixture of carbon and heteroatoms, or a nitrile, alkene, or ketone functional group, or a combination thereof. **Ex. 1039** at ¶[21]. However, I note that all of the exemplary embodiments of *Wright '451* relate to catalytic dehydration of 1-butanol to form 1-butene. See **Ex. 1039** at 24-33.

71. *Wright '451* also discloses that the modified  $\gamma$ -alumina catalysts can be silanized. **Ex. 1039**, ¶[68]-¶[72]. However, unlike *Wright '788*, *Wright '451* discloses examples in which the modified  $\gamma$ -alumina catalysts are both unsilanized

or silanized. See **Ex. 1039** at 24-33 (Examples 4, 7, and 20-22 utilize silanized catalysts).

72. *Wright '451* also focuses on maintaining high conversion and high 1-butene selectivity for extended time-on-stream. In order to simulate extended periods of time-on-stream, *Wright '451* performed experiments in which the exemplary catalysts were: 1) tested immediately after preparation; 2) tested after heating for 60 hours at 400 °C under a continuous feed of 93 wt-% water and 7 wt-% 1-butanol; and 3) after a second 60 hours at 400 °C under a continuous feed of 93 wt-% water and 7 wt-% 1-butanol. Calcium-promoted  $\gamma$ -alumina catalysts calcined at 550 °C for 6 hours (i.e., Examples 18 and 19) maintained or even improved conversion efficiency after aging but exhibited decreased 1-butene regioselectivity. *Wright '451*, p. 31, Table 2. A calcium-promoted  $\gamma$ -alumina catalyst calcined at 500 °C and silanized (Examples 24 and 26) showed butanol conversion above 95% and 1-butene regioselectivity of 94% to 95% after almost 20 days of time-on-stream.

73. Dr. Taber repeatedly pointed to the presence of heptane in the dehydration of bio-1-butanol by *Wright '451* as providing further reason a POSITA would have relied on *Wright '451* as more relevant prior art than *Wright '788* because “the POSITA would know that adding the heptane to the 1-butanol would drive the water out of the 1-butanol.” **Ex. 2007**, p. 141, l. 19 – p. 143, l. 9; p. 145, l.

15 – 147, l. 10; p. 150, ll. 9-14. I disagree. The addition of 2 wt-% heptane in *Wright '451* had no impact whatsoever on the miscibility of water in bio-1-butanol, as the heptane was completely miscible with the reactant solution of 88 wt% 1-butanol, 2 wt% heptane, and 10 wt% water. See **Ex. 1039**, ¶¶ [0042], [0043], [0045], [0046]. What is more, as noted above, during the dehydration reaction, the reactants are entirely in the vapor-phase as they pass through the reactor. A POSITA would understand that any  $\gamma$ -alumina catalyst, whether modified, base-treated, calcium-promoted, silanized, or treated in some other way, will cause no reaction of the heptane, and therefore heptane was present solely as an internal reference standard in order to better quantify the amount of 1-butene formed by the dehydration reactions using gas chromatograph (GC). Heptane is particularly useful for this because at 2 wt-% it is entirely miscible in the bio-1-butanol/water reagent mixture, and the boiling point of heptane (98 °C) is similar to that of water (100 °C) and 1-butanol (117 °C). This would have been readily apparent to a POSITA reviewing *Wright '451* and *Wright '788*, and would have had no impact on a POSITA's view of either reference being more “relevant” than the other.

74. Of note, *Wright '451* includes Comparative Example 6, which is a catalyst prepared according to the procedure of *Wu (Ex. 1025)*, wherein:

Commercially available Strem gamma-alumina powder was placed in contact with an aqueous sodium hydroxide solution (2x the wt of the gamma-alumina, 5 wt% NaOH) for 2 h, isolated by filtration, and washed several times

with water (total 4x the wt of gamma-alumina). Comparative Catalyst 6 was dried at 200 °C for a period of 24 h and then used as a dehydration catalyst (see below).

**Ex. 1039**, ¶[98]. In Comparative Example 7, the same procedure as Example 6 was used, but the catalyst was further “treated with dipenyldichlorosilane (3 g/L) and triethylamine (3 g/L) dissolved in a hexanes (total 1 L) solution. This material was washed with hexanes (1 L), then water (2 L), and dried at 200 °C for a period of 24 h.” **Ex. 1039**, ¶[99].

75. The regioselectivity and conversion provided by catalysts prepared in Examples 6 and 7 were compared to the inventive catalyst of *Wright '451*, the results of which are shown in Fig. 9, which:

[I]s a graph showing the performance (i.e. 1-butanol/dibutyl ether conversion vs. 1-butene regioselectivity) of Catalyst 8 and comparative Catalysts 6 and 7 using a feed of 88 wt% 1-butanol, 2 wt% heptane, and 10 wt% water. Run #1 is fresh catalyst, prior to Run #2 the catalyst was aged by heating 60 hours at 400 °C with a continuous feed of 93 wt% water and 7 wt% 1-butanol, and prior to Run #3 an additional 60 h (i.e. total 120 hours) of aging at 400 °C under a continuous feed of 93 wt% water and 7 wt% 1-butanol. For each run the performance of Catalyst 8 was evaluated at four temperatures: 340 °C, 360 °C, 380 °C, and 400 °C. The performance of comparative Catalyst 6 was evaluated at three temperatures: 360 °C, 380 °C, and 400 °C for each run. Comparative Catalyst 7 was evaluated at four temperatures: 340 °C, 360 °C, 380 °C, and 400 °C in Run #1 and Run 2, and at three temperatures 360 °C, 380 °C, and 400 °C in Run #3. The lowest 1-butanol conversion is always at the lowest temperature and conversion increases with temperature.

**Ex. 1039**, ¶[42]. The results with *Wu*'s catalyst (Example 6) show a conversion of about 64%, 89.4%, and about 99% with 1-butene regioselectivity of about 97%, 95.9%, and about 94%, respectively, for freshly prepared catalyst. **Ex. 1039**, Fig. 9, Table 2. However, after 60 hours and 120 hours of aging (as described above), the conversion dropped below 80% at all temperatures and the maximum regioselectivity was less than 95%. However, when silanized as in Example 7, *Wu*'s catalyst provided significantly better resistance to aging.

76. Example 8, also included in Fig. 9, shows that a calcium-promoted  $\gamma$ -alumina catalyst provides significantly better performance compared to a catalyst prepared with NaOH base. **Ex. 1039**, Fig. 9. Thus, a POSITA reading *Wright '451* would understand that a higher conversion and regioselectivity could be obtained by treating a  $\gamma$ -alumina catalyst with a calcium promoter followed by calcination compared to treatment with an alkali metal hydroxide. Further, based on *Wright '451*, a POSITA would not select *Wu*'s catalyst for use in the catalytic dehydration of 1-alcohols to form 1-alkenes with high regioselectivity and conversion.

77. In my opinion, a POSITA would view *Wright '451* and *Wright '788* as the closest prior art to the substitute claims. This is because *Wright '451* and *Wright '788* discloses catalysts that provide 1) high 1-alkene regioselectivity, 2) constant high alcohol conversion and yield, as well as selectivity for 1-alkenes, and

3) exceptional catalytic activity and regioselectivity after an extended period of time-on-stream.

**VI. REVISED SUBSTITUTE CLAIMS 29-44 ARE DESCRIBED IN THE SPECIFICATION AND ARE PATENTABLE OVER THE PRIOR ART**

78. Independent substitute claim 29 requires the following:

29. A process for making a bio-alkene, comprising:

dehydrating a bio-1-alcohol by heating the bio-1-alcohol in a continuous flow reactor at a temperature of 240 °C to 360 °C with a calcium-promoted  $\gamma$ -alumina catalyst calcined at a temperature of about 420 °C to about 480 °C, wherein the catalyst has not been silanized, and

wherein the resulting bio-alkene has a carbon chain of 6 to 10 carbon atoms and is produced in a conversion of at least 95% and a bio-1-alkene regioselectivity of 95% to about 99% from a single-pass reaction after at least 20 hours time-on-stream.

(Patent Owner's Revised Non-Contingent Motion to Amend, Appendix A.)

Notably, substitute claim 29 is a method for making a bio-alkene that requires a specific  $\gamma$ -alumina catalyst, which is calcium-promoted then calcined at a temperature of about 420 °C to about 480 °C, but not silanized. The claimed catalyst dehydrates C<sub>6</sub> to C<sub>10</sub> bio-1-alcohols with both high bio-1-alkene regioselectivity of 95% to about 99%, and with a high conversion of at least 95% with a single pass, even after a time-on-stream of at least 20 hours.

**A. Revised Substitute Claims 30-44 Are Supported by the Written Description**

79. Petitioners and Dr. Taber argue that previously presented substitute claims 29-44 fail the written description requirement based on their use of the terms “base treated” and “base-treated and calcined.” **Ex. 1038** at 14-23. I note that both of the terms “base treated” and “base-treated and calcined” are not recited in the revised substitute claims. Thus, Ground I in Petitioners’ Opposition to Inolex’s Non-Contingent Motion to Amend is rendered moot.

80. The revised substitute claims also find written support in the specification. Revised substitute claim 29 requires “a conversion of at least 95% and a bio-1-alkene regioselectivity of 95% to about 99% from a single-pass reaction after at least 20 hours time-on-stream.” As discussed above, time-on-stream (or “TOS”) is a very important factor in developing a commercially viable catalyst for conducting dehydrations. Examples 5, 11, and 12, and Fig. 7 of the ’048 patent disclose that time-on-stream periods of nearly 2 months (i.e., >1,400 h) were obtained for dehydration processes using the calcium-promoted catalyst. See **Ex. 1001** at col. 12, ll. 33-34; col. 27, ll. 20-35; col. 29, ll. 1-35, Fig. 7. The ’048 patent also discusses prior art methods in which conversion and regioselectivity were lost after a mere 20 h time-on-stream. **Ex. 1001**, col. 6, ll. 33-47. Thus, a POSITA reviewing the specification of the ’048 patent would understand that the inventors were in possession of a process that provided the claimed conversion and

regioselectivity “after at least 20 hours time-on-stream” as required by substitute claim 29.

81. In my opinion substitute claims 35, 38, 41, and 44, which require that the reactor is heated at a temperature of 315 °C to 350 °C, are also supported by the specification of the '048 patent, specifically Examples 3, 4, 5, and 11.

Examples 3-5 show that the claimed regioselectivity and conversion were obtained using calcium-promoted catalysts at reactor temperatures of 315 °C, while

Example 11 shows the same for reactor temperatures of 330 °C and 350 °C. **Ex.**

**1001**, col. 26, l. 52 – col. 27, l. 35; col. 29, ll. 1-26. Thus, a POSITA reviewing Examples 3-5 and 11 of the '048 patent would understand that the inventors were in possession of a method of using the claimed calcium-promoted catalyst to obtain the specified conversion and regioselectivity at a temperature range of 315 °C to 350 °C.

82. In sum, a POSITA reading the specification of the '048 patent in its entirety would understand that the inventors were in possession of the claimed methods of making a bio-alkene.

**B. Substitute Claim 29 is Patentable Over *Le* and *De Bruyn***

83. As discussed above, *Le* and *De Bruyn* disclose the dehydration of 1-alcohols using commercially available  $\gamma$ -alumina catalysts. According to Dr. Taber, a POSITA would have no reason to review the literature or attempt to

design new  $\gamma$ -alumina catalysts if catalysts such as those used in *Le* and/or *De Bruyn* were available. **Ex. 2007**, p. 17, ll. 1-12. While a POSITA would know there are numerous sources of commercially available  $\gamma$ -alumina catalysts, Dr. Taber testified that he did not know what catalysts are available commercially. **Ex. 2007**, p. 17, l. 13-20. Dr. Taber nonetheless testified that a POSITA would simply use one of the commercially available  $\gamma$ -alumina catalysts disclosed by *Le* or *De Bruyn*—for example ESM 110<sup>®</sup> catalyst from Eurosupport or the Pural<sup>®</sup> KR1 catalyst from Sasol. **Ex. 2001**, ¶ 56; **Ex. 2005** at p. 101, l. 21 – p. 103, l. 15; **Ex. 2007**, p. 17, ll. 1-12; p. 33, ll. 3-6; p. 126, l. 20 – p. 127, l. 10; p. 150, l. 15 – p. 151, l. 2. However, a POSITA would not know anything about how those commercially available catalysts were prepared—for example, whether commercial alumina catalysts disclosed in *Le* or *De Bruyn* had been chemically modified or calcined. See **Ex. 2001**, ¶ 40, ¶ 55, ¶ 64; **Ex. 2005** at p. 97, l. 21 – p. 99, l. 20; p. 101, ll. 6-20; **Ex. 2007**, p. 33, ll. 7-11; p. 125, l. 2 – p. 127, l. 20. And even if a POSITA were to infer that the catalysts used in *Le* or *De Bruyn* had been calcium promoted (which *De Bruyn* teaches as being a possibility), a POSITA would expect that such calcium-promoted catalysts would be limited to conversion of 59% to 77% with a single-pass reaction (as discussed in further detail below).

84. In his Second Declaration, Dr. Taber appears to agree that *Le* is silent as to the specific isomeric purity of the 1-heptene that is produced by *Le*'s

dehydration process. **Ex. 1038**, ¶46 (“the regioselectivity of greater than 95% 1-heptene, in the disclosed example, is further supported by the products of the polymerization reaction.”) Indeed, if *Le* actually disclosed the regioselectivity of the 1-heptene that was produced, then there would be no need for any “further support” or inference to be made. Indeed, when asked to specifically identify any disclosure in *Le* that described the regioselectivity of an alkene product produced by dehydration, Dr. Taber could only point to **Ex. 1007** at ¶¶ [0100], [0105], and [0109]. See **Ex. 2007**, p. 132, l. 5 – p. 133, l. 11. Based on my review of *Le*, not only do these sections of *Le* not disclose regioselectivity, a POSITA would not understand that these sections indicate experiments that had even been performed.

85. Dr. Taber also asserts that:

*Le* provides the polymerization reaction data to the POSITA, which allows the POSITA to determine inputs to the reaction. For example, [0184] of *Le* provides the composition of the inlet to the polymerization reactor after separation of the polymer products and recycling the non-reacted products. Based on Dr. Wright’s statement of how this polymerization would proceed, if there was a measurable amount of any 2- and/or 3-heptene in the mixture, it would be listed in the Table. However, even though it measures hydrogen at the ppm concentration, there is no 2-heptene listed. *Le* at [0184].

**Ex. 1038**, ¶46. I disagree. Immediately above this section, *Le* discloses that “A step of purifying the 1-heptene is performed before carrying out the polymerization step C of the process.” **Ex. 1007**, ¶ [0173]. Based on the boiling point of 1-heptene

being 93.6 °C, while the boiling points of *cis*-2-heptene and *trans*-2-heptene are 98-99 °C, a POSITA reviewing *Le* would understand that the 2-heptenes present in the product of the catalytic dehydration reaction process of *Le* (**Ex. 1007**, ¶ [0171]) would be removed by the distillation step utilized by *Le* (**Ex. 1007**, ¶¶ [0067], [0173]). Indeed, a POSITA reviewing *Le* in view of *De Bruyn* would reasonably expect that reaction of 1-heptanol as described by *Le* would proceed similarly to the dehydration of 1-hexanol and 1-octanol described by *De Bruyn*. See **Ex. 1008**, ¶¶ [0067]-[0068], Table 10, ¶¶ [0080]-[0082], Figure 2, Table 14. What is more, *De Bruyn* (like *Le*) employs a distillation step to remove unwanted impurities. **Ex. 1008**, ¶¶ [0069]-[0070], Table 11. Thus, contrary to Dr. Taber's assertion that ¶ [0184] and the following Table in *Le* suggest that 1-heptene was produced with no detectable amounts of 2- or 3-heptenes, a POSITA would recognize that the absence of 2- and 3-heptenes was due to a distillation step that was required in order to purify the product. In sum, nothing in *Le* explicitly or implicitly supports Dr. Taber's conclusion that *Le*'s process provided a 1-heptene regioselectivity of greater than 95%.

86. As discussed above, a POSITA would understand that *De Bruyn* discloses a  $\gamma$ -alumina catalyst to dehydrate 1-hexanol and 1-octanol with a 1-alkene regioselectivity of 96% to 98% and at a conversion efficiency of 59% to 77% after a single-pass reaction. See **Ex. 1008**, ¶ [0053], ¶ [0067]-¶ [0068],

Table 10, ¶ [0080]-¶ [0083], Figure 2, Table 14. That is, *De Bruyn* compensates for the low conversion provided by the commercially available catalyst by isolating the 1-alcohol from the effluent stream and recycling it back into the reactor. See **Ex. 1008**, ¶ [0053] (Example 3); **Ex. 2007**, p. 127, l. 21 – p. 128, l. 9. Recycling unreacted starting materials is commonly done in large-scale dehydration of alcohols, particularly on a large scale and/or where energy is inexpensive. **Ex. 2007**, p. 73, ll. 11-22; p. 127, l. 21 – p. 128, l. 9. Recycling reagents utilizes significantly more energy than a process capable of at least 95% conversion from a single-pass reaction. The fact that *De Bruyn* has a limited conversion efficiency of 59% to 77% after a single-pass reaction also speaks to the need in the art to develop processes and catalysts capable of much higher single-pass conversion, such as those described in the '048 patent.

87. When *Le* is read in view of *De Bruyn*, based on the catalysts disclosed in *Le*, a POSITA would reasonably expect similar results to those disclosed in *De Bruyn*. That is, *Le* only mentions commercially available  $\gamma$ -alumina catalysts (i.e., EuroSupport ESM110<sup>®</sup>, ¶¶ [0064] and [0171] and Pural<sup>®</sup> KR1, ¶[0066]). Dr. Taber nonetheless argues that *Le*'s passing mention of a barium-doped  $\gamma$ -alumina catalyst (at **Ex. 1007**, ¶ [0064]) would have motivated a POSITA to modify a commercially available  $\gamma$ -alumina catalyst such as ESM110. I disagree. First, if Dr. Taber is correct that a POSITA would read *Le* as already providing a 1-alkene

regioselectivity of greater than 95%, then a POSITA would have no reason whatsoever to further modify a commercially available  $\gamma$ -alumina catalyst. Second, in the more likely scenario in which a POSITA views *Le* as disclosing a much lower 1-alkene regioselectivity (*see above*), *Le* is entirely silent as to how a barium-doped  $\gamma$ -alumina catalyst should be prepared. Indeed, there is no discussion of how to treat the alumina with barium, whether the catalyst must be chemically or thermally treated prior to or after chemical modification with barium, or how much the regioselectivity can be improved by barium modification. Nor is this a scenario in which it would be “obvious to try” modifying a commercially available  $\gamma$ -alumina catalyst with barium, as there is not a finite number of identified, predictable solutions to the problem of catalyst modification, let alone with barium.

88. *De Bruyn* does not rectify the shortcomings of *Le*. *De Bruyn* discloses three experimental examples in which 1-hexanol (Example 3 and Example 4, Step 5) or 1-octanol (Example 5, Step 4) were dehydrated using a  $\gamma$ -alumina catalyst. Example 4, Step 5 discloses that 1-hexene was prepared from 1-hexanol using a Sasol Chemie Pural KR1  $\gamma$ -alumina catalyst with regioselectivity of 96.5% at 76.5% conversion. (See **Ex. 1008**, ¶ [0067]-¶ [0068], Table 10.) Example 5, Step 4 discloses that 1-octene was prepared from 1-octanol using a Sasol Chemie Pural KR1  $\gamma$ -alumina catalyst with regioselectivity of 97% to 98% at 59% to 73% conversion. (See **Ex. 1008**, ¶ [0080]-¶ [0083], Figure 2, Table 14.) Finally,

Example 3 discloses the preparation of 1-hexene from 1-hexanol using a  $\gamma$ -alumina catalyst with a regioselectivity of 97.5%, where the conversion efficiency was improved to 94% by recycling the unreacted 1-hexanol from the effluent stream. (See **Ex. 1008**, ¶ [0053].) Thus, while *De Bruyn* Example 3 discloses an improved conversion, this is not obtained by a single-pass reaction. In sum, a POSITA reviewing *De Bruyn* would understand that *De Bruyn* achieved an alcohol conversion efficiency in the range of 59% to 77% from a single-pass reaction, a 1-alkene regioselectivity in the range of 95% to 98%, with a chemical yield around 35% for the 1-alkene.

89. Dr. Taber then argues that “*Le* in view of *De Bruyn* specifically describes a  $\gamma$ -alumina catalyst that was prepared by calcium treatment and then calcination.” **Ex. 1038**, ¶ 50. I disagree. *De Bruyn* merely states that “many different dehydration processes are known, and are accordingly not disclosed in any detail in this specification,” and also discloses that “the catalyst may comprise a gamma-alumina catalyst or a promoted alumina catalyst e.g.  $\text{CaO} \cdot \text{Al}_2\text{O}_3$ ,  $\text{Ca}_2\text{O}_3 \cdot \text{Al}_2\text{O}_3$ .” (**Ex. 1008**, ¶ [0037].) And Dr. Taber’s citation to my testimony in this case had nothing to do with *Le* or *De Bruyn* (neither of which teaches or suggests calcination or drying of a calcium promoted catalyst), but instead concerned questions about **Ex. 1039** (*Wright* ’451). And there, my testimony at **Ex. 1040**, p. 116:4-18 concerned how to know when a calcination process was

complete, while my testimony at **Ex. 1040**, p. 131:20 – 132:14 concerned the meaning of the term “calcium loading,” a term not used anywhere in *Le* or *De Bruyn*.

90. What is more, Dr. Taber provides no discussion as to why a POSITA reviewing *Le* in view of *De Bruyn* would be motivated to use a modified  $\gamma$ -alumina catalyst, such as a calcium-promoted catalyst. Indeed, the relevant experiments described by *De Bruyn* disclose a 1-alkene regioselectivity of 97.5% (Example 3), 96.5% (Example 4, Step 5), and 97% to 98% (Example 5, Step 4). See **Ex. 1008**, ¶ [0053], ¶¶ [0067]-[0068], Table 10; ¶¶ [0080]-[0083], Figure 2, Table 14. On cross examination, Dr. Taber testified that a POSITA would only look to other prior art or attempt to create a modified  $\gamma$ -alumina catalyst if he/she could not obtain a commercially available catalyst from a company such as SASOL. **Ex. 2007**, p. 16, l. 13 – p. 17, l. 12. As any POSITA working in the field of heterogeneous catalysis would know, there are numerous sources of commercially available catalysts—from SASOL as well as many other companies.

91. Further, based on the regioselectivity disclosed by *De Bruyn*, a POSITA reviewing *Le* and *De Bruyn* together would have no reason to chemically modify the commercially available  $\gamma$ -alumina catalysts used in these references with a base in order to improve 1-alkene regioselectivity. On the contrary, if anything, the low conversions provided by *De Bruyn*’s examples would have

motivated a POSITA to modify the  $\gamma$ -alumina catalysts using *Le*, which teaches that “an acidic catalyst is used to improve the reaction yield.” **Ex. 1007**, ¶ [0064]. Therefore, in my opinion the combination of *Le* in view of *De Bruyn* would have motivated a POSITA to use an acidic catalyst, because this would have had the possibility of improving the alkene conversion without the need for recycling.

92. In sum, a POSITA reviewing *Le* and *De Bruyn* would expect that the commercially available  $\gamma$ -alumina catalysts disclosed therein could be used to dehydrate a C<sub>6</sub> to C<sub>10</sub> bio-1-alcohol with an alcohol conversion efficiency in the range of 59% to 77% from a single-pass reaction, a 1-alkene regioselectivity in the range of 95% to 98%, with a chemical yield around 35% for the 1-alkene.

93. Based on *Le*’s and *De Bruyn*’s use of commercially available  $\gamma$ -alumina catalysts to obtain high regioselectivity, a POSITA reviewing *Le* and *De Bruyn* would not have been motivated to modify the catalysts with a base-treatment and/or a chemical promoter. Nor do *Le* or *De Bruyn* teach or suggest which chemical modifications and/or calcination conditions could be used to improve chemical conversion while maintaining high regioselectivity. Thus, *Le* and *De Bruyn* in view of any background knowledge in the art would not have motivated a POSITA to utilize a calcium-promoted and calcined (but not silanized)  $\gamma$ -alumina catalyst to dehydrate a C<sub>6</sub> to C<sub>10</sub> bio-1-alcohol. Indeed, nothing in *Le* or *De Bruyn* would provide a POSITA any reasonable expectation that a calcium-

promoted  $\gamma$ -alumina catalyst calcined at about 420 °C to about 480 °C could be used to dehydrate a C<sub>6</sub> to C<sub>10</sub> bio-1-alcohol, wherein the resulting bio-alkene is produced in a conversion of at least 95% and a bio-1-alkene regioselectivity of 95% to about 99% from a single-pass reaction after at least 20 hours time-on-stream.

94. Nor would the substitute claims be obvious to try over the combination of *Le* in view of *De Bruyn* because there was not a finite number of identified, predictable solutions to the problem of improving conversion while maintaining a high 1-alkene regioselectivity.

95. In sum, the combination of *Le* in view of *De Bruyn* provides no reasonable expectation that the claimed catalyst could be used with any reasonable expectation of success to provide the claimed result. For at least these reasons, in my opinion substitute claim 29 is patentable over *Le* in view of *De Bruyn*.

**C. Substitute Claim 29 is Patentable Over *Wu* in View of *Chokkaram***

96. As discussed above, *Wu* discloses the base-modification of  $\gamma$ -alumina catalysts using an alkali hydroxide base, followed by drying. For the conditions of the dehydration reaction, *Wu* teaches that “temperatures in the range of from about 200° to 500° C. are suitable for the desired dehydration reaction to occur. It is currently preferable, however, to employ temperatures in the range of 300°-450° C.” **Ex. 1025**, col. 2:51-54. While *Wu* discloses a wide range of 1-alcohols that can

be dehydrated using the base-treated catalysts, the only examples in *Wu* concern the dehydration of 3-methyl-1-butene.

97. Example 1 in *Wu* shows that dilution of the 1-alcohol starting material (3-methyl-1-butenol) with an inert carrier gas can increase the conversion of 1-alcohol to 1-alkene (in *Wu*'s case, 3-methyl-1-butene). There, a  $\gamma$ -alumina catalyst without a carrier gas provided a conversion to 3-methyl-1-butene of 78.2% and yield of 75.3%, while conversion increased to 93.4% (and yield increased to 90%) when nitrogen carrier gas was added. See **Ex. 1025**, col. 3, ll. 7-35, Table.

98. Example 2 of *Wu*, which used a catalyst that was treated with an aqueous solution of KOH followed by washing with water and drying at 82 °C, provided 3-methyl-1-butene with 97.7% purity after fractional distillation, but no conversion (or yield) data was provided. **Ex. 1025**, col. 3, l. 36 – col. 4, l. 14. In sum, a POSITA reviewing *Wu* would understand that base-treatment and use of an inert carrier gas can improve the conversion of 1-alcohols to 1-alkenes with no adverse effects on regioselectivity.

99. Importantly, *Wu* does not mention any affirmative step of calcining any catalysts, let alone following base treatment. See **Ex. 1025**. Dr. Taber nonetheless testified that a POSITA would “know” from *Wu* that a base-treated catalyst is “always calcined.” See **Ex. 2007**, p. 118, ll. 1-5. I disagree. In my opinion, a POSITA reviewing *Wu* would have no reason to assume that any of the

catalysts described therein had been calcined. Indeed, when I and my fellow co-workers on *Wright '451* reproduced Example 2 from *Wu*, we conducted no such calcination and instead followed the procedure as provided by *Wu*. See **Ex. 1039**, ¶ [98] (Example 6). What is more, I disagree with Dr. Taber's testimony that any catalyst in *Wu*'s Example 1 had been base-treated. See **Ex. 2007**, p. 94, l. 2 – p. 96, l. 10; p. 97, l. 16 – p. 98, l. 6. *Wu*'s Example 1 could not more clearly state that “[i]n the following inventive and comparative runs, 3-methyl-1-butanol was *dehydrated over gamma-alumina in the presence or absence of nitrogen....*” **Ex. 1025**, col. 3, ll. 8-12. In contrast, Example 2 of *Wu* clearly states “[t]he following inventive run illustrates the dehydration of 3-methyl-3-butanol to 3-methyl-1-butene over *base-treated gamma-alumina in the presence of nitrogen.*” **Ex. 1025**, col. 3, ll. 36-42. In my opinion a POSITA reviewing *Wu* would not agree with any of Dr. Taber's strained interpretations of the prior art reference. What is more, if a POSITA reading *Wu* would have known that *Wu*'s catalysts were calcined, then there would be no need to combine *Wu* with *Chokkaram*, as Dr. Taber does in his Second Declaration.

100. In sum, a POSITA reviewing *Wu* would understand that a 1-alkene regioselectivity of greater than 95% could be achieved by fractionally distilling the 1-alkene product that was prepared using a  $\gamma$ -alumina catalyst that is base-treated with sodium or potassium hydroxide and dried. But a POSITA would not know

from *Wu* the conversion provided by that catalyst or the time-on-stream. See **Ex. 2007**, p. 98, ll. 16 – p. 101, l. 2.

101. Further, *Wu*'s disclosure of base-treatment with sodium or potassium hydroxide would not have motivated a POSITA to select a calcium promoter, or to calcine a calcium-promoted  $\gamma$ -alumina catalyst at a temperature of about 420 °C to about 480 °C. Further, in my opinion, a POSITA would not have combined *Wu* with *Chokkaram*, and even if such a combination were made, *Chokkaram* does not remedy any of the deficiencies of *Wu*.

102. A POSITA have not been motivated to combine *Wu* with *Chokkaram* in order to improve the conversion or time-on-stream, let alone with any reasonable expectation of success. As discussed above, *Chokkaram* discloses a detailed analysis of cobalt- and nickel-aluminate catalysts that were prepared from co-precipitates of cobalt and nickel salts with aluminum hydroxide in a molar ratio of 1:2 (Co or Ni : Al, respectively). **Ex. 1041**, p. 158, col. 2 – p. 163. Indeed, *Chokkaram* is entirely silent as to time-on-stream and states that conversion is largely a function of reaction temperature, with higher temperatures favoring greater conversion. See **Ex. 1041**, p. 165 (Tables 3-4) showing conversion increases with increasing reaction temperature.

103. A POSITA reviewing *Chokkaram* would not have been motivated to combine *Chokkaram* with *Wu*. First, *Chokkaram*'s teachings are directed to

understanding the effect of calcination temperature on fundamental changes in the crystalline structures of the cobalt- and nickel-aluminates. **Ex. 1041**, p. 158, col. 2 – p. 163 (Figures 1-4, Tables 1-2). Second, *Chokkaram* provides only two calcination temperatures for alumina—673 K (i.e., 400 °C) and 1173 K (i.e., 900 °C)—the latter of which is only used for preparing alumina crystals and is not a temperature that a POSITA would think to use for calcining a base-treated or promoted  $\gamma$ -alumina catalyst. In view of *Chokkaram*'s teachings as a whole, a POSITA would have no reason to combine *Wu* with *Chokkaram* because *Wu* is directed to developing modified  $\gamma$ -alumina catalysts while *Chokkaram* is directed to understanding the chemistry of cobalt- and nickel-aluminates. In my opinion, it is only through hindsight that a POSITA reviewing *Wu* would land on *Chokkaram* as a reference having any relevance to preparing base-treated or promoted alumina catalysts.

104. Further, even if a POSITA were to combine *Wu* with *Chokkaram*, a POSITA would not arrive at the presently claimed invention with any reasonable expectation of success. Indeed, a POSITA reviewing *Chokkaram* would understand that calcination can be performed at 400 °C. But a POSITA would have no reasonable expectation that calcination of a base-treated  $\gamma$ -alumina catalyst, let alone a calcium-promoted  $\gamma$ -alumina catalyst would have any predictable effect on

the resulting conversion of 1-alcohols to 1-alkenes, 1-alkene regioselectivity, or time-on-stream.

105. Moreover, the revised substitute claims require “a calcium-promoted  $\gamma$ -alumina catalyst calcined at a temperature of about 420 °C to about 480 °C, wherein the catalyst has not been silanized,” and both *Wu* and *Chokkaram* are wholly silent as to teaching the use of calcium-promoted  $\gamma$ -alumina catalysts or the effect that calcination of such a calcium-promoted catalyst has on conversion or 1-alkene regioselectivity. Thus, *Wu*, alone or in view of *Chokkaram*, provide a POSITA with no reasonable expectation that “a calcium-promoted  $\gamma$ -alumina catalyst calcined at a temperature of about 420 °C to about 480 °C, wherein the catalyst has not been silanized” could have any effect on the performance (positive or negative) of the dehydration of 1-alkanols, as measured by conversion, regioselectivity, or time-on-stream.

106. What is more, *Wright '451* teaches away from the use of alkali-base modification of  $\gamma$ -alumina catalysts as taught by *Wu*. See **Ex. 1039**, ¶¶ [42], [98], Table 2, Fig. 9. This is because the alkali-base modification does not maintain robust conversion as a function of time-on-stream. Instead, *Wright '451* shows that conversion as a function of time-on-stream can be maintained by silanization or the use of a calcium-promoted  $\gamma$ -alumina catalyst. Further, *Wright '451* also shows that base-treatment followed by calcination, as in comparative examples 10-11 does not

improve the time-on-stream, as regioselectivity decreased for Example 10 and conversion decreased significantly in Example 11. **Ex. 1039**, ¶¶[43], [102]-[103], Table 2, Fig. 10. Therefore, a POSITA reviewing *Wright '451* would have no reason to select *Wu* as a reference for further research into  $\gamma$ -alumina catalysts for the dehydration of bio-1-alcohols with high conversion, high 1-alkene regioselectivity, and extended time-on-stream.

107. In sum, a POSITA would not have been motivated to combine *Wu* and *Chokkaram*. The combination of *Wu* in view of *Chokkaram* provides no reasonable expectation that the claimed catalyst could be used with any reasonable expectation of success to provide the claimed result, and the prior art *Wright '451* teaches away from *Wu*. For at least these reasons, in my opinion substitute claim 29 is patentable over *Wu* in view of *Chokkaram*.

#### **D. Substitute Claim 29 is Patentable Over *Wright '788***

108. As discussed above, *Wright '788* discloses a  $\gamma$ -alumina catalyst that is first treated with an aqueous base solution, washed with water, dried, and then reacted with a chlorosilane, washed with a hydrocarbon solvent, and air-dried. (See **Ex. 1009**, ¶ [0020].) *Wright '788* discloses an exemplary embodiment in which a  $\gamma$ -alumina catalyst was base-treated, dried, and then silanized by reacting with a 2 wt-% solution of dipenyldichlorosilane. (See **Ex. 1009**, ¶ [0034].) After air-drying, the silanized catalyst dehydrated 1-butanol to provide butene with an

alcohol conversion of 96% to 98%, a 1-butene regioselectivity of 94% to 95%, and a chemical yield of 1-butene of approximately 90%. (See **Ex. 1009**, ¶ [0034], Table 1.)

109. In contrast to *Wright* '788's teachings, substitute claim 29 requires a "a calcium-promoted  $\gamma$ -alumina catalyst calcined at a temperature of about 420 °C to about 480 °C, wherein the catalyst has not been silanized." A POSITA would not view *Wright* '788 as teaching or suggesting both calcium-treatment and calcination. *Wright* '788 fails to teach or suggest modification of a  $\gamma$ -alumina catalyst with a calcium promoter followed by calcination, and *Wright* '788 would not have provided a POSITA any reasonable expectation that the claimed catalyst would provide the requisite conversion, bio-1-alkene regioselectivity, and TOS stability. Further, a POSITA would be motivated to use a silanized catalyst, as described by *Wright* '788, whereas substitute claim 29 specifically requires that "the catalyst has not been silanized." Therefore, the specific  $\gamma$ -alumina catalyst required by substitute claim 29 would not have been obvious to a POSITA based on *Wright* '788.

110. Based on *Wright* '788's teaching to use a base-treated and silanized  $\gamma$ -alumina catalyst, *Wright* '788 in view of any background knowledge in the art would not have motivated a POSITA to utilize a calcium-promoted  $\gamma$ -alumina catalyst that is then calcined (but not silanized) to dehydrate a C<sub>6</sub> to C<sub>10</sub> bio-1-

alcohol. What is more, a POSITA would not have had any reasonable expectation that using the claimed calcium-promoted  $\gamma$ -alumina catalyst that is then calcined (but not silanized) would provide the claimed combination of high alkene conversion, high bio-1-alkene regioselectivity, and chemical yield of bio-1-alkene. Finally, a POSITA would have viewed the claimed results as unexpected over the results obtained using *Wright '788*'s catalyst to dehydrate bio-1-octanol.

111. In sum, *Wright '788* provides no motivation to use the claimed catalyst with any reasonable expectation of success, and the claimed catalyst provides unexpected results compared to *Wright '788*. For these reasons, in my opinion substitute claim 29 is non-obvious and patentable over *Wright '788*.

112. Further, to the extent that the PTAB views *Wright '788* as the closest prior art to the substitute claims, then the substitute claims provide unexpected results over *Wright '788*.

113. Based on *Wright '788*'s disclosure that C<sub>5</sub> to C<sub>23</sub> 1-alkanols can be similarly dehydrated (**Ex. 1009** at ¶ [0017]), a POSITA would have attempted to dehydrate a C<sub>6</sub> to C<sub>10</sub> bio-1-alcohol using the catalyst of *Wright '788*. Experiments reported in the '048 patent as Example 10 were conducted in which 1-octanol was dehydrated using a  $\gamma$ -alumina catalyst that was first treated with a base, dried, silanized, and then calcined at 400 °C. (See **Ex. 1001** at p. 23, col. 28, ll. 51-67; **Ex. 1006** at pp. 61-62 of 719; **Ex. 1005** at pp. 42-43 of 236.) The results of these

experiments show that the claimed combination of 1-alkene regioselectivity and alcohol conversion to alkenes could not be achieved with a  $\gamma$ -alumina catalyst that was first treated with a base, dried, silanized, and then calcined at 400 °C. (See **Ex. 2004**.) Referring to **Ex. 2004**, a series of 16 experiments were conducted in which the catalyst remained the same, while the pump rate (mL/min) was varied. The data show that the base-treated, silanized, and calcined  $\gamma$ -alumina catalyst provided under certain conditions a bio-1-octene regioselectivity of 92% or higher, but with a conversion of at most 89%. (See **Ex. 2004**, Run 4.) Similarly, where the base-treated, silanized, and calcined  $\gamma$ -alumina catalyst provided an octene conversion of at least 95%, the bio-1-octene regioselectivity dropped between 61% and 85%. (See **Ex. 2004**, Runs 1-3.) In my opinion, a POSITA would view these results as unexpected, particularly given the high regioselectivity and conversion with which the base-treated and silanized  $\gamma$ -alumina catalyst of *Wright '788* dehydrated 1-butanol to 1-butene.

114. Dr. Taber argues that the comparative data in Example 10 of the '048 patent and **Ex. 2004** is not relevant because calcium acetate was not used to prepare the comparative modified catalyst, calcination was performed at 400 °C (and not at optimum temperature identified by the inventors of 440 °C), and the temperature was 325 °C (not 315 °C). **Ex. 1038**, ¶¶53-¶54. I disagree. It is important to understand that Example 10 of the '048 patent was conducted to determine

whether the catalyst disclosed by *Wright* '788 would in fact provide a similar conversion and 1-alkene regioselectivity for bio-1-octanol as was obtained by *Wright* '788 for bio-1-butanol. As such, the experiment attempted to adhere to the teachings of *Wright* '788, without introducing additional inventive concepts from the inventions described in the '048 patent.

115. Against this backdrop, Example 10 is an apt comparison showing that the silanized catalysts of *Wright* '788—even with calcination—do not provide the same high conversion and 1-alkene regioselectivity for the dehydration of bio-1-octanol as is disclosed in *Wright* '788 for bio-1-butanol. For example, the catalysts in *Wright* '788 and Example 10 of the '048 patent were both washed with a base, dried (but not calcined), and then silanized. Compare **Ex. 1009**, ¶ [0034] with **Ex. 1001**, col. 28, ll. 54-61. *Wright* '788 generally discloses washing with a base such as sodium or potassium hydroxide and the Example in *Wright* '788 is silent as to which base was used. See **Ex. 1009**, ¶¶ [0020], [0034]. *Wright* '788 makes no mention of calcium hydroxide, calcium acetate, or any other Group II metal base, and therefore the use of calcium acetate would have gone beyond the teachings of *Wright* '788. Regarding calcination and the calcination temperature in Example 10 of the '048 patent, *Wright* '788 makes no mention of calcination of any catalysts. With respect to the calcination temperature, Dr. Taber himself argues that the prior art as a whole teaches that optimum calcination temperatures are between 400 °C

and 550 °C. **Ex. 1038**, ¶26. Therefore, despite *Wright* '788's not calcining the silanized catalyst, calcining of such silanized catalyst at 400 °C is fairly representative of the prior art teachings as a whole—particularly according to Dr. Taber's testimony that a POSITA would understand that all modified catalysts are calcined and Chokkaram's purported teaching of a preferred calcination temperature of 400 °C. Further, the prior art is entirely silent as to any advantages or benefits provided by calcining a  $\gamma$ -alumina catalyst, let alone a calcium-promoted  $\gamma$ -alumina catalyst at a preferred temperature range of about 420 °C to about 480 °C, or more preferably about 440 °C to about 460 °C. Therefore, Dr. Taber is plainly incorrect that a comparative example should have included calcination at a preferred temperature identified by the inventors themselves. Finally, with respect to the reactor temperature, *Wright* '788 discloses that the dehydration of bio-1-butanol was performed at a reactor temperature of 380 °C and a flow rate of 0.8 mL/min (**Ex. 1009**, ¶ [0034]), while Example 10 of the '048 patent conducted the dehydration of bio-1-octanol at a reactor temperature of 325 °C to 335 °C and flow rates from 0.1 to 2.0 mL/min. See **Ex. 2004**. However, Dr. Taber testified that based on his knowledge, a POSITA would at best be able to control a reactor temperature within plus or minus 20 °C of a target temperature. **Ex. 2007**, p. 160, l. 19 – p. 163, l. 9. While I disagree with his analysis, based on his testimony a POSITA would view a reactor temperature of 315 °C as entirely

overlapping with a reactor temperature of 325 °C to 335 °C. Further, it was known that for a specific catalyst, at any given reaction temperature the regioselectivity and conversion are largely a function of flow-rate. Therefore, because Example 10 of the '048 patent varied the flow rates over a wide range, Example 10 is an apt representation of the conversion and 1-alkene regioselectivity provided by the *Wright* '788 catalyst for 1-octanol.

116. In sum, the results in Example 10 showing a bio-1-octene regioselectivity of 92% or higher, but with a conversion of at most 89% (**Ex. 2004**, Run 4), or an octene conversion of at least 95%, and bio-1-octene regioselectivity between 61% and 85% (**Ex. 2004**, Runs 1-3), are entirely unexpected based on the much higher 1-alkene regioselectivity and conversion demonstrated by *Wright* '788 for bio-1-butanol. For at least these reasons, in my opinion substitute claim 29 is patentable over *Wright* '788.

**E. Substitute Claim 29 is Patentable Over *Wright* '451**

117. As discussed above, *Wright* '451 discloses the use of calcium-promoted  $\gamma$ -alumina catalysts that are prepared by modifying a  $\gamma$ -alumina catalyst with a  $\text{Ca}^{+2}$  compound. **Ex. 1039** at ¶[14]. Whereas *Wright* '788 only includes a single example of dehydrating bio-1-butanol with high-conversion, regioselectivity, and time-on-stream using a base-treated and silanized  $\gamma$ -alumina catalyst, Examples 8, 9, and 12-24 of *Wright* '451 disclose catalysts that utilize

calcium-promoted  $\gamma$ -alumina with or without silanization that dehydrate bio-1-butanol with high-conversion, regioselectivity, and time-on-stream. *Wright '451* also teaches that calcium-promoted  $\gamma$ -alumina catalysts:

[C]an be further heated ... to provide drying or calcination of the catalyst. The drying or calcination can be carried out in vessel **203** or **205** at temperatures of about 100 °C to about 900 °C, or about 200 °C to 600 °C for a time period of about 1 h to about 8 h, or about 2 h to 6 h.

**Ex. 1039** at ¶[67]. *Wright '451*'s Examples 3, 8-22, 24, and 26 perform drying and/or calcination of various catalysts at 120 °C, 200 °C, 500 °C, 550 °C, and/or 600 °C for times of 30 min to 24 hours. **Ex. 1039** at pp. 24-33 (Examples 2-4, 8-22, and 24). The most frequently used calcination temperature in *Wright '451* is 550 °C (Examples 2-4 and 8-22), while a single example in *Wright '451* used a lower temperature of 500 °C (Example 24).

118. *Wright '451* discloses exemplary embodiments in which  $\gamma$ -alumina catalysts are calcium-promoted and then calcined at 500 °C, 550 °C, or 600 °C. See **Ex. 1039**, Examples 3, 8, 9, and 12-19. Such calcium-promoted catalysts dehydrated 1-butanol to provide butene with an alcohol conversion of greater than 95% and a 1-butene regioselectivity of 95% to about 99%. See **Ex. 1039**, p. 31 (Table 2).

119. Based on my review of the extensive prior art that has been submitted in this proceeding, *Wright '451* is the closest prior art to the substitute claims, which does not appear to be in dispute. **Ex. 2007**, p. 151, ll. 3-6.

120. Dr. Taber and Petitioners allege that the substitute claims would have been obvious to a POSITA over *Wright '451* alone. As a co-inventor on both *Wright '451* and the '048 patent, in my opinion there are several key differences between *Wright '451* and the substitute claims at issue in this proceeding. First, and foremost, *Wright '451*'s examples are focused solely on bio-1-butanol. While *Wright '451* discloses the dehydration of longer chain 1-alcohols as being possible, a POSITA would need to perform experiments using *Wright '451*'s catalysts to dehydrate longer chain 1-alcohols in order to know that similar conversion, 1-alkene regioselectivity, and time-on-stream could be obtained. Second, *Wright '451* teaches a reactor temperature of about 220 °C to about 440 °C, and that “[f]or each run the catalyst performance was evaluated at four temperatures: 340 °C, 360 °C, 380 °C, and 400 °C. The lowest 1-butanol conversion is always at 340 °C and conversion increases with temperature.” **Ex. 1039**, ¶¶ [13], [42]. Third, *Wright '451* discloses calcination of the modified  $\gamma$ -alumina catalysts “at temperatures of about 100 °C to about 900 °C, or about 200 °C to 600 °C” (**Ex. 1039** at ¶[67]) and in the examples teaches drying and/or calcination of various

catalysts at 120 °C, 200 °C, 500 °C, 550 °C, and/or 600 °C (**Ex. 1039** at pp. 24-33, examples 2-4, 8-24).

121. Substitute claim 29 requires:

A method for making a bio-alkene, comprising:

dehydrating a bio-1-alcohol by heating the bio-1-alcohol in a continuous flow reactor at a temperature of 240 °C to 360 °C with a calcium-promoted  $\gamma$ -alumina catalyst calcined at a temperature of about 420 °C to about 480 °C, wherein the catalyst has not been silanized, and

wherein the resulting bio-alkene has a carbon chain of 6 to 10 carbon atoms and is produced in a conversion of at least 95% and a bio-1-alkene regioselectivity of 95% to about 99% from a single-pass reaction after at least 20 hours time-on-stream.

122. To the extent that the PTAB views *Wright '451* as the closest prior art to the substitute claims, then the substitute claims provide unexpected results over *Wright '451*.

123. First, substitute claim 29 requires that the calcium-promoted  $\gamma$ -alumina catalyst is calcined at a temperature of about 420 °C to about 480 °C. As discussed in the '048 patent, 1-octanol was dehydrated with  $\gamma$ -alumina catalyst that was first treated with a calcium acetate (i.e., promoter) solution, followed by calcination in air at 440 °C or 500 °C for 12 hours. Dehydration conducted at 315 °C provided conversion (to octenes) of 97.2% and 95.5%, respectively, with a

regioselectivity of 97.4%<sup>4</sup> and 93.1% (per Table 2). **Ex. 1001**, col. 26, l. 10 – col. 27, l. 9 (Examples 1-3). Thus, the '048 patent demonstrates that the higher calcination temperature of 500 °C (as taught by *Wright '451*) resulted in a *less selective* catalyst. That is, a calcium-promoted catalyst calcined at the lower temperature of 440 °C exhibited markedly better regioselectivity for 1-octene and also exhibited higher conversion compared to the same catalyst calcined at 500 °C. Based on *Wright '451*'s disclosure of preferred calcination temperatures of 200 °C to 600 °C, and examples at 500 °C and 550 °C, in my opinion a POSITA would view this result as entirely unexpected.

124. What is more, a POSITA would view the results provided by the lower calcination temperature of 440 °C as a difference in kind and not one of degree. Indeed, in this field obtaining even a 1-2% improvement in either conversion or 1-alkene regioselectivity can provide significant benefit from the perspective of reactor design and operating costs—for example, by eliminating a recycling stream. Here, 1-octene regioselectivity increased from 93.1% to 96.7% and conversion increased from 95.5% to 97.2%. A POSITA would view these improvements as significant for either variable, but to see improvements in both is very significant and highly unexpected.

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<sup>4</sup> I note that the calculation of 1-Octene Selectivity for Example 1 in Table 2 appears to be incorrect and should be 96.7%.

125. Second, substitute claim 29 requires dehydrating a bio-1-alcohol by heating the bio-1-alcohol in a continuous flow reactor at a temperature of 240 °C to 360 °C. As discussed in the '048 patent, dehydration reactions conducted at 330 °C and 350 °C provided conversion (to 1-, 2-, and 3-hexenes) of 96.3% and 96.9%, respectively, with a regioselectivity for the formation of 1-hexene of 95.8%<sup>5</sup> and 96.8% (as reported in Table 5, below).

TABLE 5

| Catalyst    | Reactor Temperature (° C.) | 1-Hexene (%) | 2-Hexenes & 3-Hexene (%) | 1-Hexanol (%) | 1-Hexene Selectivity (%) |
|-------------|----------------------------|--------------|--------------------------|---------------|--------------------------|
| EXAMPLE 11A | 330                        | 92.8         | 3.5                      | 1.8           | 95.8                     |
| EXAMPLE 11B | 350                        | 93.8         | 3.1                      | 1.7           | 96.8                     |
| EXAMPLE 11C | 370                        | 90.1         | 3.2                      | 1.6           | 96.6                     |

**Ex. 1001**, col. 29, ll. 1-25 (Example 11). Surprisingly, increasing the reactor temperature to 370 °C reduced conversion (to hexenes) to 93.3% (from 96.9% at 350 °C), while maintaining a regioselectivity of 96.6% (as shown in Table 5, above). This temperature sensitivity was unexpected, particularly in view of prior art calcium-promoted catalysts such as those disclosed in *Wright '451* that provide conversion in excess of 95% at 380 °C. See **Ex. 1039**, Figs. 10-12.

126. Based on *Wright '451*'s teaching of reactor temperature at about 220 °C to about 440 °C, and examples at 340 °C, 360 °C, 380 °C, and 400 °C, a

<sup>5</sup> I note that the calculation of 1-Hexene Selectivity for Example 11A in Table 5 appears to be incorrect and should be 96.4%.

POSITA would also view this result as unexpected. Specifically, *Wright '451* teaches that the lowest 1-butanol conversion is always at 340 °C and *conversion increases with temperature.*” **Ex. 1039**, ¶¶ [13], [42]. Nothing in *Wright '451* would have taught or suggested to a POSITA that conversion of 1-hexanol would decrease at 370 °C.

127. What is more, a POSITA would view increase in conversion at lower temperatures as a difference in kind and not one of degree. As discussed above, even a 1-2% improvement in conversion can provide significant design and operational benefits. Here, 1-hexene conversion increased from 93.3% to 96.9% while maintaining regioselectivity. A POSITA would view this improvement as significant.

128. In sum, the results in Examples 3 and 11 showing the surprising effect of the claimed calcination temperature and reactor temperature, respectively, are entirely unexpected based on the teaching of *Wright '451*. For at least these reasons, in my opinion substitute claim 29 is patentable over *Wright '451*.

#### **F. Substitute Claims 30-44 Are Patentable Over the Prior Art**

129. Substitute claims 30-44 depend directly or indirectly from independent substitute claim 29 and incorporate all of its limitations. Thus, for the same reasons discussed above with respect to substitute claim 29, dependent claims 30-44 are also non-obvious and patentable over the prior art.

**G. Substitute Claims 34, 37, 40, and 43 Are Additionally Patentable Over the Prior Art of Record**

130. Substitute claims 34, 37, 40, and 43 depend from claims 33, 36, 39, and 42, respectively, which require that “the calcium-promoted  $\gamma$ -alumina has a CaO content of about 1% to about 2%” and more narrowly reflect the examples of the '048 patent. Substitute claims 34, 37, 40, and 43 further require that “prior to the dehydrating, the catalyst has been calcined at a temperature of about 440 °C to about 460 °C.” (Patent Owner’s, Revised Non-Contingent Motion to Amend at Appendix A.) As discussed above, the disclosures of *Le* in view of *De Bruyn* or *Wu* in view of *Chokkaram*, would not have motivated a POSITA to use a calcium-promoted  $\gamma$ -alumina catalyst that is then calcined at a temperature of about 420 °C to about 480 °C. Nor does *Wright* '788 disclose or suggest the use of a calcium-promoted  $\gamma$ -alumina catalyst that is then calcined at a temperature of about 440 °C to about 460 °C. Finally, *Wright* '451 discloses calcination of a calcium-promoted  $\gamma$ -alumina catalyst at a preferred temperature range of 200 °C to 600 °C, with specific calcination conditions disclosed at 500 °C, 550 °C, and 600 °C. Thus, the closest prior art to the revised substitute claims is *Wright* '451’s disclosure of calcination of a calcium-promoted  $\gamma$ -alumina catalyst at 500 °C.

131. As set forth in the '048 patent specification, a calcium-promoted  $\gamma$ -alumina catalyst that is calcined at a temperature of about 440 °C to about 460 °C, provides a bio-1-alkene regioselectivity of 97%, alcohol conversion of 97%, and a

chemical yield of 94%; whereas, calcination at a temperature of 500 °C reduced bio-1-alkene regioselectivity to 93%, alcohol conversion to 95.5%, and the chemical yield of 1-octene drops to 88%. (See **Ex. 1006** at p. 43 of 719, ¶ [0076], Table 1; **Ex. 1005** at p. 24 of 236, ¶ [0076], Table 1.) In my opinion, a POSITA would find the fact that a lower calcining ceiling temperature afforded an optimum catalyst in terms of both regioselectivity and conversion to be a truly unexpected result. And nothing in *Le* in view of *De Bruyn*, *Wright '788*, *Wright '451*, or *Wu* in view of *Chokkaram*, optionally in view of the background knowledge in the art, teaches or suggests that base treatment and calcination at a temperature of about 440 °C to about 460 °C can improve 1-alkene regioselectivity, alcohol conversion, and chemical yield of the target 1-alkene (particularly at reactor at a temperature of 240 °C to 360 °C). For this additional reason, claims 34, 37, 40, and 43 are non-obvious over the prior art of record in this proceeding, including *Le* in view of *De Bruyn*, *Wright '788*, *Wright '451*, or *Wu* in view of *Chokkaram*.

**H. Substitute Claims 35, 38, 41, and 44 Are Additionally Patentable Over the Closest Prior Art, *Wright '451***

132. Substitute claims 35, 38, 41, and 44 depend from claims 34, 37, 40, and 43, respectively, which require that “the calcium-promoted catalyst has been calcined at a temperature of about 440 °C to about 460 °C” and more narrowly reflect the examples of the '048 patent. Substitute claims 35, 38, 41, and 44 further require that “the reactor is heated at a temperature of 315 °C to 350 °C.” (Patent

Owner's, Revised Non-Contingent Motion to Amend at Appendix A.) As discussed above, *Wright '451* teaches the use of a calcium-promoted  $\gamma$ -alumina catalyst that is held at a reactor temperature at about 220 °C to about 440 °C, with examples at 340 °C, 360 °C, 380 °C, and 400 °C. *Wright '451* also teaches that the lowest 1-butanol conversion is always at 340 °C and *conversion increases with temperature.*" **Ex. 1039**, ¶¶ [13], [42].

133. In contrast, substitute claims 35, 38, 41, and 44 more narrowly reflect the unexpected results provided in Example 11 of the '048 patent in which the conversion of 1-hexanol at 330 °C and 350 °C was 96.3% and 96.9%, respectively, while a reactor temperature of 370 °C reduced conversion (to hexenes) to 93.3% (from 96.9% at 350 °C). **Ex. 1001**, col. 29, ll. 1-25. As discussed above, this temperature sensitivity was unexpected, particularly in view of prior art calcium-promoted catalysts such as those disclosed in *Wright '451* that provide conversion in excess of 95% at 380 °C. See **Ex. 1039**, Figs. 10-12.

134. For this additional reason, in my opinion claims 35, 38, 41, and 44 are non-obvious over the closest prior art of record in this proceeding, *Wright '451*.

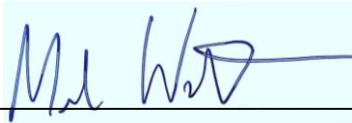
## **VII. CONCLUSION**

135. In view of the foregoing, I conclude that substitute claims 29-44 of the '048 patent would not have been obvious to a POSITA in view of the prior art cited in Petitioners' Opposition to Patent Owner's Motion to Amend. Accordingly,

for the reasons discussed herein it is my opinion that revised substitute claims 29-44 of the '048 patent are patentable over the prior art of record.

I declare under penalty of perjury that, to the extent of my knowledge and belief, the foregoing is true and correct. I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the understanding that knowing and willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of title 18 of the United States Code.

Dated June 13, 2024

A handwritten signature in blue ink, appearing to read "Michael Wright", is written over a light blue rectangular background. A horizontal line extends to the right from the end of the signature.

Michael E. Wright, Ph.D.