

UNITED STATES PATENT AND TRADEMARK OFFICE

BEFORE THE PATENT TRIAL AND APPEAL BOARD

TOPSOE, INC.,
Petitioner

v.

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

Inter Partes Review No. IPR2025-01173

REPLY DECLARATION OF PROF. DR. HARALD KLEIN

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

I. INTRODUCTION AND BACKGROUND

1. I previously submitted a declaration (EX1003) dated June 28, 2025 in support of Petitioner Topsoe, Inc. in IPR2025-01173. A copy of my curriculum vitae was attached to my prior declaration as EX1004.

2. In my prior declaration I provided opinions concerning the invalidity of claims 1-6, 11, and 12 of U.S. Patent No. 11,673,805 (“the ’805 Patent”), specifically with respect to the series configuration of the endothermic reforming step and the autothermal reforming step. My prior declaration concluded that claims 1, 11, and 12 are anticipated by Reinertsen (EX1005) (Ground 1), claims 1-6, 11, and 12 are obvious over the combination of Reinertsen and Darde (EX1007; English translation EX1008) (Ground 2), and claims 1-6, 11, and 12 are obvious over the combination of Rytter (EX1009) and Darde (Ground 3).

3. I understand that Patent Owner Air Liquide submitted a Patent Owner Response dated April 27, 2026 (Paper 28) (“POR”), supported by the declaration of Dr. Joseph D. Smith (EX2021). I disagree with Dr. Smith’s opinions, and have prepared this supplemental declaration to address what I consider to be fundamental errors in his opinion and analysis of the prior art.

4. I have also reviewed the transcript of the deposition of Dr. Smith taken in the proceeding on July 7, 2026, which I cite in this declaration as “Smith Tr.” (EX1032).

5. I was previously deposed by counsel for Patent Owner on April 2, 2026 in Houston, TX, and I understand a copy of the deposition transcript was submitted in this IPR proceeding by Patent Owner as EX2023. In this declaration, I also clarify certain answers from my deposition that the POR quotes out of context.

6. To assist the Board in comparing my analysis with Dr. Smith’s I have adopted the same citation conventions Dr. Smith used in his declaration: I cite Reinertsen (EX1005) by paragraph number; Darde (EX1008) by page and line number; Rytter (EX1009) by exhibit page (“p.__/35”) and line number; and the deposition transcripts by page and line. For the Air Liquide Technology Handbook (EX1018), which Dr. Smith did not cite, I use the exhibit’s page numbers (“p.__/108”) so that the Board can easily locate the cited material.

7. I am competent to testify under oath with regards to this supplemental declaration, and will do so if called as a witness, as follows herein.

II. LEGAL STANDARDS

8. I was previously informed by counsel for Topsoe of the legal standards surrounding anticipation and obviousness, which I set forth in my original declaration (EX1003) at ¶¶14-25.

9. I understand that Patent Owner has argued that a POSA would not be motivated to combine Reinertsen and/or Rytter with Darde because a POSA would consider certain performance characteristics to be inferior. Specifically, Dr. Smith alleges that the inclusion of PSA H₂ separation, instead of a Pd membrane separation process, followed by cryogenic CO₂ separation would result in reduced H₂ and CO₂ purities and capture rates.

10. I address the factual inaccuracies of Dr. Smith's opinions below. I do not offer opinions on the law; however, counsel for Topsoe has informed me that the relevant inquiry in an obviousness analysis is not whether the prior art combination reflects the best engineering choice, but whether the asserted combination was taught, suggested, or otherwise within the predictable grasp of a POSA. I understand the Supreme Court of the United States has addressed this issue directly: "When there is a design need or market pressure to solve a problem and there are a finite number of identified, predictable solutions, a person of ordinary skill has good reason to pursue the 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." *KSR Int'l Co. v. Teleflex Inc.*, 550 U.S. 398, 421 (2007).

11. I also understand that Patent Owner has argued that a POSA would not be motivated to combine Reinertsen and/or Rytter with Darde if Reinertsen/Rytter teaches away from the combination. Counsel for Topsoe has informed me that the

legal doctrine of “teaching away” is valid; however, I disagree that Reinertsen/Rytter teaches away from an obviousness combination with Darde, that is the addition of Darde’s PSA H₂ separation upstream of Reinertsen/Rytter’s cryogenic CO₂ separation. For the reasons discussed below, I find the opposite to be true—Reinertsen and Rytter both encourage the specific PSA H₂ separation followed by cryogenic CO₂ separation process scheme disclosed in Darde.

III. GROUND 1: REINERTSEN EXPRESSLY TEACHES PSA H₂ SEPARATION UPSTREAM OF CRYOGENIC CO₂ SEPARATION

12. I previously described the factual bases for the anticipation of claims 1, 11, and 12 of the ’805 Patent by Reinertsen. *See* EX1003 at ¶¶57-70 (description of Reinertsen); ¶¶106-172 (anticipation analysis).

13. I understand that Dr. Smith has opined that Reinertsen does not satisfy claim elements [1.5] and [1.6] of claim 1 and claim 11. I disagree with these opinions. Although Reinertsen does not identify PSA H₂ separation upstream of cryogenic CO₂ separation in any of its “embodiment examples” listed in Table 1 (Fig. 10)—a point I acknowledged at my deposition (EX2023, 67:12-68:9)—Reinertsen nonetheless provides specific details for implementing PSA as an alternative to Pd-membrane H₂ separation.

A. Reinertsen expressly teaches PSA H₂ separation on the gas output from the WGS reactor, which is necessarily upstream of Cryogenic CO₂ Separation

14. I previously described how Reinertsen clearly discloses that PSA is a “feasible” alternative to Pd-membrane for H₂ separation. EX1003, at ¶67; *see also* EX1005, ¶[0012] (“Preferably, the hydrogen separation process comprises a PSA process.”); ¶[0097] (“The schemes vary in . . . use of Pd-membrane or PSA for hydrogen separation. . . .”); ¶[0056] (“Still, it can be feasible to use PSA for some hydrogen production plants.”). I understand from Dr. Smith’s declaration that he does not dispute this fact.

15. However, Dr. Smith does not acknowledge in his declaration that Reinertsen specifically discloses that PSA H₂ separation would occur on the output of the water gas shift reactor (“WGS”), which would necessarily be upstream of cryogenic CO₂ separation. EX1005, ¶[0085] (“Embodiments may alternatively use PSA to separate hydrogen from the gas output from the WGS reactor.”). This sentence exactly parallels Reinertsen’s description of its preferred membrane two paragraphs earlier: “Embodiments preferably separate hydrogen from the gas output from the WGS reactor using a membrane. In particular, a Pd-film membrane may be used.” EX1005, ¶[0083]. This confirms that PSA is a direct substitute for the Pd-membrane at the same position in the process (Unit 12 of Figures 3-7), upstream of any CO₂ separation. Paragraph [0085] further describes the PSA operating on the

full, CO₂-containing shifted gas: “CO, CO₂ and CH₄ are adsorbed, letting the hydrogen pass through at process pressure” and “[n]ot to overload the adsorbent, water is condensed before the PSA unit”.

16. Dr. Smith’s contrary opinion—that the “alternatively” sentence of ¶[0085] refers to “alternative embodiments” of the Figure 2 type, in which amine CO₂ removal precedes the PSA (EX2021, ¶37; POR at 6-8)—cannot be reconciled with Reinertsen’s own text. In Figure 2, the PSA (Unit 10) does *not* receive “the gas output from the WGS reactor.” Reinertsen states expressly that in Figure 2, the PSA “separates the hydrogen from the gas 82 *that has already been depleted of CO₂* by the [amine type separation] process 8.” EX1005, ¶[0103] (emphasis added). The only configuration disclosed in Reinertsen in which a PSA separates hydrogen “from the gas output from the WGS reactor”—the words of ¶[0085]—is therefore one in which the PSA occupies the Unit-12 position of Figures 3-7, upstream of CO₂ separation. In my opinion, Dr. Smith’s reading does not place ¶[0085] “in context”; it instead rewrites it.

17. Dr. Smith’s deposition testimony confirms this plain reading. He agreed that ¶[0085] “does say after from the water-gas shift means it’s after the water-gas shift” (Smith Tr. 150:9-151:8), and that a reading under which the PSA “is right after the water-gas shift” is a reading a POSA “would believe . . . as well” (Smith Tr. 191:16-192:14). I also note that claim elements [1.5] and [1.6] do not

require the PSA to sit immediately after the WGS reactor with no intervening equipment; they require PSA separation of the fourth (shifted) synthesis gas stream, with cryogenic separation of the resulting residual stream. The cooling and water condensation between the WGS reactor and the PSA taught by Reinertsen (¶[0085]), Rytter (EX1009, p.12/35, l.29), and Darde (EX1008, p.9/23, l.37-p.10, l.3, steps e)-g)) do not change that analysis: under any reading Dr. Smith conceded, the PSA operates on the shifted, CO₂-containing gas upstream of CO₂ separation, which is what the claims recite.

B. Reinertsen expressly teaches Cryogenic CO₂ Separation after any H₂ Separation Unit

18. I understand that Dr. Smith does not dispute that Reinertsen expressly teaches cryogenic CO₂ separation.

19. However, Dr. Smith does not acknowledge in his declaration that cryogenic CO₂ separation occurs downstream of H₂ separation in each of the five Reinertsen embodiments that use cryogenic CO₂ separation. *See* EX1005, Embodiment Example 2 (Figure 4, Element 13; ¶¶[0109]-[0110]), Embodiment Example 4 (Figure 6, Element 13; ¶¶[0114]-[0116]), Embodiment Examples 5-7 (Figure 7, Element 13; ¶¶[0122]-[0123]). I understand that Dr. Smith admitted this fact in his deposition. Smith Tr. 117:9-118:18 (Embodiment Examples 2, 4, and 5-7 each show the cryogenic separation unit downstream of the hydrogen removal unit); 213:5-19 (“Q. And in terms of the order of steps, when cryogenic CO₂ removal is

used, it's always downstream of the hydrogen removal. Would you agree with that?

. . . A. I believe that's correct.”).

20. In my opinion, that ordering is not a coincidence. As I discussed in my original declaration at ¶238, a POSA would have readily understood that separating H₂ gas upstream of a cryogenic CO₂ separation necessarily reduces the total amount of gas fed to the cryogenic CO₂ separation unit, reducing the physical size of the cryogenic unit, resulting in capital expenditure (CAPEX) and operating expense (OPEX) reductions. Dr. Smith agreed with my analysis in his deposition, and further admitted that a POSA would understand that the placement of cryogenic CO₂ separation after H₂ removal follows a “best practice” because the larger volume component (H₂ in steam methane reformer plants) would be separated first: “[O]ne of the best practices is that you remove the component with the highest volume percent in the mixture first . . . if hydrogen has got the most composition, most volume, then you remove hydrogen first.” Smith Tr. 194:14-195:3. Dr. Smith further agreed that, in a steam-reforming plant, hydrogen is “most likely” the higher-volume component of the shifted gas, and that “a POSA would be led to remove the hydrogen first before CO₂ removal That is most likely correct.” Smith Tr. 195:4-12; see also Smith Tr. 55:6-17, 56:7-13 (a POSA would apply best practices in ordering separation processes); 61:15-62:6 (the order of separations “can be a best practice”). Although Dr. Smith noted that best practices are a “starting point” subject

to case-specific evaluation (Smith Tr. 195:13-196:3), that starting point is precisely the ordering recited in the claims.

C. Reinertsen expressly teaches that PSA H₂ Separation upstream of Cryogenic CO₂ Separation requires compression (Claim 11)

21. Consistent with PSA H₂ separation upstream of cryogenic CO₂ separation (claim elements [1.5] and [1.6]), Reinertsen also expressly teaches the need for compression of the H₂-depleted feed to the cryogenic CO₂ separation unit (as required by claim 11).

22. Dr. Smith does not acknowledge that Reinertsen expressly teaches that selecting PSA H₂ separation results in “CO₂ being released at low pressure, e.g. atmospheric pressure, and so there is a subsequent need for compression and cooling.” EX1005, ¶[0056]. In addition to satisfying dependent claim 11, this “compression and cooling” of the H₂-depleted syngas stream exiting PSA also confirms that PSA H₂ separation necessarily occurs upstream of cryogenic CO₂ separation (which requires compression and cooling). Dr. Smith conceded this at his deposition when confronted with ¶[0056] itself: “So there is a subsequent need for compression and cooling. Agree completely. . . . We do need a compression step. In the PSA unit we have atmospheric CO₂ coming out. If you’re going to use it, you’re going to compress it . . . you’re going to use it to separate it cryogenically.” Smith Tr. 161:11-162:22. Asked whether one reason for that compression “would be for

the CO₂ syngas to be fed to a cryogenic CO₂ unit,” Dr. Smith answered: “Agreed.” Smith Tr. 162:23-163:4.

23. Rather than addressing Reinertsen’s express teaching regarding the compression requirement, Patent Owner focuses on an embodiment from Reinertsen that is not pertinent here—the selection of Pd-membrane for H₂ separation upstream of cryogenic CO₂ separation. As I explained in my deposition, although it may be theoretically possible not to require additional compression before the cryogenic CO₂ separation unit if the pressure is already high enough for cryogenic CO₂ separation, that is only relevant for Pd-membrane embodiments that release CO₂ at higher pressures. That is not relevant to PSA embodiments where the H₂-depleted feed to the cryogenic CO₂ separation unit is necessarily at low pressure. This must require compression, as expressly stated by Reinertsen. In my opinion, the underlying physics permit no alternative: a PSA adsorbs the CO₂ (with CO and CH₄) at the elevated feed pressure and releases it by “swing[ing] to low pressure to desorb the adsorbed molecules.” EX1009, p.12/35, ll.26-27. Darde quantifies it—the PSA offgas is at a pressure “below 2 bar(a)” (EX1008, p.8/23, ll.8-11) and must be compressed to “between approximately 20 and 100 bar(a)” for cryogenic purification (¶[0027])—and Patent Owner’s own handbook lists typical PSA offgas pressures of 1.1 to 10 bara, while the H₂ product “is delivered at a pressure close to feed pressure” (EX1018, p.38). Dr. Smith agreed that a POSA choosing PSA would

know that “you are not going to need a hydrogen compressor, but you are going to need a compressor to compress . . . the CO₂-rich stream for cryogenic separation” (Smith Tr. 115:11-116:1), and that “the hydrogen coming off of a PSA unit is a higher pressure and the CO₂ is a lower pressure” (Smith Tr. 159:5-8).

24. Because the POR supports its no-compression argument with fragments of my deposition testimony (POR at 10-13, 48-49), I clarify that testimony here: (a) My testimony at EX2023, 99:6-9 that Figure 4 shows no compressor between units 12 and 13 described Figure 4 as drawn—the Pd-membrane embodiment—and, as I explained elsewhere in the same deposition, even in the membrane case some compression may still be required depending on the natural gas supply pressure (EX2023, 81:20-83:10); it says nothing about the PSA path. (b) My testimony at EX2023, 104:2-11 was expressly conditional—“[i]f the pressure would be high enough”—a condition that can never be satisfied by a PSA tail gas released at approximately atmospheric pressure (EX1005, ¶[0056]). (c) The approximately 32 bar figure I discussed at EX2023, 106:22-108:23 is the pressure of the shifted gas at the WGS outlet—the PSA feed, and hence approximately the pressure of the hydrogen product (EX1005, ¶[0085]; EX1018, p.38)—not the pressure of the PSA offgas. The POR transplants that number onto the wrong stream, as Dr. Smith’s own testimony confirms (Smith Tr. 159:5-8).

D. The CO₂ Compression requirement would not discourage a POSA from selecting PSA over Pd-Membrane for H₂ Separation

25. Dr. Smith attempts to characterize Reinertsen as discouraging the selection of PSA over Pd-membrane for H₂ separation because of the compression requirement, discussed above. In my opinion, this is incorrect, and it overlooks that Reinertsen expressly teaches that Pd-membrane H₂ separation requires compression of product H₂. EX1005, ¶[0100] (“... produced H₂ from the membrane unit is at 3 bar and compressed to 350 bar for export.”)

26. A POSA would understand that the two H₂ separation alternatives (Pd-membrane vs. PSA) creates a direct tradeoff between compressing product H₂ (using Pd-membrane H₂ separation) vs. compressing the CO₂-rich feed to the cryogenic CO₂ separation unit (using PSA H₂ separation). Dr. Smith does not address this tradeoff in his declaration.

27. In my opinion, Reinertsen itself frames this tradeoff expressly—and in terms favoring technologies, like PSA, that keep the hydrogen at pressure: “As it is more demanding to pressurize hydrogen than CO₂, separation technologies that provide hydrogen at high pressure are sometimes preferred. This benefit can, however, be outweighed by the efficiency of a hydrogen-permeable membrane.” EX1005, ¶[0088]. PSA is the disclosed technology that provides hydrogen at high pressure. EX1005, ¶[0085] (hydrogen passes through “at process pressure”); EX1018, p.38. Rytter contains the same teaching nearly verbatim. EX1009, p.13/35,

11.2-9. A reference that expressly identifies the claimed option as “sometimes preferred”—and describes the countervailing membrane benefit as one that merely “can” outweigh it—does not criticize, discredit, or discourage that option.

28. As I explained in my deposition, a POSA would understand that H₂ compression is much more complicated than CO₂ compression, in both CAPEX and OPEX, as compared to CO₂ compression because hydrogen is the molecule with the lower molecular weight. EX2023 at 124:22-125:8. More specifically:

- H₂'s molecular weight is approximately 2 kg/kmol, versus approximately 44 kg/kmol for CO₂. Because each compression stage imparts comparatively little pressure rise to so light a gas, hydrogen service requires many stages, or reciprocating machines, with interstage cooling; the specific compression work per unit mass of H₂ is far higher than for CO₂. See EX2023, 124:22-125:8; Smith Tr. 167:15-168:8 (molecular weights of 2 and 44).
- One of the biggest challenges relating to H₂ compression is metal degradation (called H₂ embrittlement), increasing the risk of cracking or failure under cyclic loading—this necessitates specialized alloys, coatings, and design practices for compressor shafts, valves, and seals that are unnecessary in CO₂ compression systems. Dr. Smith agreed that H₂ embrittlement “does not exist” for CO₂ compression (Smith Tr. 167:1-7), and that material selection and

special compressor seals are required, adding capital and operating cost (Smith Tr. 143:12-144:4).

- H₂ molecules are very small and have high diffusivity, which leads to leakage through tiny clearances, more sophisticated sealing requirements, and greater emphasis on leak detection. By contrast, CO₂ is much easier to contain. Dr. Smith agreed. Smith Tr. 167:8-171:7.
- H₂ compression involves substantially more demanding staging and thermal-management design than CO₂ compression to comparable delivery conditions, with more stages and interstage cooling required for the reasons above.
- H₂ compression has more demanding safety considerations. Hydrogen has very low ignition energy, a wide flammability range in air (about 4–75% by volume), and creates a nearly invisible flame. These characteristics require additional safeguards in compressor design, ventilation, and monitoring. By contrast, the primary hazard with CO₂ is asphyxiation in confined spaces rather than fire or explosion. Dr. Smith agreed these hazards attend H₂ compression, that “CO₂ doesn’t have that type of hazard”, and that these safety considerations bear on the choice of the Figure 4 (Pd-membrane) scheme. Smith Tr. 142:13-144:9, 168:10-170:12.
- The magnitude of the H₂ compression power is also far greater: hydrogen for export or transport must reach approximately 350 bar (EX1005, ¶[0100]) or,

for gaseous transport applications, on the order of 600-900 bar (EX2023, 126:8-14), whereas the CO₂ need only be compressed to the cryogenic unit's operating pressure (approximately 20-100 bar(a), EX1008, p.8/23, ll.24-30) or CO₂-delivery conditions (e.g., 16 bar, EX1005, ¶[0100]). Hence, due to the different end pressures, the required compression power for H₂ compression is higher than for CO₂ compression. However, the suction pressure is usually more important, for both H₂ and CO₂ compression. Therefore, a POSA would rather select the highest possible suction pressure for H₂ compression, which is achieved by a PSA for H₂ separation, whereas with a Pd membrane process the suction pressure for H₂ compression is low.

29. Although Dr. Smith did not address these facts in his declaration, he agreed with my analysis in his deposition, including the safety concerns specific to H₂ compression. Smith Tr. 142:13-144:9 (H₂ compression required for the Figure 4 scheme; unique safety considerations; special seals; contributes to CAPEX and OPEX; a consideration in choosing the Figure 4 scheme); 167:1-171:7 (embrittlement, diffusivity, flammability, leak detection; no CO₂ analogue). Indeed, when my deposition testimony at EX2023, 124:22-125:8 was read to Dr. Smith, he adopted it without qualification: "I do. . . . it's more complicated to compress hydrogen than it is to compress CO₂, and I agree with that"; a POSA "would weigh

. . . that trade-off” (“Sure”); and the tradeoff “definitely would affect CapEx and OpEx.” Smith Tr. 164:22-166:4.

30. In my opinion, a POSA weighing these tradeoffs (CAPEX, OPEX, and safety) would have viewed PSA as at least an equally suitable—and commercially the standard—choice over Pd-membrane for H₂ separation. PSA is the commercial choice for H₂ separation in syngas production. *See* EX2023 at 118:11-23. This is confirmed by Patent Owner Air Liquide’s own prior art *Technology Handbook* (Feb. 2018), which identifies >70 PSA references “in operation or under construction” (EX1018 at 38) while listing no Pd-membrane H₂ separation technology at all among its 64 technologies. The Air Liquide Technology Handbook’s only H₂ membrane offering (EX1018 at 42) is a polymeric hollow-fiber membrane—not a Pd-membrane—used to recover H₂ from refinery purge gas, delivering the H₂ into the low-pressure permeate at a purity of “>99% vol,” versus the PSA’s high-pressure product at purities up to 99.9999% (EX1018 at 38). Dr. Smith confirmed the handbook lists no reference installations for its membrane product (Smith Tr. 183:19-184:4), that he is aware of no commercial Pd-membrane installation in operation or under construction for a steam-reforming plant (Smith Tr. 180:14-181:7), and that in 2020 “palladium membrane was the exception, PSA hydrogen removal was the rule” (Smith Tr. 204:11-15).

31. On the point of suitability of the options Reinertsen teaches, I recognize that Reinertsen also reports, in discussing its own comparative example, that vendor pricing showed that “for hydrogen separation, PSA is considerably more costly than using a Pd-membrane”, with a larger footprint for PSA. EX1005, ¶¶[0118]-[0119]. That unit-cost observation does not change my opinion. It is an intra-reference cost preference offered to explain why Reinertsen’s authors favored their membrane examples over their own amine-plus-PSA comparative example; it is not a statement that PSA is unworkable or should be avoided—the same reference simultaneously calls PSA “feasible” (¶[0056]) and “[p]referabl[e]” for the hydrogen separation process (¶[0012]). It also addresses only the purchase cost of the separation unit itself, not the system-level compression tradeoff that Reinertsen ¶[0088] and Rytter expressly identify and that Dr. Smith agreed a POSA would weigh, with effects on both capital and operating cost (Smith Tr. 164:22-166:4). And a vendor-price projection premised on a technology that existed only at pilot scale (EX2023, 118:11-23; Smith Tr. 180:14-181:7) would have been understood by a POSA as a projection, not commercial experience. Expressing a preference among disclosed alternatives is not a teaching away from any of them.

IV. GROUND 2: DARDE DETAILS THE PSA H₂ SEPARATION UPSTREAM OF CRYOGENIC CO₂ SEPARATION PROCESS SCHEME DESCRIBED BY REINERTSEN

32. I previously described the factual bases for the obviousness of claims 1-6, 11, and 12 of the '805 Patent by Reinertsen combined with Darde. *See* EX1003 at ¶¶57-70 (description of Reinertsen); ¶¶78-81 (description of Darde); ¶¶163-229 (obviousness analysis of Reinertsen combined with Darde).

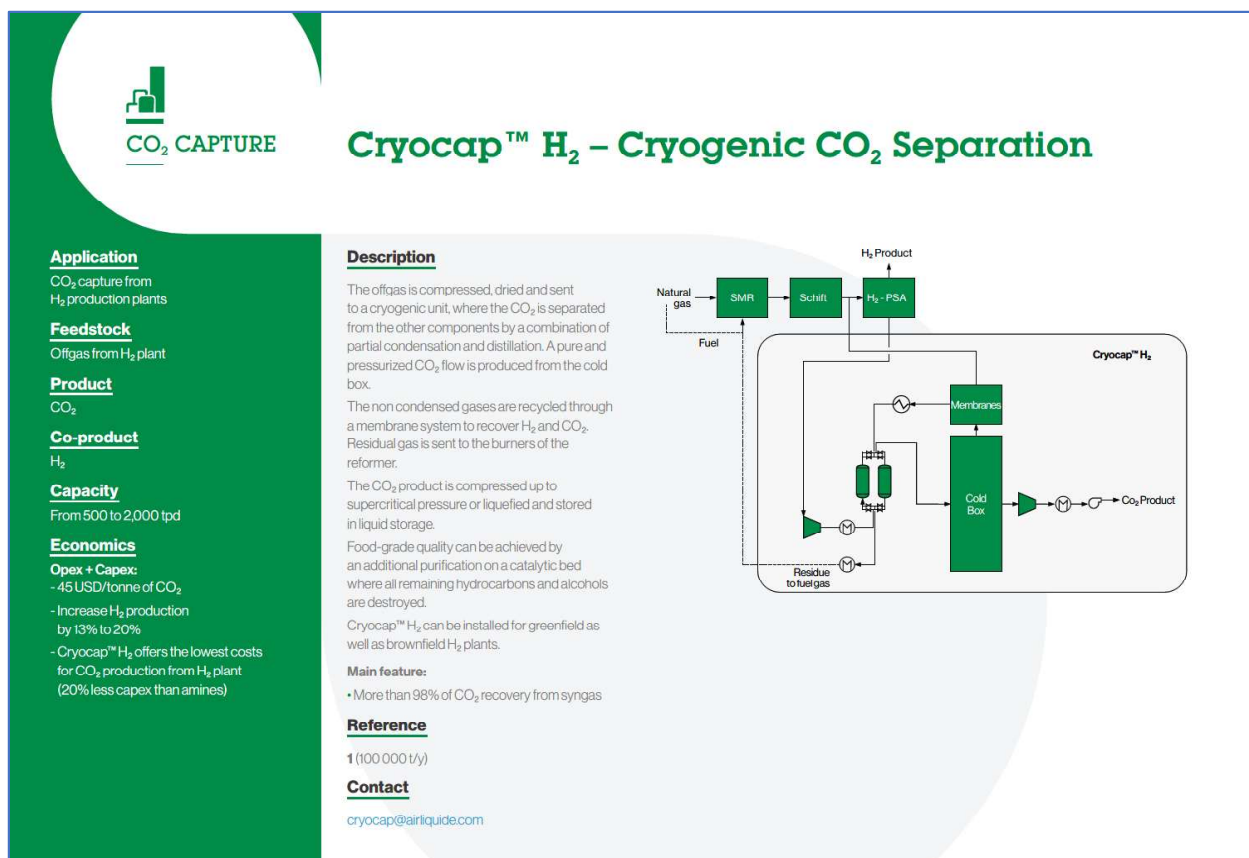
A. Motivation to combine Reinertsen with Darde is expressly provided by Reinertsen

33. I understand that Dr. Smith opined that a POSA would not have been motivated to combine Reinertsen with Darde. I disagree with this argument. As discussed above in ¶¶13-23, Reinertsen specifically teaches: (a) using PSA for H₂ separation as an alternative to Pd-membrane; (b) placing PSA H₂ separation in the process flow on the gas output of the WGS reactor; (c) always placing cryogenic CO₂ separation after H₂ separation; and (d) compressing the CO₂-rich syngas from the PSA before cryogenic CO₂ separation. Darde's placement of PSA H₂ separation upstream of cryogenic CO₂ separation embodies each of these teachings from Reinertsen.

34. As discussed above, Reinertsen specifically teaches that PSA is a feasible alternative to Pd-membrane for H₂ separation. *See* ¶¶14, 27 above. A POSA would understand these statements as encouragement, not discouragement from utilizing PSA H₂ separation.

35. In my opinion, Dr. Smith's deposition confirmed this point. When asked directly, he conceded: "Q. So when Reinertsen says you can use PSA hydrogen as an alternative, a person of ordinary skill in the art would know how to apply that in the Reinertsen process. Would you agree with that? A. I would." Smith Tr. 204:16-21. In my opinion, that admission—from Patent Owner's own expert—concedes that a POSA both could and would know how to implement PSA H₂ separation in Reinertsen's process, which is the combination presented in Ground 2 of my declaration (EX1003), and with it a reasonable expectation of success.

36. Darde (in addition to disclosing the elements of dependent claims 2-6) confirms the process flow described by Reinertsen—utilizing PSA H₂ separation upstream of cryogenic CO₂ separation. Darde's process flow represents basic knowledge of a POSA as of the critical date of the '805 Patent, which is readily confirmed by Patent Owner Air Liquide's own prior art *Technology Handbook* (Feb. 2018). EX1018 at p.24/108 (describing Air Liquide's commercial Cryocap™ H₂ process involving PSA H₂ separation upstream of cryogenic CO₂ separation: a steam methane reformer with shift conversion, followed by a H₂ PSA, with the PSA offgas "compressed, dried and sent to a cryogenic unit" for CO₂ capture):



Dr. Smith agreed that Cryocap™ H₂ shows “a cryogenic CO₂ unit positioned after a PSA hydrogen removal,” published two years before the ’805 Patent’s earliest claimed priority date, and that this ordering “is within a toolbox of a person of ordinary skill in the art.” Smith Tr. 129:7-135:3 (esp. 132:3-7, 134:16-135:3).

37. I note that Dr. Smith argues (for both Grounds 2 and 3 of my declaration (EX1003)) that my “finite set of known design choice” (i.e., finite options) analysis rests on a “false” premise because Reinertsen and Rytter allegedly disclose more than two H₂ separation options—pointing to absorption, “temperature swing” adsorption, and combinations of solid and liquid membranes. EX2021, ¶¶72, 98. In my opinion, this argument conflates CO₂-separation techniques with H₂ separation.

Reinertsen’s absorption discussion is explicitly about removing CO₂, the acid gas (EX1005, ¶[0082]), not about producing a hydrogen product. The “combination of solid membranes and liquid membranes” achieves “a rapid permeation of CO₂, while H₂ is kept back” (¶[0085])—again a CO₂-selective route, not an additional H₂-separation technology—and the temperature-swing sentence of ¶[0085] describes a variant within the same adsorption family as PSA. For separating a hydrogen product from shifted syngas, Reinertsen identifies exactly two technologies—a hydrogen-selective membrane, preferably a Pd-membrane (¶¶[0011], [0083]-[0084]), and PSA (¶¶[0012], [0063], [0085])—the same either/or pair presented in Table 1 (¶[0097]; EX2023, 67:12-68:9). That framing matched commercial reality: Dr. Smith agreed PSA was “the proven way commercially to separate hydrogen from syngas” (Smith Tr. 203:11-14), that a POSA would have been “much more familiar with PSA technology than membrane separation technology” (Smith Tr. 185:20-186:2), and that PSA had reached a technology-readiness level of roughly 8-10 versus roughly 5-7 for membranes (Smith Tr. 178:16-179:12, 185:13-19).

B. Dr. Smith’s “Inferior H₂ Purity and Recovery” argument is incorrect and would not have discouraged a POSA from using PSA

38. Dr. Smith argues that Reinertsen already performs at a 99.9% purity using Pd-membrane separation, and Reinertsen and Darde only state that PSA separation produces a 99% H₂ purity. EX2021 at ¶¶49, 59. In my opinion, a POSA would understand that this singular metric is incorrect—PSA represents the standard

for high-purity H₂ separation in the industry and H₂ purity would not serve to discourage using PSA.

39. As an initial matter, the premise that Reinertsen “already performs” at these levels mistakes the modeling assumption for a result. Reinertsen states its simulation assumptions expressly: “in amine cases, it is assumed that 100% of inlet CO₂ is removed by the unit, in membrane cases, 93% H₂ separation *is assumed*” EX1005. ¶[0100] (emphasis added). The “H₂ recovery factor” shown for the membrane cases in Table 2 of Figure 11 simply reflects that assumed input. A POSA would recognize that an assumed figure for technology that had never operated beyond pilot scale (EX2023, 118:11-23; Smith Tr., 178:16-179:12, 180:14-181:7) is not a demonstrated benchmark that an alternative must exceed. By contrast, a POSA would have understood that commercial PSA achieves H₂ purities up to 99.9999%, with H₂ recovery rates of 60 to 90% as a design-selectable parameter. These correct PSA performance metrics would have been readily verifiable by a POSA from a number of sources, including Patent Owner’s own prior art *Technology Handbook* (Feb. 2018). EX1018 at p.38/108 (“Hydrogen up to 99.9999%”; “H₂ recovery rate: 60 to 90%”); *see also* EX1019 (Riquarts, 99.999% H₂ purity and more than 90% recovery) at p.3/6; EX1016 (Appl, 99.9999+ vol% H₂ purity).

40. Dr. Smith’s comparison also sets a minimum comparison of a floor against a maximum. Darde’s “greater than 99%” is a claimed minimum—“a stream

of hydrogen . . . having a purity of at least 99%” (EX1008, p.10/23, ll.1-3, step g); *see also* EX1008, p.18/23, ll.18-20)—not a statement of the maximum purity the disclosed PSA can achieve. Patent Owner’s own Ground 3 purity benchmark confirms this point: Rytter’s hydrogen specification of “ ≥ 99.97 mol% for fuel cells” is satisfied in Rytter’s Figure 3 simulation, in which the hydrogen is separated by PSA (the simulation’s “PSA off-gas (152)”). EX1009, p. 15/35, ll.24-28. In my opinion, the very purity figure Patent Owner cites as evidence of PSA’s inferiority was itself achieved with PSA.

41. Nor do single-pass recovery differences translate into lost hydrogen in these integrated processes. In Reinertsen, the PSA rest gas is either used as fuel for feed preheating (EX1005, ¶[0103]) or recycled to the reformers at rates of at least 50%, 80%, or 90% (*id.*, Abstract, ¶¶[0057], [0061]), and Reinertsen teaches that lower CO conversion “can be rectified by recycle of rest gas” (*id.*, ¶[0081]); Rytter likewise recycles the off-gas “for maximum carbon efficiency” (EX1009, p.15/35, ll.20-21). Hydrogen slipping to the PSA tail gas is thus recovered or its energy value captured—it is not “lost.” Moreover, the 86% PSA recovery in Reinertsen’s comparative example sits within the 60-90% commercial design range published by Patent Owner itself (EX1018 p.38/108).

42. Dr. Smith admitted in his deposition that PSA produces very high purity H₂ product and that purity would not serve as a basis for discouraging using PSA for

H₂ separation. Smith Tr. 198:17-199:5 (reading the 99.9999% figure from EX1018: “That’s what it says”); 197:15-198:11 (PSA purity is a design parameter; “99, 99.9 is not unreasonable to expect”); 203:5-6 (“99 versus 99.9 versus 99.9999, that’s pretty high”); and—directly contrary to his declaration, where purity was his deciding consideration (EX2021, ¶59)—Smith Tr. 202:1-6 (purity “won’t be a deciding factor for me”).

43. Patent Owner also quotes Reinertsen’s statement that a plant is “not favorably designed for separating a pure CO₂-stream for storage.” POR at 25-26; EX2021, ¶75 (quoting EX1005, ¶[0094]). In context, that sentence describes a historical ammonia-plant design—steam reforming with PSA hydrogen fed, with nitrogen, to Haber-Bosch synthesis—that included no CO₂ capture unit at all. EX1005, ¶¶[0093]-[0094]. The criticism is of venting CO₂, not of PSA H₂ separation, and not of PSA upstream of cryogenic capture—the configuration Patent Owner itself commercialized as Cryocap™ H₂ (EX1018 p. 24/108). Rather, Reinertsen’s ¶[0094] articulates the design need—capture the CO₂—that motivates adding cryogenic capture downstream of a PSA.

C. Darde’s CO₂ purity and capture rate metrics are not material to the obviousness analysis

44. Dr. Smith argues that because Darde focuses on steam production and capturing condensates, Darde’s failure to discuss CO₂ capture rates would discourage a POSA from combining Reinertsen with Darde.

45. In my opinion, this argument is misplaced because Darde has only been cited (with respect to claim 1 and 11) as a secondary reference demonstrating PSA H₂ separation upstream of cryogenic CO₂ separation—a process flow scheme specifically described by Reinertsen. Reinertsen itself teaches CO₂ purity and capture rates associated with cryogenic CO₂ separation, which would remain unchanged. To the extent Darde teaches lower CO₂ purity or capture rates than Reinertsen, a POSA would understand it is not due to upstream PSA H₂ separation. In any event, Dr. Smith’s suggestion that a POSA would “suspect” unfavorable performance from Darde’s silence on capture rates (EX2021, ¶¶48-52) does not withstand scrutiny. As an initial matter, capture rate and purity are distinct metrics, and Dr. Smith concedes that Darde expressly discloses favorable *purity* on both sides of the separation—hydrogen “greater than 99%” (EX1008, p.16/23, ll.7-8) and, for the cryogenic unit, “CO₂ purities greater than 99%” (EX1008, p.8/23, ll.20-21). His argument therefore rests solely on Darde’s silence as to *capture rate*. But that silence reflects only Darde’s different focus—steam production and condensate handling—and is not evidence of poor performance. It is affirmatively contradicted by Patent Owner’s own documents: Air Liquide’s commercial embodiment of cryogenic capture applied to PSA offgas—Cryocap™ H₂—advertises “[m]ore than 98% of CO₂ *recovery* from syngas.” EX1018 p. 24/108 (emphasis added).

V. GROUND 3: RYTTER, LIKE REINERTSEN, POINTS A POSA TO PSA H₂ SEPARATION UPSTREAM OF CRYOGENIC CO₂ SEPARATION

46. I previously described the factual bases for the obviousness of claims 1-6, 11, and 12 of the '805 Patent by Rytter combined with Darde. *See* EX1003 at ¶¶71-77 (description of Rytter); ¶¶230-285 (obviousness analysis of Rytter combined with Darde). I understand Dr. Smith responds at EX2021, ¶¶83-114. Many of his Rytter arguments repeat the arguments addressed above for Grounds 1 and 2, and my responses apply equally here. I address the Rytter-specific points below.

47. In my opinion, Rytter points a POSA directly to the claimed arrangement. Its clauses recite “performing a water-gas-shift on the gas output from the reforming processes; and separating hydrogen from the shifted gas.” EX1009, p.4/35, ll.32-33. Its description of PSA is materially identical to Reinertsen’s: CO, CO₂ and CH₄ are adsorbed, “letting the hydrogen pass through at process pressure,” with water “condensed before the PSA unit.” EX1009, p.12/35, ll.21-29. It states that “[e]mbodiments include using any known technique for hydrogen and carbon dioxide separation.” EX1009, p.12/35, l.30. And it expressly discloses the claimed ordering: “Embodiments also include the hydrogen being separated from the gas mixture before the carbon dioxide is separated from the gas mixture” EX1009, p.20/35, ll.7-10. Rytter also contains the same no-fixed-order disclaimer as Reinertsen, quoted in ¶53 below. EX1009, p.25/35, ll.20-25.

48. Dr. Smith’s “six options” argument for Rytter (EX2021, ¶98) is incorrect for the reasons discussed in ¶37 above. Rytter’s absorption discussion, like Reinertsen’s, is about removing CO₂, the acid gas (EX1009, p.11/35, l.30-p.12/35, l.9); its “solid membranes and liquid membranes” combination achieves “a rapid permeation of CO₂, while H₂ is kept back” (EX1009, p.12/35, ll.19-20)—a CO₂-selective route, not an H₂-separation technology. For producing a hydrogen product from the shifted gas, Rytter, like Reinertsen, identifies a hydrogen-permeable membrane (preferably Pd) and PSA. EX1009, p.12/35, ll.10-18, 21-29; p.13/35, ll.7-9.

49. Dr. Smith’s argument that amine CO₂ capture outperforms cryogenic capture (EX2021, ¶¶92, 94-96) is not pertinent and, in any event, is contradicted by the references. The claims recite cryogenic CO₂ separation; the pertinent question is whether the claimed configuration was an obvious one, not whether an unclaimed amine alternative might also have been chosen. And the art itself commends the cryogenic choice: Reinertsen prefers cryogenic separation (EX1005, ¶[0059]), describing it as “a cost and energy efficient method” that delivers the CO₂ “directly in the liquid form” needed for deposition (¶¶[0060], [0086]); its recycle embodiments exist precisely to raise carbon-capture efficiency (¶[0061]); and Patent Owner itself selected cryogenic capture for its commercial Cryocap™ technology

(EX1018, pp.7, 24). A POSA selecting cryogenic CO₂ capture follows, rather than departs from, the art—and follows Patent Owner’s own commercial practice.

50. Dr. Smith’s related assertion that, if the order were reversed, a POSA “would have selected” a Pd-membrane rather than PSA (EX2021, ¶¶93, 99-102, 114) rests on the purity and pressure premises rebutted in ¶¶39-42 above. Rytter itself describes the membrane’s efficiency as a benefit that merely “can” outweigh the hydrogen-at-pressure advantage of technologies like PSA (EX1009, p.13/35, ll.8-9)—a two-way tradeoff between the only two identified H₂-separation options. Commercially, the selection actually made at scale, overwhelmingly, was PSA. See ¶¶29, 36 above. And Patent Owner’s own 99.97 mol% purity benchmark was achieved with PSA. See ¶39 above; EX1009, p.15/35, ll.20-28.

51. For claim 11 (EX2021, ¶¶110-114), the analysis in ¶¶21-24 above applies with equal force. If PSA is used for the H₂ separation, the CO₂-rich tail gas is inherently released at desorption (low) pressure regardless of the approximately 32 bar feed pressure—that is how PSA regeneration works (EX1009, p.12/35, ll.26-27)—so compression (and cooling) of that stream before cryogenic separation is required, exactly as Rytter (p.13/35, ll.1-4: the CO₂ “has to be brought to high pressure and in liquid form” for pipeline transport, with injection pressures of 50-300 bar), Reinertsen (¶[0056]), and Darde (EX1008, p.8/23, ll.8-11, 24-30) each teach, and as Dr. Smith conceded (Smith Tr. 115:11-116:1, 161:11-163:4). Patent

Owner's no-compression theory again depends on Pd-membrane retentate physics, and its use of my "32 bar" testimony transplants the PSA feed pressure onto the PSA offgas. See ¶24(c) above.

52. Finally, Dr. Smith argues the combination would require modifying Rytter's WGS operation, and that Darde's offgas pressure (below 2 bar(a)) is "incompatible" with Rytter's process, defeating any reasonable expectation of success. EX2021, ¶¶104-107. Rytter itself teaches that the WGS arrangement follows from the separation choice: only high-temperature WGS may be applied before a Pd-membrane (which requires elevated temperature), with the option of low-temperature WGS after the membrane and partial recycle, whereas conventional shift is used otherwise. EX1009, p.11/35, ll.25-29. Reinertsen teaches the same accommodation for PSA: "it still might be feasible to operate only one WGS reactor," and "[u]ltra-high conversion of CO is not always needed, and can be rectified by recycle of rest gas." EX1005, ¶¶[0080]-[0081]. Dr. Smith agreed that selecting WGS severities, temperatures, and reactor counts are "all true" design levers and "tools in the toolbox" of a POSA. Smith Tr. 81:1-10. And the "incompatibility" of a 2 bar(a) offgas with a cryogenic unit is not, in my opinion, an incompatibility—it is the compression step that all three references teach and that claim 11 expressly recites.

VI. CRITIQUE OF MY “ROUTINE OPTIMIZATION” TESTIMONY IGNORES THE EXPRESS TEACHINGS IN REINERTSEN, RYTTER, AND DARDE

53. Patent Owner and Dr. Smith contend that my failure to conduct detailed modelling is fatal to my opinions concerning obviousness. However, Reinertsen and Rytter make clear that the choice and order of process steps is within the skill of a POSA. For example, both Reinertsen and Rytter state:

The flow charts and descriptions thereof herein should not be understood to prescribe a fixed order of performing the method steps described therein. Rather, the method steps may be performed in any order that is practicable. Although the present invention has been described in connection with specific exemplary embodiments, it should be understood that various changes, substitutions, and alterations apparent to those skilled in the art can be made to the disclosed embodiments without departing from the spirit and scope of the invention as set forth in the appended claims.

EX1005, ¶[0223]; EX1009, p.25/35, ll.20-25.

54. Reinertsen also expressly acknowledges that a POSA would understand how to compare process alternatives based on known boundary conditions:

It is understood that the applied conditions are reasonable for comparing different process schemes, but that a variety of other conditions can be used; e.g. for different natural gas compositions, requirements for delivery of hydrogen and CO₂, and site specific conditions like possibilities for integration with other process units and the availability of electricity from the grid.

EX1005, ¶100.

55. Dr. Smith agreed with these principles in his deposition: chemical engineers possess a “toolbox” of standard options and best practices (Smith Tr. 76:9-

77:15, 81:1-10); a POSA can determine the compression requirements for these arrangements by hand or with standard tools such as Aspen—“[a] person skilled in the art would be able to do that” (Smith Tr. 113:3-22); the PSA-then-cryogenic ordering is “within a toolbox of a person of ordinary skill in the art” (Smith Tr. 134:16-135:3); best practice would lead a POSA to remove the hydrogen first (Smith Tr. 194:8-195:12); and, when Reinertsen offers PSA as an alternative, “a person of ordinary skill in the art would know how to apply that in the Reinertsen process” (Smith Tr. 204:16-21).

56. Specific to PSA H₂ separation, both Reinertsen and Rytter each expressly teach numerous process considerations for accommodating PSA H₂ separation upstream of cryogenic CO₂ separation. For example, Reinertsen teaches:

- PSA H₂ separation would occur on the gas output of the water gas shift (WGS) reactor, and water condensation would be utilized before the PSA unit to prevent overloading of the adsorbent. EX1005, ¶85.
- WGS operation may vary across embodiments, and WGS severity would be adapted if substituting PSA for Pd-membrane H₂ separation. EX1005, ¶¶80-81.
- The CO₂-rich product stream from the PSA unit would be released at low pressure, and therefore requires subsequent compression and cooling before cryogenic CO₂ separation. EX1005, ¶56.

57. Similarly, Rytter also discloses essentially the same process considerations for accommodating PSA H₂ separation:

- PSA H₂ separation would be performed on the shifted gas output from the WGS reactor. EX1009, p.4/35, ll.32-33.
- Water condensation would be utilized before a PSA unit to prevent overloading of the adsorbent. EX1009, p. 12/35, l.29.
- WGS severity would be adapted depending on the choice of PSA or Pd-membrane for H₂ separation. EX1009, p. 11/35, l.25-29.
- PSA H₂ separation produces H₂ product at high pressure for storage and transportation; if CO₂ is separated at high pressure, downstream compression energy is saved. EX1009, p. 13/35, l.2-9.

58. As I noted in the legal-standards section of my original declaration, I have been informed that the proper obviousness analysis asks whether a person of ordinary skill in the art would have had a reasonable expectation of success, not absolute predictability of success, in arriving at the claimed invention by combining the prior art. EX1003, ¶¶22-25. Applying that standard here, and for the reasons set out below, it is my opinion that the choice and order of these process steps was within the ordinary skill of a person in the art, and that a person of ordinary skill would have had a reasonable expectation of success in implementing PSA H₂ separation upstream of cryogenic CO₂ separation in Reinertsen and Rytter.

59. Each of these routine engineering considerations is independently confirmed by Darde, further demonstrating that a POSA would have had a reasonable expectation of success in utilizing PSA H₂ separation in Reinertsen and Rytter. For example, Darde teaches:

- WGS is utilized to convert unwanted CO to additional H₂ and CO₂. EX1008, p.7/23, ll.30-34.
- PSA H₂ separation would occur downstream of the WGS reactor, with the same intervening colling and water condensation steps taught by Reinertsen and Rytter. EX1008, p.9/23, l.37-p.10/23, l.3 (steps e)-g)).
- PSA H₂ separation produces an H₂ product of substantially pure hydrogen (EX1008, p.7/23, ll.36-37) at the high PSA feed pressure. (EX1005, ¶[0085]; EX1009, p. 12/35. Ll.27-28; EX1018, p.38).
- PSA H₂ separation produces a CO₂-rich gas that requires compression for downstream cryogenic CO₂ separation. EX1008, p.8/23, ll.24-30.

60. I also note that Dr. Smith, while criticizing my methodology, applied the same methodology himself. His declaration identifies no simulation, no comparative energy or cost analysis, and no design study of his own; his performance comparisons consist of reading values from Reinertsen's Figure 11—values that reflect the assumed inputs of ¶[0100] (see ¶38 above)—and quoting purity figures whose significance he disclaimed at his deposition (see ¶41 above).

At deposition, he confirmed that he has never himself simulated a PSA unit. Smith Tr. 17:20-18:8.

61. Finally, because the POR characterizes my deposition testimony on this subject as a concession (POR at 24-25, 29), I clarify it. On redirect, I explained why a detailed calculation is unnecessary to evaluate the disclosed options: “[T]o find a general judgment of all the options and the pros and the cons of all the options, it’s not required to do a detailed calculation.” EX2023, 124:14-21; *see id.* 123:17-125:19. The pros and cons of the two disclosed H₂-separation options—pressure allocation, compression duty, purity, maturity, safety—are stated in the references themselves and were common knowledge to a POSA, as Dr. Smith’s own testimony confirms.

VII. DR. SMITH’S OWN TESTIMONY CONFIRMS MY OPINIONS

62. Dr. Smith’s deposition confirmed each of the facts that I stated in my original declaration:

- that the only dispute for claim 1 over Reinertsen “is the order of PSA hydrogen and cryogenic CO₂” (Smith Tr. 190:17-191:7);
- that whenever Reinertsen uses cryogenic CO₂ separation, it is downstream of the hydrogen removal (Smith Tr. 213:5-19);
- that best practice would lead a POSA to remove the hydrogen—the largest-volume component—first (Smith Tr. 194:8-195:12);

- that H₂ compression is more complicated and more costly than CO₂ compression, with unique safety burdens, exactly as I testified (Smith Tr. 142:13-144:9, 164:22-166:4, 167:1-170:12);
- that selecting PSA entails compressing the CO₂-rich stream for cryogenic separation (Smith Tr. 115:11-116:1, 161:11-163:4);
- that PSA was the commercial “rule” and delivered purities he called “pretty high,” such that purity “won’t be a deciding factor” (Smith Tr. 198:17-204:15); and
- that a POSA “would know how to apply” Reinertsen’s PSA alternative “in the Reinertsen process” (Smith Tr. 204:16-21).

63. Together with the express teachings of Reinertsen, Rytter, and Darde, addressed above, these admissions confirm the opinions stated in my original declaration, all of which I reaffirm.

VIII. DEMONSTRATIVE: PRIOR ART ANTICIPATION AND OBVIOUSNESS

64. In order to visually demonstrate the anticipation and obviousness of claim 1 of the '805 Patent, I have created the demonstrative chart attached as Exhibit A to this declaration.

65. As shown in Exhibit A, attached to this declaration Reinertsen’s disclosed H₂-separation alternatives (Pd-membrane or PSA) and CO₂-separation alternatives (amine or cryogenic) occupy fixed positions in the disclosed process

flow—H₂ separation on the gas output of the WGS reactor, with CO₂ separation downstream—and the parallel disclosures of Rytter and Darde map to the same positions. Exhibit A is compiled solely from the record evidence cited in this declaration and in my original declaration and contains no information beyond that record evidence.

IX. ACKNOWLEDGEMENT

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



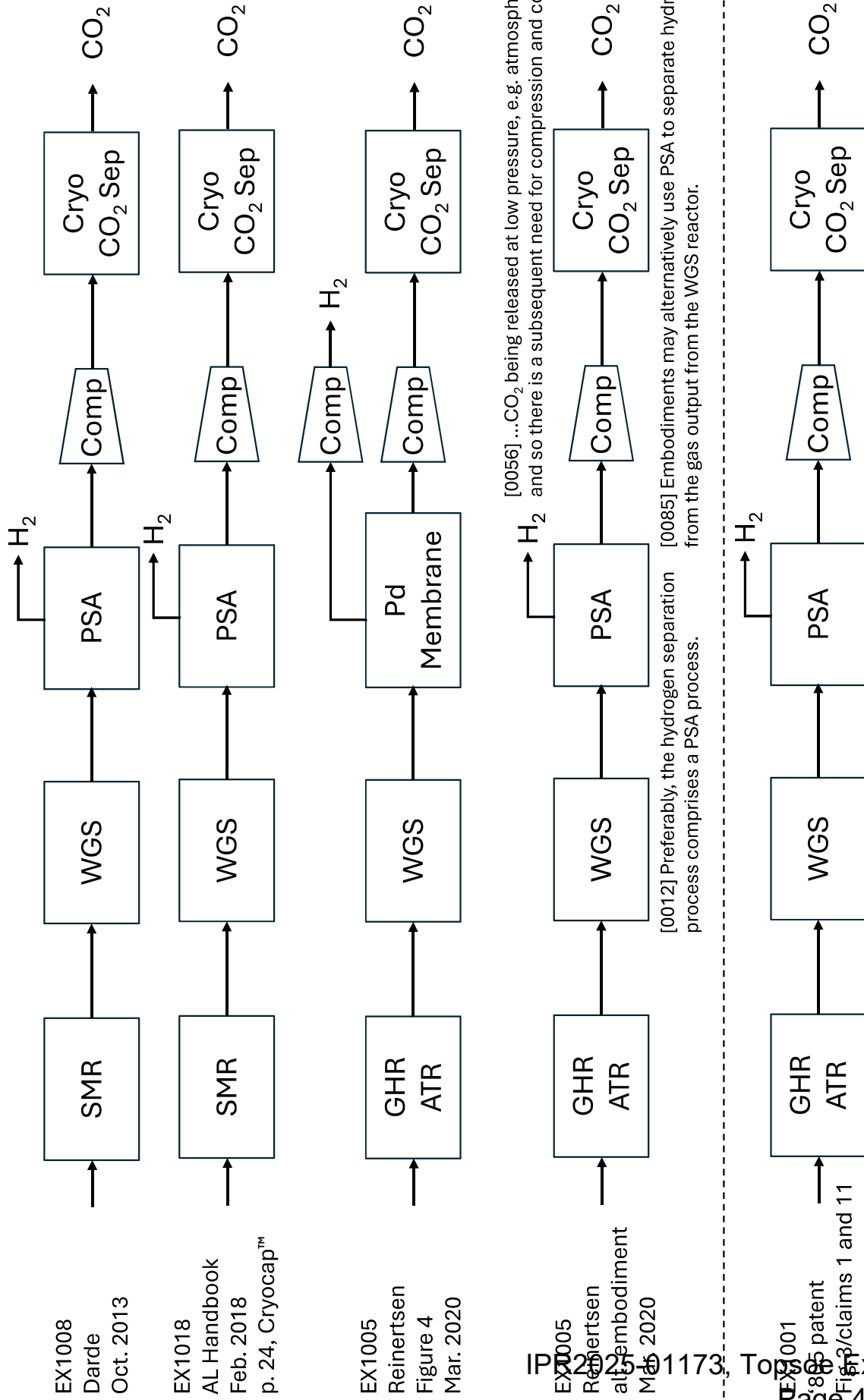
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Demonstrative Chart

Exhibit A

Prof. Dr.-Ing. Harald Klein, 11.07.2026



[0056] ...CO₂ being released at low pressure, e.g. atmospheric pressure, and so there is a subsequent need for compression and cooling.

[0012] Preferably, the hydrogen separation process comprises a PSA process. [0085] Embodiments may alternatively use PSA to separate hydrogen from the gas output from the WGS reactor.