

UNITED STATES PATENT AND TRADEMARK OFFICE

BEFORE THE PATENT TRIAL AND APPEAL BOARD

AIR PRODUCTS AND CHEMICALS, INC.,
Petitioner

v.

EVONIK OPERATIONS GMBH and EVONIK CORPORATION,
Patent Owner

Case IPR2024-00611
Patent 10,471,380

SUPPLEMENTAL DECLARATION OF DR. RICHARD W. BAKER

I, Dr. Richard W. Baker, of Redwood City, California, do hereby declare that:

1. I am over the age of 18 and competent to make this supplemental declaration.
2. On February 22, 2024, I submitted a declaration that discussed my background, qualifications, compensation, the technology at issue and my opinions.
3. I provide this supplemental declaration to address certain points made by Evonik in its December 12, 2024 Patent Owner Response and in Dr. Geise's December 12, 2024 declaration, Exhibit 2008.

I. ADDITIONAL MATERIALS CONSIDERED

4. In formulating my opinions in this supplemental declaration, I have considered the following materials, in addition to the materials identified in my February 22, 2024 declaration:

- a. December 12, 2024 Patent Owner Response
- b. December 12, 2024 Declaration of Geoffrey Geise (Exhibit 2008)
- c. Dr. Richard W. Baker December 8, 2024 deposition transcript (Exhibit 2009)
- d. AP-1046: Huang, *Pressure ratio and its impact on membrane gas separation processes*, Journal of Membrane Science 463 (2014) 33–40
- e. AP-1047: Baker, *Natural Gas Processing with Membranes: An Overview*, Ind. Eng. Chem. Res. 2008, 47, 2109-2121

- f. AP-1048: Bhatti, *Polymeric Membranes Used in Natural Gas Processing to Separate Carbon Dioxide: the Facing Problems and Solutions*, Journal of Environmental Science and Engineering, ISSN 1934-8932, Jan. 2010, Volume 4, No.1 (Serial No. 26)
- g. AP-1049: Staudt-Bickel, *Improvement of CO₂/CH₄ separation characteristics of polyimides by chemical crosslinking*, Journal of Membrane Science 155 (1999) 145-154

II. THE SIMULATIONS IN PRISKE '380, SCHOLZ'S FIGURE 7 AND UNGERANK'S COMPARATIVE EXAMPLE 1

5. Starting in the 1990s, computer simulations, with programs like Aspen and ChemCad, became commonly used in the gas membrane separation field. Computer simulations then and now can help optimize membrane gas separation systems.

6. A POSITA would have understood that simulations can be set up in different ways, with different assumptions, data, and inputs.

7. I am very familiar with simulating membrane gas separation systems, having run several thousands of such simulations in my career, over decades.

'380 Priske patent simulations

8. All the examples in the '380 Priske patent are simulations, as discussed in column 15, lines 34-35, 45.

9. For example, column 22, lines 9-10 in the '380 Priske patent explain that for Example 4 and Comparative Example 4, the "parameters of the simulations are given in Table 9." There is similar language referring to the simulations for

several of the other Examples and Comparative Examples, for example in column 22, lines 45-46, column 23, lines 20-21, and column 23, lines 57-58.

10. In all the examples in the '380 Priske patent, the crude gas flowrates (17) are 1.78 m³/hr or less. In most of the examples, they are 1 m³/hr.

11. For example, Reference Example 1 in the '380 Priske patent states that the "Comparative-Example of WO 2012/000727 A1 was reproduced," which I understand is Ungerank's Comparative Example 1. In Ungerank's Comparative Example 1, the crude gas stream flowrate is 1.78 m³/hr. In Example 1.2 in the '380 Priske patent, the flowrate of the crude gas stream is shown in Table 1 to be 1.78 m³/hr, and Table 3 shows the same crude gas flowrate, 1.78 m³/hr, for Priske's Comparative Example 1. Tables 5 and 7 show crude gas stream flowrates of 1 m³/hr for the corresponding examples. Tables 9 and the similar tables do not expressly show the crude gas stream flowrates, but the crude gas stream flowrates can be calculated by subtracting the flowrates of streams 9 and 10 from the flowrate of stream 5 (with reference to the Figure on the first page of the '380 Priske patent). For example, in Table 9 for Example 4 and Comparative Example 4, the crude gas stream flowrate is 1 m³/hr, as shown in each by subtracting the flowrates of streams 9 and 10 from the flowrates of stream 5.

Scholz simulations

12. All the systems in Scholz are simulations, using GAMS, which a POSITA would have understood from Scholz's Abstract and Scholz discussion throughout the paper.

13. GAMS was not nearly as well-known as Aspen and ChemCad in 2014, but is a simulation method. On pages 2 and 4, Scholz refers to its "simulations."

14. Scholz is an academic paper, and its simulations are based assumptions such as isothermal operation and ideal gases. Ungerank, on the other hand, was from Evonik, a known commercial membrane supplier.

15. For these reasons and others, a POSITA would have viewed Ungerank's Comparative Example 1 as closer to a real-world system than Scholz's Figure 7 systems.

Ungerank Comparative Example 1 simulation

16. I understand Evonik has explained that Ungerank's Comparative Example 1 is a simulation.

17. In my opinion, a POSITA would have understood or strongly suspected Ungerank's Comparative Example 1 is a simulation, for several reasons.

18. Ungerank's Comparative Example 1 is a relatively small system, and many of the concentrations, pressures and flowrates are provided with three

significant digits, which is typical of a simulation. In contrast, in a real world system, it would be exceedingly difficult and unusual to measure these variables with such precision. A POSITA would have understood that in real world systems, these variables would typically be measured with only one or two significant digits. For example, in laboratory experiments, flowrate measurements are typically off by as much as 2%, and in commercial systems, flowrate measurements are typically off by as much as 5%.

19. Also, the mass balances in Ungerank's Comparative Example 1 sum almost exactly. For example, the crude gas stream flowrate is $1.78 \text{ m}^3/\text{hr}$. The second retentate stream flowrate is $0.895 \text{ m}^3/\text{hr}$ and the third permeate stream flowrate is $0.885 \text{ m}^3/\text{hr}$, which sum to exactly $1.78 \text{ m}^3/\text{hr}$ with three significant digits. A POSITA would have understood this would be highly unlikely in any real world experiment or commercial system, but typical and indicative of a simulation.

The membrane properties and surface areas in Ungerank's Comparative Example 1's simulation and in Scholz's simulations

20. A POSITA would have understood that simulations of membrane gas separation systems do not have actual membranes or membrane modules. Instead, simulations are based on membrane properties (such as selectivity, permeance and surface area) that are either input into the simulation or calculated in the simulation. Some simulation methods also allow configuration properties to be

input, for example, cross-current flow for spiral wound membrane configurations or counter-current flow for hollow fiber membrane configurations.

21. Ungerank's Comparative Example 1 does not specify if the membranes' areas were input into the simulation or calculated in the simulation.

22. Ungerank's Comparative Example 1 states that "Each stage consisted of a hollow fiber membrane module consisting of hollow polyimide fibers from UBE (NMB01 A type)." In reality, since Ungerank's Comparative Example 1 is a simulation, properties of polyimide membranes were used, not actual membranes or membrane modules. In other words, in Ungerank's Comparative Example 1, identical polyimide membrane properties (aside from the surface areas, which are not specified) were used in each stage.

23. A POSITA would have understood that with the crude gas flowrate of 1.78 m³/hr in Ungerank's Comparative Example 1, any type of commercial membrane modules would have been far too large for that system.

24. Similar to Ungerank's Comparative Example 1, in the Scholz Figure 7 simulations, a POSITA would have understood that identical membrane properties (except for the surface areas) were used in each stage. Scholz explains on page 5 that the Figure 7 systems are based on the "properties of cellulose acetate membranes with a selectivity of 20." On page 6, Scholz also states "For a

membrane reflecting the characteristics of a cellulose acetate membrane with a CO₂/CH₄ selectivity of 20 the results are depicted in Fig. 7.”

25. Ungerank’s Comparative Example 1 states the “membranes used exhibit a moderate mixed gas selectivity for carbon dioxide over methane of 20.” A POSITA would have understood the terms “low,” “moderate” and “high” were not exact terms in 2014 with respect to selectivity, and their meanings depended on the context and the time.

26. The membranes used in Ungerank’s Comparative Example 1 simulation have lower selectivity (“mixed gas selectivity for carbon dioxide over methane of 20”) than the membranes used in Ungerank’s Examples 1-4, and a POSITA would have understood this from reading Ungerank. A POSITA would have also understood in 2014 that Ungerank’s Comparative Example 1 was from 2011, and more higher-selectivity membranes had become available in the three years in between. For example, in 2014, on page 9 of Scholz, the membranes in Figure 7 “with a CO₂/CH₄ selectivity of 20” are described as “low selective.”

27. For these reasons, a POSITA in 2014 would have understood the membranes in Ungerank’s Comparative Example 1 to be lower-selectivity—or in other words, as membranes with average or standard selectivity for biogas—as opposed to higher-selectivity membranes at the time, for example, the “highly selective” membranes that are the bases for Ungerank’s Examples 1-4.

28. In my deposition, I discussed how a POSITA “might have already figured out” that in Ungerank’s Comparative Example 1, stage 2’s membrane surface area was larger than stage 1’s membrane surface area. For a fixed feed gas flow rate across the membrane surface, as the membrane becomes larger, more of the permeable component (CO_2) passes through the membrane. This depletes the feed gas of the permeable component (CO_2) and enriches it in the retained component (CH_4). Thus, as the membrane becomes larger, a disproportionate amount of the retained component (CH_4) begins to pass through the membrane. The net effect is that a larger membrane area results in lower concentrations of the permeable component (CO_2) in the permeate stream. Conversely, smaller membranes result in higher concentrations of the permeable component (CO_2) in the permeate stream. In terms of Ungerank’s Comparative Example 1, that means that a smaller membrane in stage 1 would result in a higher concentration of CO_2 in the first permeate stream (6).

29. A POSITA would have understood that in Ungerank’s Comparative Example 1, to achieve the relatively high concentration of CO_2 (99.0%) in the third permeate stream (11), a relatively large driving force would be needed in stage 3, meaning there must be a relatively high concentration of CO_2 in the first permeate stream (6). A POSITA would therefore have understood that stage 1’s membrane must be relatively small.

30. For stage 2, the same principle applies. However, the second permeate stream (9) is recycled. In other words, a POSITA would have understood that in Ungerank's Comparative Example 1, to achieve the relatively high concentration of methane (98.5%) in the second retentate stream (8), a relatively large membrane would be needed in stage 2 to allow much of the permeable component (CO₂) to pass through it. A relatively large membrane in stage 2 would also allow a relatively large amount of methane to pass through stage 2's membrane into the second permeate stream (9), but that stream, including the methane therein, is recycled and combined with the crude gas stream (17) so that it processed again.

31. Scholz's Figure 7 systems had larger stage 2 membrane areas than the stage 1 membrane areas. That was consistent with the intuition a POSITA would have had about the relative sizes of Ungerank's Comparative Example 1's stage 2 and stage 1 areas. This would have given a POSITA additional reason to believe it was appropriate to look to Scholz's Figure 7 to better understand the relative surface areas of stages 1 and 2 in Ungerank's Comparative Example 1.

32. As I discussed in my deposition, for a POSITA looking for more information on the relative sizes of the membranes in Ungerank's Comparative Example 1, looking to Scholz would have been "the cheapest sort of research." In other words, a POSITA would have expeditiously and inexpensively looked to the relative surface areas in Scholz's Figure 7, for the reasons I have explained.

Qi and Henson's Figure 9 from 2000

33. In my opinion, in looking to sources to better understand the relative surface areas in Ungerank's Comparative Example 1, a POSITA in 2014 would not have looked to Qi and Henson's Figure 9, which was from 14 years earlier—as opposed to looking to Scholz's Figure 7 systems from 2014.

34. The system in Qi and Henson's Figure 9 has components in the crude gas stream (such as hydrogen sulfide and ethane) that are not in the crude gas streams in Ungerank's Comparative Example 1 and Scholz's Figure 7 (which both have the same components (methane and carbon dioxide)). Also, Qi and Henson's Figure 9 has two compressors, as opposed to Ungerank's Comparative Example 1 and Scholz's Figure 7, which both have one compressor and very similar system configurations.

35. A POSITA would have found Scholz's Figure 7 systems more similar to Ungerank's Comparative Example 1 than Qi and Henson's Figure 9, both in terms of the time frame and the systems' characteristics.

III. THE “CAPACITY” RATIOS IN UNGERANK'S COMPARATIVE EXAMPLE 1 AND IN SCHOLZ'S FIGURE 7 ARE EQUAL TO THE SURFACE AREA RATIOS

36. Claim 1 in the Priske '380 patent requires that “the total capacity of the gas separation membranes in the retentate separation stage is higher than the total capacity of the gas separation membranes in the feed stream separation stage,

the total capacity being measured for nitrogen Grade 4.8 under standard conditions as defined in the description.”

37. The Priske '380 patent defines “capacity” in column 2, lines 53-57 “as the product of the membrane surface and the permeance of the membrane at operating temperature that is determined for nitrogen (Grade 4.8) under standard conditions.”

38. In other words, as I explained in my February 22, 2024 declaration, the reference in Priske’s claim 1 to measuring capacity under certain conditions is the result of and depends on measuring the membranes’ permeances at those conditions.

39. When membranes have identical properties (aside from their surface areas), the membranes’ permeances, measured at any set of conditions (i.e., as long as the conditions are the same), are necessarily identical.

40. In Scholz’s Figure 7 and in Ungerank’s Comparative Example 1, since all those systems have identical membrane properties in each system (except for their surface areas), the membranes’ permeances in each system, measured “for nitrogen Grade 4.8 under standard conditions as defined in the description,” are necessarily identical.

41. As a result, in Scholz’s Figure 7 and in Ungerank’s Comparative Example 1, the capacity ratio of the membranes in any two stages is equal to the

surface area ratio in those two stages, because the equal permeances cancel each other out:

$$\frac{Capacity2}{Capacity1} = \frac{area2}{area1} * \frac{permeance}{permeance}$$

42. In the two systems in Scholz's Figure 7, the membrane surface areas in stage 2 are larger than in stage 1. Accordingly, for the reasons explained above, those systems meet the capacity requirement in Priske '380 claim 1.

43. For the same reasons, if in Ungerank's Comparative Example 1, the membrane in stage 2 were larger than in stage 1, that would also meet the capacity requirement in Priske '380 claim 1.

IV. A POSITA IN 2014 WOULD HAVE HAD REASONS TO USE LOWER-SELECTIVITY MEMBRANES

44. In my February 22, 2024 declaration, I discussed reasons a POSITA in 2014 would have used lower selectivity membranes, for example, on offshore platforms and in situations where operators desired to minimize upfront costs.

45. A paper I co-authored that was published in mid 2014, *Pressure ratio and its impact on membrane gas separation processes*, explains an additional reason a POSITA would have used lower-selectivity membranes. AP-1046. As explained in the paper, some applications cannot be economically operated with high feed-to-permeate pressure ratios, and in those situations, the "limited pressure ratio means that the optimum membrane may not be the one with the highest

selectivity; rather, a membrane with lower selectivity, but tailored to the pressure ratio, may result in a more economical process.”

46. A paper I co-authored that was published in 2008, *Natural Gas Processing with Membranes: An Overview*, explains an additional reason a POSITA would have used high permeance, smaller, lower selectivity membranes. AP-1047. As explained in the paper, “High skid costs arise because of the many pressure vessels, pipes, flanges, and valves that are required. One way to reduce membrane skid cost is to increase the permeance of the membranes, allowing a smaller membrane area to be used to treat the same volume of gas. All membrane manufacturers pursue this approach. Increasing the feed gas pressure also reduces the membrane area required, and, hence, skid size, but at the expense of larger compressors (higher capital cost) and increased energy consumption (higher operating cost).”

V. RESPONSES TO OTHER EVONIK STATEMENTS

Methane content in third permeate stream in Ungerank’s Comparative Example 1

47. In its Patent Owner Response, I understand that Evonik contends that a POSITA would have been dissuaded from looking to Ungerank’s Comparative Example 1 because it has 1% methane in the third permeate stream.

48. There are different applications for carbon dioxide, with varying specifications, so I do not agree that a POSITA would have found 1% methane in a

carbon dioxide stream to be categorically problematic.

49. If 1% methane in a carbon dioxide stream were concerning to a POSITA, it is routine in industrial systems and relatively inexpensive to pass streams containing small amounts of organic compounds through a flare, thermal oxidizer or other equipment to decrease the organic concentration.

Priske '380 claim 12 – pure-gas versus mixed-gas selectivity

50. Claim 12 in the Priske '380 patent requires that “the gas separation membranes in the feed stream separation stage have a pure gas selectivity for CO₂/CH₄ of from 20 to 100.” Ungerank’s Comparative Example 1 states that the “membranes used exhibit a moderate mixed gas selectivity for carbon dioxide over methane of 20.”

51. I explained in my February 22, 2014 declaration that in the types of membranes at issue, pure-gas selectivity is higher than mixed-gas selectivity, but a POSITA would have understood that the pure-gas selectivity for the types of membranes in Ungerank’s Comparative Example 1 would not be more than 500% higher than the mixed-gas selectivity.

52. Evonik seems to dispute this in its Patent Owner Response, in part because the data I cited on pure-gas selectivities versus mixed-gas selectivities was for cellulose acetate membranes, but Ungerank’s Comparative Example 1 is based on properties of polyimide membranes.

53. I do not agree with Evonik's criticisms. For the types of membranes under discussion, a POSITA would clearly understand that the pure gas selectivities would not be more than five times greater than the mixed gas selectivities.

54. Ungerank's Comparative Example 1 is based on polyimide membrane properties including a "mixed gas selectivity for carbon dioxide over methane of 20," and, for example, in 2010, in "*Polymeric Membranes Used in Natural Gas Processing to Separate Carbon Dioxide: the Facing Problems and Solutions*," Bhatti reported CO₂/CH₄ pure gas selectivities for polyimide membranes of 29 to 57. AP-1048.

55. Similarly, in 1999, in *Improvement of CO₂/CH₄ separation characteristics of polyimides by chemical crosslinking*, Staudt-Bickel reported pure gas and mixed gas selectivities for CO₂/CH₄ for several types of polyimide membranes, in Table 3 and Figure 6. At 20 atmospheres (approximately the feed pressure in Ungerank's Comparative Example 1), the mixed gas selectivities in Figure 6, for example, for the 6FDA-*m*PD/DABA 9:1 copolyimides are approximately 45 to 50; and their pure gas selectivities in Table 3 are 63.3 and 65.3. AP-1049.

Priske '380 claim 9

56. Claim 9 in the Priske '380 patent requires that "the combined gas

volume recycled with the second permeate stream and the third retentate stream is from 65 to 85% of the volume of the crude gas stream.” Ungerank’s Comparative Example 1 states that the “sum of recycled gas streams (9) and (10) is ... 86.7% based on the amount of gas to be separated supplied.”

57. The difference between 86.7% and 85% ($((86.7 \div 85) - 1) * 100$) is 2%.

58. I have been asked to calculate the differences between (i) 0.3% and 0.31% ($((0.31 \div 0.3) - 1) * 100$), which is 3.3%; (ii) 0.3% and 0.25% ($((0.3 \div 0.25) - 1) * 100$), which is 20%; (iii) 0.8% and 0.75% ($((0.8 \div 0.75) - 1) * 100$), which is 6.7%; and (iv) 0.94% and 0.8% ($((0.94 \div 0.8) - 1) * 100$), which is 17.5%.

I declare under penalty of perjury under the laws of the United States of America that the forgoing is true and correct. I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief our believe to be true; and further that these statements were made with the knowledge that 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: April 11/2025

By: Richard W. Baker
Richard W. Baker