

Case Number: T 0283/95 - 3.4.2

D E C I S I O N
of the Technical Board of Appeal 3.4.2
of 3 July 1996

Appellant: PRAXAIR INC.
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Respondent: Bayer AG, Leverkusen
(Opponent) Konzernverwaltung RP
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Decision under appeal: Decision of the Opposition Division of the European Patent Office posted 6 February 1995 revoking European patent No. 0 297 542 pursuant to Article 102(1) EPC.

Composition of the Board:

Chairman: E. Turrini
Members: C. Black
M. Lewenton

Summary of Facts and Submissions

- I. European patent No. 0 297 542 (Application number 88 110 380.8) was revoked by a decision of the Opposition Division on the ground that its subject-matter, although novel, did not involve an inventive step.
- II. In reaching its decision the Opposition Division took into account the following documents filed by the opponents 1 and 2:
- D1 H. W. Habgood, "Adsorptive and gas chromatographic properties of various cationic forms of zeolite X", Canadian Journal of Chemistry, Vol. 42(1964), pages 2340 to 2350
- D2 US-A-3 140 933
- D3 Leaflet UOP, MOLSIV Adsorbent Type 13X
- D4 US-A-2 882 244
- D5 DD-A-0 154 690
- D6 R. M. Barrer et al. in Ion exchange and the thermodynamic properties of intracrystalline sorption, 1958, pages 464 to 483
- D7 Neftekhimiya, 19691 IX, (18), pages 790 to 795, G. V Tsitsishvihi et al.

D8 J. Chem. Phys. 78(12), 15. June 1983, Tadashi
Tokuhiro et al.,

and the following documents filed by the patent proprietor:

D9 Molecular sieves, Paper read at the conference held
at the School of Pharmacy (University of London) 4-
6th April, 1967, Habgood et al. "Cyclopropane to
Propylene over X- and Y-zeolites"

D10 US-A-3 140 932.

III. The present appeal lies against this decision.

IV. Oral proceedings were appointed. In a communication pursuant to Article 11(2) RPBA the Board expressed its provisional opinion on the case, which was that the claimed subject-matter arguably did not involve an inventive step having regard to the disclosure in D2 taking into account the common general knowledge of the person of average skill in the art. However the lapse of nearly 23 years between the publication date of D2 and the priority date of the patent in suit seemed to require some explanation.

V. With a response to the Board's communication, the appellant (patent proprietor) filed affidavits from the inventor and an independent expert, seeking to meet the points raised by the Board.

VI. At the oral proceedings the appellant requested that the decision under appeal be set aside and the patent maintained in amended form on the basis of sets of claims

according to a main or auxiliary request. The respondents (opponents 1 and 2) requested that the appeal be dismissed.

VII. Claim 1 according to the main request reads as follows:

"Pressure swing adsorption process for selectively separating nitrogen from a gas mixture consisting essentially of nitrogen and oxygen which comprises contacting said gas mixture with a crystalline zeolite X adsorbent

- having a framework $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio from 2.0 to 2.5,
- having at least 90 per cent of its AlO_2 tetrahedral units associated with lithium cations, and
- exhibiting a greater difference in loading for nitrogen, measured at room temperature and at an adsorption pressure of 200 kPa (1500 torr) and at a desorption pressure of 20 kPa (150 torr), than the difference in loading exhibited under the same conditions of measurement by an otherwise identical adsorbent material of which only 86 per cent of its AlO_2 tetrahedral units are associated with lithium cations,

at a process temperature of from 15 to 70°C and at a process pressure swinging between a higher adsorption pressure and a lower desorption pressure, said pressures being within the range of 6.7 kPa to 1333 kPa (50 torr to 10 000 torr)."

Claim 1 according to the auxiliary request differs from the above in that "at least 90 per cent" is replaced by "at least 94 per cent".

VIII. The argumentation of the appellant may be summarised as follows:

The object of the invention is to provide a pressure swing adsorption (hereinafter PSA) process having

- high nitrogen selectivity over oxygen at the adsorption pressure,
- high delta nitrogen loading i.e. loading difference between the adsorption pressure and the desorption pressure in pressure swing adsorption process cycles, and
- high nitrogen loading
- within particularly useful temperature and pressure ranges.

The average skilled person, faced with such a problem, would, from the disclosure in D2, rather be led away from the claimed subject-matter.

In the first place, most of the data in D2 relates to operation at sub-zero temperatures, in particular -78°C , and from the data in Tables I and II it can be derived that any rise in separation factor with increasing temperature levels off at approaching 0°C . This in combination with the

known reduction in capacity, or loading, with increasing temperature (also demonstrated by the data in Tables I and II) would deter the skilled person from investigating operation at the temperature range required by Claim 1.

Moreover, data from Praxair records, depicting the type of data known in this field in 1987 or before, indicates that at 23°C, the nitrogen loading increases only marginally, if at all, with increasing exchange for the zeolites CaNaX, LiCaX and LiSrX, illustrated by Exhibits 2, 3 and 4 accompanying the letter dated 3 June 1996. Further data from Praxair records (Exhibits A and B) accompanying the letter dated 24 June 1996, shows that the nitrogen loading did not rise with increasing exchange at -80°C, and that X zeolites with 90 per cent and 94.4 per cent lithium exchange were inferior to zero exchanged sodium zeolite X in terms of nitrogen loading.

Column 5, lines 41 to 45 of D2 indeed suggests that higher lithium exchange confers higher selectivity. However, the average skilled person is aware that a higher selectivity does not automatically translate into a more effective separation, because at the low desorption pressure, high selectivity makes it more difficult to desorb nitrogen from the zeolite. D2 is in fact silent concerning delta loading.

The only reference in D2 to operation at ambient temperatures occurs in column 6, lines 14 to 20. This paragraph however follows on from a paragraph describing a temperature swing adsorption (TSA) process and the average skilled person will interpret the reference to temperatures as high as 30°C as relating to such a process.

The average skilled person can moreover be assumed to be aware of D10 as well as D2 since these are parallel patents. From D10 he learns that both barium and strontium exchanged zeolite X have a higher separation factor than that of lithium exchanged zeolite X, so that the latter would not be the zeolite of choice for further investigation.

Therefore for the average skilled person the disclosure in D2 held no promise, not the least in view of the cost of achieving higher exchange rates and this is one explanation of the gap of 23 years between the publication of D2 and the priority date of the patent in suit.

The gist of the respondents' counter-argumentation is that the feature "exhibiting a greater difference in loading ... associated with lithium cations" has no basis in the description, because the passage on page 6, lines 35 to 46, stated to provide such support, only compares 94 per cent and 99 per cent lithium exchanged zeolite X with zero exchanged zeolite X and not with 86 per cent lithium exchanged zeolite X. Otherwise the only difference between the subject-matter of Claim 1 and the disclosure in D2 is that D2 does not in words disclose that at least 90 per cent of the AlO_2 tetrahedral units is associated with lithium ions. D2 does however disclose 40 per cent, 86 per cent and approaching 100 per cent, and points to the benefits associated with high degrees of exchange. As to the twenty three year gap between D2 and the priority date of the patent in suit, D2 would appear to have been ahead of its time.

Reasons for the Decision

1. The appeal is admissible.
2. Claim 1 according to the main request is derived from a combination of Claims 1, 2, 3, 5 and 6 of the granted patent. The further feature "exhibiting a greater difference ... associated with lithium ions" is stated by the appellant to have a basis in the description, page 6, lines 35 to 46. The respondents dispute this, arguing that in the table contained in the cited passage, no comparison is made with 86 per cent exchanged zeolite X as is required by this feature of Claim 1. The Board notes that lines 35 to 39 refer to superiority of highly exchanged LiX over 86 per cent exchanged LiX; however since the appeal fails for other reasons, the Board has elected not to decide on this issue.

The basis for the limitation contained in Claim 1 according to the auxiliary request ("at least 94 per cent" instead of "at least 90 per cent") is to be found in the table in the above-cited passage.

3. Novelty of the claimed subject-matter (both requests) will be apparent from the discussion of inventive step.
4. For the evaluation of inventive step, D2 is undisputedly the most appropriate prior art. It discloses a PSA process - see the paragraph

bridging columns 2 and 3 which indicates both PSA and TSA, and the paragraph bridging columns 5 and 6 in conjunction with the following paragraph, the latter indicating that TSA is an alternative to PSA as previously described. D2 also discloses the separation of nitrogen from oxygen, e.g. in air - see e.g. the title. Zeolite X is stated to be the preferred adsorbent - column 4, lines 60 to 62, and has a $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of 2.5 ± 0.5 to 1 (see the formula in column 5). The temperature range disclosed in D2 is $+30^\circ\text{C}$ to -150°C (see e.g. the claims) overlapping that required by Claim 1 (both requests) and the pressure range disclosed is 0.5 to 5 atmospheres, which embraces that required by Claim 1.

D2 further discloses that the benefit of improved selectivity is related to the degree of exchange with lithium ions (column 5, lines 41 to 44) and that one would therefore prefer to have relatively high degrees of exchange approaching 100 per cent. D2 goes on to state that this degree of exchange is not always achievable for various reasons and that a lithium exchange of as low as 40 per cent can improve the nitrogen oxygen separation factor. D2 nevertheless discloses 86 per cent exchanged lithium zeolite X in Tables I and II.

5. The subject-matter of Claim 1 therefore differs from the disclosure in D2 (a) in that the $\text{SiO}_2/\text{Al}_2\text{O}_2$ molar ratio is restricted to the range 2.0 to 2.5 (b) in requiring that at least 90 per cent of the AlO_2 tetrahedral units are associated with lithium cations, this degree of exchange not being specifically disclosed in D2 and (c) in that the

claim includes the feature seeking to emphasise the improved delta loading.

6. The Board's assessment of the problem which the invention seeks to solve is as follows. Difference (c) - see paragraph 5 above - reflects the importance the appellant attaches to the corresponding aspect of the object of the invention as set out in paragraph VIII above. In the Board's view all four aspects of the said object of the invention are obvious desiderata for the person of average skill in the art. It would seem to be self-evident that if nitrogen is to be separated from oxygen the adsorbent should show a high selectivity for nitrogen over oxygen. It is equally self-evident that the adsorbent should have a high capacity, or loading, for nitrogen; in this respect it is pointed out that an adsorbent with low capacity for nitrogen and zero capacity for oxygen could be said to be 100 per cent selective for nitrogen, but a separation process using such an adsorbent would not be efficient. Again a high capacity for nitrogen during the adsorption step is of little value if the nitrogen is not readily released in the desorption step, so that the average skilled person is aware of the need for high delta nitrogen loading. Finally it is clearly more convenient to operate at ambient temperatures than at sub zero temperatures.

In view of the foregoing the Board sees the object of the invention as seeking to improve on the PSA process known from D2 in respect of the above-mentioned desiderata, and the question to be answered is whether an inventive step can be seen in the differences between the subject-matter of Claim 1 and the disclosure in D2.

7. The appellant argues that the framework ratio range of 2.0 to 2.5 to which difference (a) relates results in an increase in nitrogen loading and separation factor which is greater than could be expected from the presence of the increased number of cations in the lower portion of the range disclosed in D2. In the Board's view the representation of the $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio as 2.5 ± 0.5 is in itself a disclosure of the range of 2.0 to 2.5 required by Claim 1. But even if it could be argued that this were not the case, the sub-range cannot be seen as a selection from the wider range disclosed in D2, since it does not meet the requirements for a selection. These requirements are set out in a number of decisions of the Boards of Appeal and summarised in Case Law of the Boards of Appeal I.C.4.2. In particular the sub-range is not narrow in comparison with that disclosed, nor does it meet the requirement of being sufficiently far removed from the known range. Therefore this difference does not contribute to inventivity.

8. The appellant further argues, in respect of difference (b), that the average skilled person would interpret "approaching 100 per cent" (see paragraph 4 above) in the light of the disclosed value of 86 per cent (description relating to Tables I and II) and, in view of the cautionary statement that high degrees of exchange are not always achievable, would not be led to investigate the use of zeolites having at least 90 per cent of the AlO_2 tetrahedral units associated with lithium atoms.

In the Board's view there is nothing in D2 to suggest to the average skilled person that 86 per cent is an upper practicable limit. It is moreover apparent from D6

(Table I), D7 (Table I and the references throughout to zeolite X) and D8 (calculation from data in Table I) that although substitution of lithium ions for sodium ions in zeolite X was time consuming, there were no insuperable difficulties in achieving degrees of exchange of over 90 per cent up to about 100 per cent before the priority date of the patent in suit. Accordingly, in the Board's opinion, the average skilled person will interpret D2, column 5, lines 41 to 45 as it stands, that is, the greater the degree of exchange, the higher the selectivity, or separation factor. D2 further discloses, in column 6, lines 14 to 20, that because of the high separation factors and high capacity afforded by the described zeolites, the adsorption step may be carried out at ambient temperatures as high as 30°C. The teaching of D2 therefore leads the average skilled person to investigate the use of up to 100 per cent lithium exchanged zeolite X in a PSA process for separating nitrogen and oxygen in the reasonable expectation of achieving an improvement in respect of capacity and separation factor, so that this feature is an obvious extension of what is specifically disclosed in D2.

9. It is true that apart from a hint in column 6, lines 45 to 57 (see paragraph 11 below) D2 is silent concerning delta loading, which as previously stated is reflected in Claim 1 in the feature corresponding to difference (c). As regards the desorption step, D2 discloses that the claimed process may be carried out as disclosed in another cited application, with desorption of nitrogen at sub-atmospheric pressures. The drafter of D2 presumably considered that no more needed to be said about the desorption step, which explains the silence concerning delta loading.

What Claim 1 requires is that the delta loading is higher for 90 per cent lithium exchanged zeolite X than for 86 per cent exchanged zeolite X. However, as has been shown in paragraphs 7 and 8 above, a PSA process utilising a zeolite X having the $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio and degree of lithium exchange required by Claim 1 is obvious from the teaching of Claim 1. In the Board's opinion a zeolite X meeting these requirements will also inherently meet the delta loading requirement. This is supported by the statement in the patent in suit, page 6, lines 34 to 39 referring to the superiority of the highly exchanged LiX over known lithium exchanged zeolites, not only in capacity for nitrogen, but also in terms of delta loading, which indicates that high capacity and high delta loading are both inherent properties of highly exchanged LiX zeolites.

10. The operating pressures and temperatures of the claimed PSA process have been referred to in paragraph 4 above. The pressure range required by Claim 1 is not seen as an inventive selection over that disclosed in D2, and this has not been argued, but merely a convenient range the choice of which falls within the competence of the average skilled person. The temperature range of 15 to 70°C indeed extends beyond that disclosed in D2 (ambient temperatures as high as 30°C) but again it has not been argued that there could be anything inventive in operating above 30°C. D2 gives the teaching that the disclosed zeolites permit operating at convenient ambient temperatures and the skilled person is free to establish an upper maximum temperature, the main constraint being the known reduction in capacity with increasing temperature.

11. The appellant has sought to show that the average skilled person would find little, if any, encouragement to operate according to the requirements of Claim 1. The Board's view is that the person of average skill in the art reading D2 will pay little, if any, attention to data relating to sub-zero temperatures or to the data illustrated in Exhibits 2, 3 and 4 for different zeolites. The teaching which is of interest to the said skilled person is that the separation factor and capacity of the disclosed lithium exchanged zeolites are so high that adsorption at ambient temperatures is possible, the more so the higher the exchange rate, and in investigating the teaching of D2 further he would confine his attention to operation at ambient temperature.

In this respect the Board's interpretation of the paragraph in column 6, lines 14 to 20 of D2 is that it is a general statement about the disclosed zeolites and is not restricted to the TSA process which is the subject of the previous paragraph.

The Board accepts that the skilled person aware of D2 will also be aware of D10, which is a parallel parent having the same applicant, assignee, application date and publication date, and, **mutatis mutandis**, a similarly worded description. It is true that D10 discloses separation factors for barium and strontium exchanged zeolites which are higher than that disclosed in D2 for 86 per cent lithium exchanged zeolite X. Against this however can be set the higher loading capacity of the lithium exchange zeolite X. Further one piece of disclosure to be found in D2 and not in D10 occurs in column 6, lines 21 to 57. This

relates to the advantageous relationship between separation factor and temperature for lithium exchanged zeolite X, and arguably hints at a satisfactory delta loading resulting therefrom. The disclosure in D2 is accordingly at least as promising as that of D10.

12. All in all, the Board's view is that the grant of a patent on the basis of Claim 1 would prevent the average skilled person from doing what is no more than is derivable from the teaching of D2. The Board has considered the 23 year gap between the publication date of D2 and the priority date of the patent in suit, and also the apparent commercial success related to the claim process. These factors alone are not sufficient to demonstrate the presence of an inventive step, and a plausible explanation of the time gap is the cost of manufacturing highly exchanged zeolites, which can be assumed to have become relatively lower with advances made over the years. The appellant's main request must therefore be rejected.
13. The same considerations apply to Claim 1 according to the auxiliary request. D2 refers to degrees of exchange approaching 100 per cent and no inventive step can be seen in the limitation to at least 94 per cent as compared to at least 90 per cent.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar: The Chairman:

P. Martorana

E. Turrini