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Datasheet for the decision of 30 May 2008

Case Number: T 0615/05 - 3.3.06

Application Number: 95309112.1

Publication Number: 0718024

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Language of the proceedings: EN

Title of invention:
Removal of carbon dioxide from gas streams

Patentee:
Linde, Inc.

Opponent:
L'AIR LIQUIDE, S.A.
Praxair Technology, Inc.
AIR PRODUCTS AND CHEMICALS, INC.

Headword:
Removal of carbon dioxide/LINDE

Relevant legal provisions:
EPC Art. 56

Keyword:
"Inventive step: no (all requests)"

Decisions cited:
-

Catchword:
-

Case Number: T 0615/05 - 3.3.06

DECISION
of the Technical Board of Appeal 3.3.06
of 30 May 2008

Appellant:
(Patent Proprietor)

Linde, Inc.
575 Mountain Avenue
Murray Hill, NJ 07974 (US)

Representative:

Wickham, Michael
The Linde Group
Intellectual Property Department
The Priestley Centre
10 Priestley Road
The Surrey Research Park
Guildford, Surrey GU2 7XY (GB)

Respondents:
(Opponent)

L'AIR LIQUIDE, S.A.
pour l'étude et l'exploitation
des procédés Georges Claude
75, quai d'Orsay
F-75321 Paris (FR)

Representative:

Pittis, Olivier
L'Air Liquide, S.A.
Service Brevets & Marques
75, Quai d'Orsay
F-75321 Paris Cedex 07 (FR)

(Opponent)

Praxair Technology, Inc.
39 Old Ridgebury Road
Danbury, Ct. 06810-5113 (US)

Representative:

Schwan - Schwan - Schorer
Patentanwälte
Bauerstrasse 22
D-80796 München (DE)

(Opponent) AIR PRODUCTS AND CHEMICALS, INC.
7201 Hamilton Boulevard
Allentown, PA 18195-1501 (US)

Representative: Burford, Anthony Frederick
Beck Greener
Fulwood House
12 Fulwood Place
London WC1V 6HR (GB)

Decision under appeal: **Decision of the Opposition Division of the European Patent Office posted 4 April 2005 revoking European patent No. 0718024 pursuant to Article 102(1) EPC 1973.**

Composition of the Board:

Chairman: P.-P. Bracke
Members: G. Dischinger-Höppler
U. Tronser

Summary of Facts and Submissions

I. This appeal is from the decision of the Opposition Division to revoke the European patent No. 0 718 024 relating to the removal of carbon dioxide from gas streams.

II. Three notices of opposition had been filed against the granted patent, wherein the Opponents sought revocation of the patent on the grounds of, inter alia, Article 100(a) EPC for lack of inventive step (Article 56 EPC). The oppositions were based, amongst others, on documents

D3 S.S. Khvoshchev et al., "Adsorption of Carbon Dioxide on Zeolites in relation to the Content of Cations in their Crystal Lattices" in Russian Journal of Physical Chemistry, 42(1), 1968, pages 87 to 90;

D4 EP-A-0 284 850;

D9 R.M. Barrer, "Zeolites and Clay Minerals as Sorbents and Molecular Sieves", 1978, Academic Press, London · New York, pages 206 to 216;

D13 EP-A-0 196 103;

D14 C.G. Coe et al., "Molecularly Engineered, High-Performance Adsorbent", in Perspectives in Molecular Sieve Science, 1988, American Chemical Society, Chapter 30, pages 470 to 491;

and

D20 G.H. Kühl, "Crystallization of low-silica faujasites (SiO_2/Al_2O_3~2.0)", published in 1987, pages 451 to 457.

III. The decision under appeal was based on amended claims according to a main and five auxiliary requests.

IV. In its decision, the Opposition Division revoked the patent for lack of inventive step of the subject-matter of Claim 1 of all requests in view of the disclosure of document D4 as the closest prior art in combination with the teaching of document D3.

V. This decision was appealed by the patent Proprietor, now Appellant, who filed - under cover of a letter dated 11 August 2005 setting out its statement of grounds of appeal - documents A3 to A5 and amended sets of claims in a new main request and in five auxiliary requests.

The Opponents, now Respondents, filed amongst others document

D29 US-A-4 603 040.

VI. Upon requests made by all parties, oral proceedings before the Board of Appeal were held on 30 May 2008, in the course of which the Appellant withdrew the main request as well as the second and fourth auxiliary requests and renumbered the former first auxiliary requests as its main request and the former third and fifth auxiliary request as its first and second auxiliary request, respectively.

Claim 1 of the main request reads:

"1. A method of removing carbon dioxide from a gas stream comprising carbon dioxide and gases less polar than carbon dioxide, comprising subjecting said gas stream to adsorption, using type X zeolite having a silicon to aluminium atomic ratio up to 1.15, at a temperature in the range of -50 to 80°C and a carbon dioxide partial pressure not greater than 5 mbar, wherein the adsorption is part of a temperature swing adsorption process and wherein the exchangeable cations of the type X zeolite are sodium ions."

Claim 1 of both auxiliary requests differs therefrom in that the term "up to 1.15" has been replaced, namely by "in the range of 1.0 to 1.1" (first auxiliary request) and by "in the range of 1.0 to 1.02" (second auxiliary request).

VII. The Appellant, orally and in writing, made in essence the following submissions:

- The Appellant's own experimental results set out in the examples of the patent in suit and in documents A3 to A5 showed a surprising increase of carbon dioxide (CO_2) adsorption obtained by the claimed process at low CO_2 partial pressures as compared with the comparative example of document D4 disclosing a sodium exchanged zeolite X (NaX) having a Si/Al atomic ratio of 1.25 as adsorbent.

- Document D4 suggested another solution to the technical problem of improving CO_2 adsorption, namely the use of barium exchanged zeolite X (BaX) instead of NaX.

- Therefore, the claimed invention was not the result of a "one-way-street" situation. Instead, the technical problem actually solved by the subject-matter claimed in the main and first auxiliary requests in view of the disclosure of document D4 consisted in an improvement at low CO_2 partial pressures of the CO_2 adsorption of a magnitude that was not predictable from the prior art.

- There was nothing in the prior art to suggest solving this problem by using sodium exchanged low silica zeolite X (NaLSX) having a Si/Al atomic ratio up to 1.15, let alone 1.10 for the following reasons:

- At low CO_2 partial pressures, document D3 did not show a significantly improved CO_2 adsorption for NaLSX having a Si/Al atomic ratio of 1.11 instead of 1.18 nor indicate an improvement within the general group of NaX zeolites with decreasing Si/Al atomic ratios.

- A skilled person would not have considered in figure 1 of document D3 the curve for NaLSX of the ratio 1.11 since it had a wrong shape. Typical CO_2 adsorption curves were those shown in document A3. Therefore, the skilled person was not influenced by document D3. This was corroborated by the long period of time between the publication date of document D3 and the priority date of the patent.

- Further, the skilled person would have known from prior art documents that low silica zeolite X (LSX) was difficult to prepare so that any improved CO_2 uptake would not have been considered advantageous.

- Concerning the second auxiliary request, the technical problem actually solved consisted in an enhanced economic attractiveness of the claimed process due to the reduced need for refrigeration. However, the prior art did not foreshadow the claimed solution of that problem.

VIII. The Respondents, orally and in writing, argued that the claimed subject-matter differed from the comparative example shown in document D4 only in that NaLSX having a Si/Al atomic ratio of up to 1.15 was used instead of NaX having a Si/Al atomic ratio of 1.25. The technical problem solved by this difference was to improve the CO_2 adsorption. However, it was known from documents D3, D9 and D14 that the adsorption capacity of zeolite X increases with decreasing Si/Al atomic ratio. Concerning the availability of NaLSX at the priority date of the patent in suit, the Respondents referred amongst others to documents D13, D20 and D29.

IX. The Appellant requested that the decision under appeal be set aside and the patent be maintained on the basis of the claims according to the main request or one of first and second auxiliary requests, these requests corresponding respectively with the first, third and fifth auxiliary requests filed under cover of the letter dated 11 August 2005.

The Respondents requested that the appeal be dismissed.

Reasons for the Decision

1. *General issues*

The question of whether the amendments made to the claims of all requests are admissible under Article 123(2) EPC or whether the claimed subject-matter is novel in view of the cited prior art (Article 54 EPC) need not be gone into since, eventually, the appeal fails for lack of inventive step.

Concerning an objection under Article 84 EPC with respect to the term "exchangeable cations", the Board agrees with the Appellant insofar as this term is understood to indicate those cations which may be exchanged by usual ion exchange processes and concludes that it is clear to those skilled in the art.

2. *Inventive Step*

2.1 The patent in suit and the claimed subject-matter relate to the removal of CO_2 from gas streams, such as air, by a temperature swing adsorption (TSA) process which is a process wherein regeneration of the adsorbent is carried out at a temperature higher than the adsorption temperature (paragraphs 1, 3, 11 and 21).

2.2 It is indicated in the description of the patent in suit to be common in the art to remove carbon dioxide from air by adsorption on a type X zeolite, such as zeolite 13X. It was agreed that this particular zeolite corresponds to NaX having a Si/Al atomic ratio in the order of 1.25.

2.3 All parties based their line of argument for evaluating inventive step on document D4, specifically on the comparative example disclosed therein, as the closest prior art.

This example discloses a process using NaX of a Si/Al atomic ratio of 1.25 as adsorbent for CO_2 from air wherein the adsorption is carried out in a TSA process at a temperature of 5°C and a CO_2 partial pressure of 3.25 mbar (350 ppm CO_2 in air at 98 psia) (see page 5, lines 5 to 52 and Claim 1).

2.4 The Board agrees that document D4 is a suitable starting point for the assessment of inventive step since it starts from the same prior art as the patent in suit, which prior art differs from the claimed process only in that the NaX used as adsorbent is one wherein the Si/Al atomic ratio is 1.25 instead of up to 1.15.

2.5 It is explained in the patent in suit that CO_2 adsorption on zeolite 13X is preferably carried out at temperatures of about 5°C or below since it becomes unfeasible at temperatures of above 20°C. Due to the tendency for adsorption bed temperatures to increase considerably during the adsorption process, it was necessary to take measures for cooling which, however, reduces the overall efficiency of the process (paragraph 4).

According to the patent in suit, the technical problem consists, therefore, in the provision of a process of enhanced economic attractiveness wherein the need for refrigeration is completely eliminated or significantly reduced (paragraph 5).

The examples of the patent in suit show that the adsorbent NaLSX having a Si/Al atomic ratio of 1.02 used in accordance with the claimed process has a considerably increased CO_2 adsorption capacity at 5°C, 35°C and 50°C and at various CO_2 partial pressures between 2 and 300 mbar when compared with the prior art zeolite NaX having a Si/Al atomic ratio of 1.25.

The examples further show that for CO_2 partial pressures up to 5 mbar the level of adsorption achieved by NaSLX at a temperature of 50°C is comparable to that achieved with the conventional adsorbent NaX at 5°C. In contrast to the latter, the claimed process is thus still commercially attractive at higher temperatures, so that it is credible that refrigeration energy can be saved under such conditions.

2.6 However, the claims of all requests cover embodiments where considerable refrigeration is required since the adsorption is carried out at temperatures down to minus 50°C. Consequently, Claim 1 of the second auxiliary request also covers embodiments requiring refrigeration despite the fact that in this case, the process is limited to the using of an adsorbent (LSX of an Si/Al atomic ratio of 1.02) for which the above effect has been shown to occur at higher temperatures.

The Board is, therefore, not satisfied that the technical problem of increased economy due to reduced refrigeration mentioned in the patent in suit is solved within the whole scope of the claims.

2.7 The Respondents accepted that the examples of the patent in suit substantiate an advantage due to the increased CO_2 adsorption capacity.

Further, it is undisputed that document D4 does not hint at an improvement of CO_2 adsorption with decreasing Si/Al atomic ratio. On the contrary, it teaches another solution to the same technical problem, namely that CO_2 adsorption can be improved if BaX having a Si/Al ratio of 1.25 is used as adsorbent instead of NaX of the same ratio (page 6, Table I and lines 20 to 26).

This means, however, that the technical problem of improved CO_2 adsorption with respect to the conventional NaX zeolite has already been solved by document D4.

2.8 Therefore, the technical problem actually solved by the process claimed in all requests in view of the disclosure of document D4 may be seen in the provision of a further process for improving the adsorption capacity when removing CO_2 from gases like air.

2.9 However, in the Appellant's opinion, the true technical problem was to achieve an improvement by using an adsorbent having a capacity for CO_2 adsorption of a magnitude that was not predictable from the prior art at the priority date of the patent in suit.

This technical problem is not accepted since it partially anticipates the solution of the problem of improving the adsorption capacity, namely by selecting a particular adsorbent (see Case Law of the Boards of Appeal, 5th edition 2006, chapter I.D.4.3.1). Further, the Board shares the Respondent's opinion that this formulation of the technical problem is not objective since it is not merely based on the differences between the closest prior art and the claimed subject-matter but includes additional state of the art concerning adsorption capacities of adsorbents other than those used in the closest prior art (see Case Law of the Boards of Appeal, 5th edition 2006, chapter I.D.4.1).

2.10 It remains to be decided whether, in view of the available prior art documents, it was obvious for someone skilled in the art to solve the above stated technical problem of providing a further process for improving the adsorption capacity (2.8 above) by the means claimed, namely by using as adsorbent NaX having a Si/Al atomic ratio of up to 1.15 in accordance with the main request, or up to 1.10 or 1.02, respectively, in accordance with the auxiliary requests (point VI above).

2.11 The Appellant argued that a skilled person would have chosen BaX as suggested in document D4 and would not have been influenced by any other prior art.

However, this argument ignores the above stated technical problem which is based on the principle that earlier solutions to a given technical problem do not preclude later attempts to solve the same problem in another way (see Case Law of the Boards of Appeal, 5th edition 2006, chapter I.D.4.5).

The fact that the technical problem of improving CO_2 adsorption with respect to the conventional NaX zeolite has already been solved does not, therefore, prevent a skilled person to look for further solutions of that same problem. Thus, a skilled person would consider further prior art and in particular prior art relating to CO_2 adsorption on zeolites, such as document D3.

2.12 Document D3 relates to a scientific study of the adsorption of CO_2 on various synthetic zeolites and the general finding that the number of cations per unit mass of the dehydrated zeolite determines the extent of adsorption of CO_2 at low saturations (see page 87, abstract). In particular, it has been found to be a general phenomenon for all zeolites that at low saturations the degree of adsorption of CO_2 increases with increase of the number of cations (page 90, left-hand column, lines 3 to 7).

It is uncontested and well-known in the art that the number of cations increases as the atomic ratio Si/Al decreases in a specific group of zeolites. Hence, document D3 teaches that the capacity for CO_2 adsorption of a given type of zeolite increases as the Si/Al atomic ratio decreases.

This phenomenon is demonstrated, inter alia, for the group of synthetic faujasites of different Si/Al ratios. As agreed by the parties, it belongs to the common general knowledge of those skilled in the art that faujasites are type X and Y zeolites which differ from each other only in that the Si/Al ratio is up to 1.5 for the former and above 1.5 for the latter (see also e.g. document D13, page 5, last paragraph).

Figure 1 on page 88 of document D3 illustrates CO_2 adsorption isotherms at 18°C on synthetic faujasites having Si/Al atomic ratios of 1.11, 1.18, 1.51 and 2.05 (table on page 88). It is apparent that within the measured pressure range from 1 to about 50 mm Hg, there exists a general trend for an increase in CO_2 adsorption with decreasing Si/Al atomic ratio.

2.13 The Board concludes, therefore, that a skilled person would generally expect that using, in the process of document D4, a NaX zeolite having a Si/Al atomic ratio lower than 1.25, would improve the CO_2 adsorption capacity over the conventional NaX zeolite.

2.14 The Appellant argued that it had been demonstrated in the examples of the patent in suit and in document A3 to A5 that a surprising increase of the CO_2 adsorption capacity was obtained if the Si/Al atomic ratio was in the range of up to 1.15, 1.10 or especially 1.02 instead of 1.25 as in the NaX zeolite used in the prior art. It was apparent that the effect was particularly pronounced at low CO_2 partial pressures up to 5 mbar.

In contrast, at such pressures no increase in CO_2 adsorption capacity could be detected in Figure 1 of document D3 if NaLSX having a Si/Al atomic ratio of 1.11 (curve 1) instead of 1.18 (curve 2) was used as the adsorbent. As no other examples of LSX zeolites were considered, document D3 did not teach any general trend for type X zeolites. In addition, a skilled person would realise from the shape that the curve for NaLSX having a Si/Al atomic ratio of 1.11 was wrong.

Those skilled in the art further knew that NaLSX having a Si/Al atomic ratio of as low as 1.11 was difficult to prepare at the time of document D3. The respective content of document D3 would, thus, have been ignored by the skilled artisan.

This was corroborated by the fact that despite the long period of more than 25 years between the publication date of document D3 and the priority date of the patent in suit, there was no evidence for an attempt in the art to use LSX for the removal of CO_2 from gases.

2.15 The Board is not convinced by these arguments if only for the reason that document D3 is quite clear when it concludes in 1968 that - as a general rule - the capacity of a given type of zeolite for CO_2 adsorption increases as the Si/Al atomic ratio decreases (point 2.12). There is no indication that this rule is invalid at low CO_2 pressures and, indeed, the Board perceives from Figure 1 in document D3 an increase in CO_2 absorption even for curve 1 (Si/Al ratio 1.11) when compared with curve 2 (Si/Al ratio 1.18) at a pressure of 5 mbar and below.

However, even if a skilled person would have had some doubts for whatever reason of the correctness of curve 1 in document D3, he would have known at the priority date of the patent in suit that the above general rule was at least noteworthy since it has been confirmed in 1978 by the teaching in document D9 that the energy of a faujasites type zeolite of binding CO_2 molecules increases with increasing charge density in the structure (page 215, last full paragraph and Table 22 on page 216) and in 1988 by the teaching of document D14 where it is found for calcium exchanged zeolite X (CaX) as adsorbent and nitrogen molecules as the adsorbate that CaLSX zeolite having the theoretical minimum Si/Al atomic ratio of 1.0 and hence the maximum possible number of exchangeable cations has considerably more sites accessible for molecules to be adsorbed than CaX of the standard Si/Al atomic ratio of 1.25 (page 470, abstract and last paragraph to page 471, line 1; page 489, Figure 7).

Apart from that, the Board observes that the effect in this pressure range is also quite clear from curves 2 to 4 alone which relate to faujasites having a Si/Al atomic ratio of 1.18, 1.51 and 2.05, respectively. It may be true that curves 3 and 4 (Si/Al ratio 1.51 and 2.05) belong to the subgroup of type Y zeolites. Nevertheless, they differ from the other subgroup of the faujasites, the type X zeolites, only in that the Si/Al ratio is higher (above 1.5; see point 2.12). There is no hint in document D3 towards a discontinuous behaviour between zeolites X and Y with respect to CO_2 absorption in relation to the Si/Al ratio.

The argument of the Appellant that NaSLX having a Si/Al ratio as low as 1.11 was not readily available at the date of document D3 is in the Board's opinion irrelevant since it was available at the priority date of the patent in suit. This is apparent from e.g. documents D13, D20 and D29, all published in 1986 or 1987 and relating to a novel method for the preparation of maximum aluminium X type zeolite having the lowest possible Si/Al ratio of 1.0 which overcomes the preparation problems in the prior art mentioned therein (document D13, abstract and page 6, lines 1 to 7; document D20, abstract, page 451, left-hand column, 'introduction' and page 452, left-hand column first full paragraph; and document D29, abstract and column 2, last paragraph to column 3, line 24).

In the Board's opinion, it is therefore not possible to conclude that the claimed subject-matter is not obvious merely from the period of time which lapsed between the publication date of document D3 and the priority date of the patent in suit without ignoring any other reasons those skilled in the art had for not applying NaLSX as adsorbent in the method of document D4.

2.16 The Board concludes therefore that, at the priority date of the patent in suit, a skilled person had ample reasons to expect that, following the teaching of document D3 (point 2.12) and applying in the method of document D4 NaLSX having a Si/Al atomic ratio of at most 1.15, 1.1 or 1.02, respectively, instead of one having a ratio of 1.25, would improve the CO_2 absorption capacity even at low CO_2 partial pressures.

Whether the magnitude of the improvement as shown in the examples of the patent in suit and in documents A3 to A5 was also to be expected or not is irrelevant since it results from the obvious performance of the method and no further measures have to be taken in order to obtain them.

2.17 For these reasons, the Board finds that the subject-matter of Claim 1 of all the Appellant's requests does not comply with the requirements of Articles 52(1) and 56 EPC.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar:	The Chairman:
M. Kiehl	P.-P. Bracke