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**Datasheet for the decision  
of 6 May 2011**

**Case Number:** T 0237/09 - 3.3.06

**Application Number:** 99936039.9

**Publication Number:** 1085935

**IPC:** B01D 53/047

**Language of the proceedings:** EN

**Title of invention:**  
Rate-enhanced gas separation

**Applicant:**  
PRAXAIR TECHNOLOGY, INC.

**Headword:**  
Gas separation with high intrinsic diffusivity  
adsorbent/PRAXAIR

**Relevant legal provisions:**  
EPC Art. 83

**Relevant legal provisions (EPC 1973):**

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**Keyword:**  
"Sufficiency of disclosure - main request, auxiliary requests  
1 - 6 (no), auxiliary request 7 (yes)"  
"Remittal to first instance"

**Decisions cited:**

-

**Catchword:**

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Case Number: T 0237/09 - 3.3.06

**D E C I S I O N**  
of the Technical Board of Appeal 3.3.06  
of 6 May 2011

**Appellant:** PRAXAIR TECHNOLOGY, INC.  
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**Representative:** Schorer, Reinhard  
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**Decision under appeal:** Decision of the Examining Division of the  
European Patent Office posted 14 August 2008  
refusing European patent application  
No. 99936039.9 pursuant to Article 97(2) EPC.

**Composition of the Board:**

**Chairman:** P.-P. Bracke  
**Members:** E. Bendl  
J. Geschwind

## Summary of Facts and Submissions

- I. The appeal is from the decision of the Examining Division to refuse the European patent application no. 99 936 039.9.
- II. The Examining Division argued in its decision, that the requirement of Article 83 EPC were not met due to the fact that a parameter for determining the mass transfer coefficient and the intrinsic diffusivity as well as the breakthrough test could not be reproduced and that also some of the adsorbents used in the process of Claim 1 could not be prepared due to lack of disclosure.
- III. The Applicant/Appellant filed an appeal against this decision, disputed the Examining Division's arguments, maintained the claims refused by the Examining Division as the main request and submitted seven sets of auxiliary requests.
- IV. The wording of Claim 1 of the **main request** is as follows:

"1. A process for the separation of nitrogen from a gas mixture including nitrogen and a less selectively adsorbable component, comprising: contacting said gas mixture in an adsorption zone with an adsorbent that is equilibrium selective for nitrogen over said less selectively adsorbable component and adsorbing said nitrogen on said adsorbent, wherein said adsorption zone comprises said adsorbent selected from the group consisting of A-zeolite, Y-zeolite, NaX, mixed cation X-zeolite, LiX, chabazite, mordenite, clinoptilolite, silica-alumina, alumina, silica, titanium silicates and

mixtures thereof, wherein said adsorbent has a mass transfer coefficient (MTC) for nitrogen of  $k_{N_2} \geq 12 \text{ s}^{-1}$  and an intrinsic diffusivity for  $N_2$ , when measured in air at 1.5 bar, 300 K and at a molar flux of  $10 \text{ mol/m}^2\text{s}$ , of  $D_p \geq 3.5 \times 10^{-6} \text{ m}^2/\text{s}$ ."

Claim 1 of the **first auxiliary request** differs from the main request in the replacement of the passage "wherein said adsorption zone comprises said adsorbent selected from the group consisting of A-zeolite, Y-zeolite, NaX, mixed cation X-zeolite, LiX, chabazite, mordenite, clinoptilolite, silica-alumina, alumina, silica, titanium silicates and mixture thereof" by "wherein said adsorbent is selected from the group consisting of type X and type A-zeolites and mixtures thereof".

The identical passage was in Claim 1 of the **second auxiliary request** amended to read "wherein said adsorbent is selected from the group consisting of monovalent cation-exchanged zeolites and mixtures thereof".

In Claim 1 of the **third auxiliary request** the cited passage was changed to "wherein said adsorbent is Li-exchanged type X zeolite".

In Claim 1 of the **fourth auxiliary request** the passage was worded "wherein said adsorption zone comprises said adsorbent selected from the group consisting of A-zeolite, Y-zeolite, NaX, mixed cation X-zeolite and LiX and mixtures thereof, wherein said zeolite is combined with a binder".

In the **fifth auxiliary request** the passage was modified to read "wherein said adsorption zone comprises said adsorbent selected from the group consisting of NaX and LiX, wherein said zeolite is combined with a binder".

In Claim 1 of the **sixth auxiliary request** the passage was amended to "wherein said adsorbent is LiX, which is combined with a binder".

Finally, in Claim 1 of the **seventh auxiliary request** the said passage was worded as follows: "wherein said adsorbent comprises beads of LiX which in the bead-forming step have been combined with a binder and subsequently were subjected to caustic digestion".

V. The main arguments of the Appellant were as follows:

Adsorbents with an intrinsic diffusivity of at least  $3.5 \times 10^{-6} \text{ m}^2/\text{s}$  have not been reported in the prior art. However, the parameter as such was known.

The application gives one concrete example how to prepare such adsorbents and teaches that inter alia binder, caustic digestion and fibre burnout may be used to prepare adsorbents according to the application-in-suit. In particular the variation of "binder content and type, the inclusion and subsequent burn-out of fibers having selected dimensions, concentrations and compositions and caustic digestion of the binder" (letter of 22 December 2008, page 11, second paragraph) may be used to modify intrinsic diffusivity. Given these tools the skilled person has to experiment to prepare adsorbents with the desired intrinsic diffusivity. This applies not only to lithium exchanged

type-X zeolites, but to all of the adsorbents mentioned in the application.

- VI. The Appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of the main request filed with letter of 29 May 2008 or one of the seven auxiliary requests filed with letter of 22 December 2008.

### **Reasons for the Decision**

#### 1. *Article 83 EPC*

- 1.1 The Appellant argued in the oral proceedings before the Board that adsorbents with an intrinsic diffusivity for  $N_2 \geq 3.5 \times 10^{-6} \text{ m}^2/\text{s}$ , when measured in air at 1.5 bar, 300 K and at a molar flux of  $10 \text{ mol}/\text{m}^2\text{s}$  have not been described in the prior art.
- 1.2 The application-in-suit contains the description of the preparation of an adsorbent called S-1, which possesses the required intrinsic diffusivity. S-1 is a lithium-exchanged type X (LiX) zeolite, which was, in the course of the production process mixed with 12% binder and subjected to caustic digestion.
- 1.3 In particular pages 31 and 32 of the application teach that the skilled person has to experiment in order to achieve an intrinsic diffusivity above  $3.5 \times 10^{-6} \text{ m}^2/\text{s}$ . No addition of binder results in an unacceptable intrinsic diffusivity (sample S-0), but a binder content of 20% followed by caustic digestion (S-2) or by fibre burnout (S-3) also does not lead to the

desired result. Only **lowering** the amount of **binder** to 12% in combination **with caustic digestion** without fibre burnout resulted in the desired intrinsic diffusivity (S-1), whereas a low amount of binder (12%) without caustic digestion leads to inferior results (S-4).

1.4 Although some experimentation is necessary to produce adsorbents when starting from the examples given in the application-in-suit, for LiX zeolite adsorbents some guidance is given by showing which materials can be used and by defining the amount of binder which may be treated by caustic digestion.

1.5 Main request

However, Claim 1 of the main request covers also adsorbents **other** than LiX zeolite. Such adsorbents with an intrinsic diffusivity of at least  $3.5 \cdot 10^{-6} \text{ m}^2/\text{s}$  have not been described so far. A method for their production is not defined in the application-in-suit, no evidence has been provided by the Appellant that such adsorbents behave exactly the same way as LiX and not even a hint is given in the application-in-suit which parameters should be changed when starting production based on known adsorbents. In addition to binder content, caustic digestion and optionally fibre burnout also the type of binder, the dimensions of the fibres, concentrations and compositions may need to be changed. Consequently the level of experimentation required to obtain adsorbents with an intrinsic diffusivity  $\geq 3.5 \cdot 10^{-6} \text{ m}^2/\text{s}$  useful for the claimed process is considered to be an undue burden to the person skilled in the art.

1.6 Thus, the requirement of Article 83 EPC is not considered to be met by the main request.

1.7 Auxiliary requests 1-5  
Identical considerations apply also to Claims 1 of auxiliary requests 1-5, because each Claim 1 of these requests encompasses at least one adsorbent other than LiX zeolite.

1.8 Auxiliary request 6  
Although the adsorbent in Claim 1 of the auxiliary request is restricted to LiX, the LiX adsorbent in auxiliary request 6 is only required to contain a binder. As is shown on page 31 of the application-in-suit, the required intrinsic diffusivity can only be achieved when subjecting the binder-containing LiX beads to caustic digestion. The application-in-suit does not teach how to prepare beads with the required intrinsic diffusivity without the caustic digestion step.

Consequently also the sixth auxiliary request is not sufficiently disclosed.

1.9 Auxiliary request 7  
Only Claim 1 of the seventh auxiliary request refers to beads of LiX containing a binder, which were also treated by caustic digestion. Since the application on pages 31 and 32 specifically discloses how a lithium exchanged type X (LiX) zeolite having the necessary intrinsic diffusivity may be prepared, the Board comes to the conclusion that the requirement of Article 83 EPC is met.



- 1.10 The Board considers therefore that the preparation of the LiX adsorbents to be used for the process of Claim 1 of the seventh auxiliary request is sufficiently disclosed.
- 1.11 In its decision to refuse the application the Examining Division furthermore argued that the interparticle void fraction and the simulation of the breakthrough test were not sufficiently disclosed.
- 1.12 Interparticle void fraction (IPVF)
- 1.12.1 According to the Examining Division the IPVF is strongly dependent upon parameters such as the packing of the bed, the bed height, particle properties, particle size distribution, particle shape and particle density and the question whether the particles should be vibrated. Since no details about these parameters are given in the application-in-suit, the skilled person would allegedly not know which conditions to select for packing the bed.
- 1.12.2 The Board cannot share this point of view. Given the lack of details about the determination of these parameters, the parameters have to be interpreted broadly, i.e. that each **suitable** interpretation can be applied: for instance vibration may be applied or not.
- 1.12.3 However, this lack of detailed information does in this case not mean that the IPVF cannot be determined. On the contrary, the determination of this parameter is rather a standard method, as is plausibly explained in the letter of appeal, item 4.5.

1.13 Simulation of the breakthrough test

1.13.1 The Examining Division argued that a skilled person can in principle determine the mass transfer coefficient (MTC), but that doubts would arise when it comes to the limits of the values used. Given the variations and uncertainty in the determination of individual parameters, the skilled person could not decide whether or not a parameters lies within the limits defined in Claim 1.

1.13.2 The Board cannot share this point of view. The Examining Division does not object to the method as such. The question whether or not test results may vary is intrinsic to all methods employing empirically determined values and rather concerns clarity than sufficiency of disclosure.

1.14 The invention as defined in the seventh auxiliary request is considered to be sufficiently disclosed.

2. Since the application was only refused on the grounds as laid down in Article 83 EPC, the case is remitted to the first instance for continuation of examination procedure, thus enabling the Applicant to defend its case before two instances.

**Order**

**For these reasons it is decided that:**

1. The decision under appeal is set aside.
  
2. The case is remitted to the first instance for further prosecution on the basis of the claims 1 to 20 of the seventh auxiliary request.

The Registrar

The Chairman

D. Magliano

P.-P. Bracke