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Datasheet for the decision of 6 May 2011

Case Number: T 0461/09 - 3.3.06

Application Number: 99908560.8

Publication Number: 1059999

IPC: B01D 53/053

Language of the proceedings: EN

Title of invention:
Pressure swing adsorption gas separation method, using adsorbents with high intrinsic diffusivity and low pressure ratios

Applicant:
PRAXAIR TECHNOLOGY, INC.

Headword:
PSA with high intrinsic diffusivity adsorbent/PRAXAIR

Relevant legal provisions:
EPC Art. 83, 123(2)

Relevant legal provisions (EPC 1973):
-

Keyword:
"Sufficiency of disclosure - main request, auxiliary requests 1 - 8 (no), auxiliary requests 9, 10 (yes)"
"Amendment beyond original disclosure - auxiliary request 9 (yes), auxiliary request 10 (no)"
"Remittal to first instance"

Decisions cited:
-

Catchword:
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Beschwerdekammern | Boards of Appeal | Chambres de recours

Case Number: T 0461/09 **-** 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: (Applicant)
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Decision under appeal:
Decision of the Examining Division of the European Patent Office posted 22 August 2008 refusing European patent application No. 99908560.8 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. 99 908 560.8.

II. The Examining Division argued in its decision that the requirement of Article 123(2) EPC would be met, but that Article 83 EPC would not be fulfilled 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 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 with regard to sufficiency of disclosure, maintained the claims refused by the Examining Division as the main request and submitted ten sets of auxiliary requests.

IV. The wording of Claim 1 of the **main request** is as follows:

"1. A pressure swing adsorption method comprising repetitions of a cycle of N steps, said method adapted to separate components of a gas mixture into at least a first component and a second component by selective adsorption of said first component into a bed of adsorbent, said method comprising the steps of:

a) during adsorption steps of a cycle, raising a pressure of a feed of said gas to said bed to an

adsorption pressure so as to enable adsorption of said first component by said adsorbent, said adsorbent exhibiting an intrinsic diffusivity for nitrogen that is equal to or greater than 3.5 x 10^{-6} m^2/s when measured in air at 1.5 bar and 300 K;

b) during desorption steps of said cycle, depressurizing said bed to a desorption pressure so as to desorb said first component from said adsorbent, a ratio of pressures of said adsorption pressure to said desorption pressure falling within a preferred range of less than 5.0."

Claim 1 of the **first auxiliary request** differs from the main request in the replacement of the passage "into a bed of adsorbent" by "into a bed of type X zeolite adsorbent".

The identical passage was in Claim 1 of the **second auxiliary request** amended to read "into a bed of monovalent cation-exchanged zeolite absorbent".

In Claim 1 of the **third auxiliary request** the cited passage was changed to "into a bed of Li-exchanged type X-zeolite adsorbent".

In Claim 1 of the **fourth auxiliary request** the passage was worded "into a bed of type X zeolite adsorbent". Additionally, at the end of the Claim the following text was attached: "wherein said bed of adsorbent has a dimension, in a direction of flow of said gas through said bed of adsorbent, of less than 1.2 m and said method performs steps a) and b) in a time of less than 40 seconds wherein preferably said dimension is less

than 0.9 m and said method performs steps a) and b) in a time of less than 30 seconds and wherein most preferred said dimension is less than 0.6 m and said method performs steps a) and b) in a time of less than 20 seconds".

Claim 1 of the **fifth auxiliary request** is identical with Claim 1 of the fourth auxiliary request, but the passage "into a bed of type X zeolite adsorbent" was replaced by "into a bed of monovalent cation-exchanged zeolite adsorbent".

Claim 1 of the **sixth auxiliary request** differs from Claim 1 of the fourth auxiliary request only in that the passage referred to before reads "into a bed of Li-exchanged type X zeolite adsorbent".

Claim 1 of the **seventh auxiliary request** is identical with Claim 1 of the main request apart from the following change: the term "into a bed of adsorbent" was replaced by "into a bed of adsorbent comprising beads of zeolite which in the bead-forming step have been combined with a binder and subsequently were subjected to caustic digestion".

Claim 1 of the **eighth auxiliary request** is identical with Claim 1 of the seventh auxiliary request, but the passage referred to before reads as follows: "into a bed of adsorbent comprising beads of type X zeolite which in the bead-forming step have been combined with a binder and subsequently were subjected to caustic digestion".

Claim 1 of the **ninth auxiliary request** is identical with Claim 1 of the seventh auxiliary request apart from the cited passage being amended to read: "into a bed of adsorbent comprising beads of LiX zeolite which in the bead-forming step have been combined with a binder and subsequently were subjected to caustic digestion, wherein the gas mixture is air, said first component is nitrogen and said second component is oxygen".

Finally, the **tenth auxiliary request** contains a total of twenty claims, the independent Claims 1 and 9 read as follows:

"1. A pressure swing adsorption method comprising repetitions of a cycle of N steps, said method adapted to separate components of a gas mixture into at least a first component and a second component by selective adsorption of said first component into a bed of adsorbent comprising beads of LiX zeolite which in the bead-forming step have been combined with a binder and subsequently were subjected to caustic digestion, wherein said gas mixture is air, said first component is nitrogen and said second component is oxygen, said method comprising the steps of:

a) during adsorption steps of a cycle, raising a pressure of a feed of said gas to said bed to an adsorption pressure so as to enable adsorption of said first component by said adsorbent, said adsorbent exhibiting an intrinsic diffusivity for nitrogen that is equal to or greater than 4.0×10^{-6} m^2/s when measured in air at 1.5 bar and 300 K;

b) during desorption steps of said cycle, depressurizing said bed to a desorption pressure so as to desorb said first component from said adsorbent, a ratio of pressures of said adsorption pressure to said desorption pressure falling within a preferred range of 1.5 to 3.5."

"9. A pressure swing adsorption system for performing a gas separation method comprising repetitions of a cycle of N steps, said separation method for separating components of a gas mixture into at least a first component and a second component by selective adsorption of said first component into a bed of adsorbent particles, wherein said gas is air, said first component is nitrogen and said second component is oxygen, said system comprising:

an adsorbent comprising beads of LiX zeolite which in the bead-forming step have been combined with a binder and subsequently were subjected to caustic digestion, said adsorbent being selective for said first component, said adsorbent exhibiting an intrinsic diffusivity for nitrogen that is equal to or greater than 3.5×10^{-6} m^2/s when measured in air at 1.5 bar and 300 K, said system exhibiting both a reduction in bed size factor
(BSF) such that the BSF is less than 226.8 kg (500 lb)/tons per day of contained oxygen and specific power consumption relative to a pressure swing adsorption system incorporating adsorbents of lower intrinsic diffusivity such that the specific power consumption is less than 7.5 kW/tons per day of contained O_2; and control means for controlling a ratio of adsorption

pressure to desorption pressure across said bed within a preferred range of less than 5.0."

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

Article 83 EPC (all requests)

Adsorbents with an intrinsic diffusivity of at least 3.5×10^{-6} m^2/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 variations in binder content, caustic digestion and fibre burnout may be used to prepare adsorbents according to the application-in-suit. 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.

Article 123(2) EPC

The combination of features of Claim 1 of the ninth auxiliary request can be derived from the application as a whole.

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 or one of the eight auxiliary requests filed with letter of 05 January 2009, or of the ninth or tenth auxiliary request filed during the oral proceedings of 06 May 2011.

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 nitrogen ≥ 3.5 x 10^{-6} m^2/s, when measured in air at 1.5 bar and 300 K, have not been described in the prior art.

1.2 The application-in-suit contains the description of the preparation of an adsorbent called Z-2, which possesses the required intrinsic diffusivity. Z-2 is a lithium-exchanged type X (LiX) zeolite, which was, in the course of the production process, mixed with a low amount of binder and subjected to caustic digestion.

1.3 Although some experimentation is necessary to produce adsorbents when starting from the examples given in the application-in-suit, for LiX adsorbents containing a binder and being subjected to caustic digestion some guidance is given by showing in the example which materials can be used and by defining the amount and kind of binder which needs to be subjected to caustic digestion.

1.4 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 x 10^{-6} m^2/s have not been described in the prior art. A method for their production is not defined in the application-in-suit, no evidence has been provided by the Appellant that such adsorbents may be prepared in exactly the same way as LiX zeolites. In the letter of 05 January

2009 it was even explicitly highlighted on page 13, item 4.9, that the adsorption rate characteristics cannot be determined until the adsorbent is formed. Thus, the person skilled in the art would have to start a series of experiments varying binder, amount of binder, caustic digestion, addition of fibre, addition of submicron latex particle and burn-out conditions to obtain adsorbents other than LiX zeolites with the desired properties.

Given the level of experimentation required to obtain non-LiX zeolite adsorbents with an intrinsic diffusivity $\geq 3.5 \times 10^{-6}$ m^2/s useful for the claimed invention, this is considered to be an undue burden to the person skilled in the art.

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

1.6 Auxiliary requests 1-2,4-5,7-8
Identical considerations apply also to Claims 1 of auxiliary requests 1-2,4-5,7-8, because each Claim 1 of these requests encompasses at least one kind of adsorbent other than the specific LiX zeolites.

1.7 Auxiliary requests 3,6
Although the adsorbent in Claim 1 of these auxiliary requests is restricted to LiX zeolite, no details about the adsorbent except the intrinsic diffusivity are given. The only example of the application-in-suit describing in detail the process of preparing adsorbents useful for the method of the present invention teaches, that the required intrinsic diffusivity is 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 third and the sixth auxiliary request are not sufficiently disclosed either.

1.8 Auxiliary requests 9,10
Only Claims 1 of the ninth and of the tenth auxiliary request refer to beads of LiX containing a binder, which beads were treated by caustic digestion.

1.9 Identical considerations apply to Claim 10 of the ninth auxiliary request and to Claim 9 of the tenth auxiliary request too.

1.10 Since the application specifically discloses how a lithium exchanged type X (LiX) zeolite according to the claimed method, having the necessary diffusivity, may be prepared, the Board considers that the application-in-suit contains sufficient information to prepare the specific LiX zeolite adsorbents used for the process/adsorption system according to Claims 1 and 10 of the auxiliary request 9 or according to Claims 1 and 9 of auxiliary request 10.

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, the lack of detailed information does in this case not mean that the IPVF cannot be determined. On the contrary, the determination of these parameters is rather a standard method, as is plausibly explained in the letter of appeal, item 4.4.

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 parameter lies within the limits defined in Claim 1.

1.13.2 The Board cannot share this point of view. As stated above, the Examining Division did not object to the method as such. The question whether or not test results show minor deviations is intrinsic to all methods employing empirically determined values and rather concerns clarity than sufficiency of disclosure.

1.14 Therefore the method according to Claims 1 or auxiliary requests 9 and 10 and the adsorption systems according to Claim 10 of auxiliary request 9 and Claim 9 of auxiliary request 10 are considered to be sufficiently disclosed.

2. Article 123(2) EPC

2.1 Ninth auxiliary request

2.1.1 Claim 1 of the ninth auxiliary request discloses among other characteristics of the method the following features:

- a specific LiX zeolite adsorbent
- the gas mixture is air, the first component is nitrogen and the second component is oxygen,
- the intrinsic diffusivity is equal to or greater than 3.5×10^{-6} m^2/s,
- the ratio of pressures of the adsorption pressure to the desorption pressure is less than 5.0.

2.1.2 The application as originally filed exemplifies the separation of air, but refers also to other gas phase separations (page 10, lines 8/9).

2.1.3 A disclosure of the combination of the features mentioned above in the application as originally filed could not be found. The only combination of air, the specific intrinsic diffusivity and the ratio of pressures can be found in Claim 5. However, this claim refers to Claim 3 defining an intrinsic diffusivity for nitrogen of equal or greater to 4.0×10^{-6} m^2/s and a pressure ratio of about 1.5 to 3.5.

2.1.4 Thus, Claim 1 of the ninth auxiliary request does not meet the requirement of Article 123(2) EPC.

2.2 Tenth auxiliary request

2.2.1 Claim 1 is based on a combination of original Claims 1,3 and 5 together with the disclosures on page 10, lines 15-17, page 15, lines 7-10 and page 18, lines 4/5.

2.2.2 Claim 9 is a combination of original Claims 11, 18 and 19 together with the disclosures on page 10, lines 15-17, page 15, lines 7-10 and page 18, lines 4/5.

2.2.3 Dependent Claims 2-10 and 12-20 derive from original Claims 2,4,6-10,12-17,20-24. Apart from the adaptation of the references, the term "pressure swing adsorption method" was replaced in Claim 17 by "system" to be in line with the wording of Claim 9.

2.2.4 Thus, the set of claims of the tenth auxiliary request meets the requirement of Article 123(2) EPC.

3. Since the application was only refused on the grounds as laid down in Articles 83 and 123(2) 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 claims 1 to 20 of the tenth auxiliary request and the description as originally filed with amended pages 2,6,7,14,15,18,22,23,30 to 35 filed with letter of 18 September 2006.

The Registrar The Chairman

D. Magliano P.-P. Bracke