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Datasheet for the decision of 17 January 2014

Case Number:	T 0789/11 - 3.3.06
Application Number:	03251245.1
Publication Number:	1340532
IPC:	B01D53/04
Language of the proceedings:	EN

Title of invention:
Process and apparatus for nitrous oxide removal

Patent Proprietor:
AIR PRODUCTS AND CHEMICALS, INC.

Opponents:
1 - Linde AG
2 - L AIR LIQUIDE SOCIETE ANONYME POUR L ETUDE ET L'EXPLOITATION DES PROCEDES GEORGES CLAUDE

Headword:
Nitrous oxide removal/Air Products

Relevant legal provisions:
EPC Art. 52(1), 54(1), 56, 114(2), 100(b), 123(2), 123(3)

Keyword:

Late filed evidence - admitted (yes) -
evidence further corroborating contested finding of the opposition division
Sufficiency of disclosure - (yes)
Novelty - (yes)
Inventive step - obvious solution (main request) - non-obvious solution (auxiliary request 1)

Decisions cited:

Catchword:

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Case Number: T 0789/11 - 3.3.06

DECISION
of Technical Board of Appeal 3.3.06
of 17 January 2014

Appellant: (Opponent 2)	L AIR LIQUIDE SOCIETE ANONYME POUR L ETUDE ET L EXPLOITATION DES PROCEDES GEORGES CLAUDE 75 Quai d'Orsay 75007 PARIS (FR)
Representative:	Beroud, Amandine L'air Liquide Direction Propriété Intellectuelle 75, quai d'Orsay F-75321 Paris Cedex 07 (FR)
Respondent: (Patent Proprietor)	AIR PRODUCTS AND CHEMICALS, INC. 7201 Hamilton Boulevard Allentown, PA 18195-1501 (US)
Representative:	Smart, Peter John Beck Greener Fulwood House 12 Fulwood Place London WC1V 6HR (GB)
Party as of right: (Opponent 1)	Linde AG Leopoldstrasse 252 80807 München (DE)
Representative:	Zahn, Christoph Linde AG Legal Services Intellectual Property Dr.-Carl-von-Linde-Strasse 6-14 82049 Pullach (DE)
Decision under appeal:	**Decision of the Opposition Division of the European Patent Office posted on 10 March 2011 rejecting the oppositions filed against European patent No. 1340532 pursuant to Article 101(2) EPC.**

Composition of the Board:

Chairman: B. Czech
Members: G. Santavicca
U. Lokys

Summary of Facts and Submissions

I. The appeal by opponent 2 lies from the decision of the opposition division rejecting the oppositions against European patent n° 1 340 532.

II. Claim 1 of the patent as granted read as follows:

"*1. A process for removing water, carbon dioxide and nitrous oxide from a feed gas stream, comprising passing the feed gas stream through a first adsorbent to adsorb water, a second adsorbent to adsorb carbon dioxide and a third adsorbent to adsorb nitrous oxide and to form a purified feed gas stream, wherein the third adsorbent has a nitrogen diffusion parameter of 0.12 sec*$^{-1}$ *or higher and a nitrous oxide capacity of 80 mmol/g/bar (79 mmol/g/atm) or higher at 30°C.*".

III. The patent in suit had been opposed on the grounds of lack of novelty and inventive step (Article 100(a) EPC) as well as insufficiency of the disclosure (Article 100(b) EPC). The prior art relied upon included:
E1: EP 1 072 300 A1;
D1: EP 0 992 274 A1;
D2: EP 1 092 465 A2;
D5: FR 2 784 599 A1;
P1: D.M. Ruthven, Principles of Adsorption and Adsorption Processes, John Wiley & Sons, Inc., 1984, pages 166 to 205.

IV. In the decision under appeal, it was *inter alia* held that:

a) The disclosure of the invention in the opposed patent met the requirement of Article 83 EPC.
b) E1 was not novelty-destroying since it did not directly and unambiguously disclose that the third

adsorbent had a diffusion parameter and a nitrous oxide capacity as defined in Claims 1, 17, 19 and 20.

c) Likewise, D2 (example 3) was not novelty-destroying, since it had not been shown that the adsorbent CECA's "Siliporite G586" mentioned therein necessarily had a nitrogen diffusion parameter in the claimed range, like the adsorbent material "*CECA Grade G586 CaX*" mentioned in a table of the patent in suit. Also, no information was directly and unambiguously derivable regarding the nitrous oxide capacity of the adsorbent.

d) Starting from either E1 or D2 as the closest prior art and taking into account the knowledge of the skilled person (reference was made to paragraph [0021] of D1) that the capacity was an essential parameter of an adsorbent, it was obvious to use an adsorbent with a high capacity, i.e. a capacitiy in the claimed range. However, since the processes of E1 and D2 already achieved very good removal of nitrous oxide, the skilled person would not be prompted by any of E1 and D2 to consider kinetic properties and to conclude therefrom that a nitrogen diffusion parameter as defined in Claim 1 should be selected. Thus, the claimed subject-matter was not obvious.

Since the oppositions were rejected, the amended set of claims according to the then pending auxiliary request 1 was not dealt with in the reasons for the decision.

V. In its statement setting out the grounds of appeal, the appellant maintained its objections regarding the insufficiency of the disclosure (concerning both the main request - patent as granted - and said auxiliary request), lack of novelty over D2 (idem) and lack of inventive step in view of D1 and D2 (main request) or D2 (auxiliary request) as the closest prior art.

VI. With its reply of 1 September 2011, the respondent (patent proprietor) re-filed the auxiliary request that had been pending before the Opposition Division and submitted two new items of evidence, namely:
P4: Data Sheet, France CECA, Molecular Sieve G5L86;
P5: Data sheet, CECA Siliporite G586 Beads 1.6-2.5 mm.

Claim 1 according to said auxiliary request 1 differs from Claim 1 as granted only in terms of the lower limit of the numerical range for the nitrogen diffusion parameter, said feature now reading

"...*third adsorbent has a nitrogen diffusion parameter of* ***0.15 sec^{-1}*** *or higher...*" (emphasis added by the Board).

The other independent Claims 16, 18 and 19 according to said auxiliary request 1 read as follows:

"*16. Apparatus for removing water, carbon dioxide and nitrous oxide from a feed gas stream comprising in fluid series connection a first adsorbent to adsorb water, a second adsorbent to remove carbon dioxide and a third adsorbent to remove nitrous oxide, wherein the third adsorbent has a nitrogen diffusion parameter of 0.15 sec^{-1} or higher and a nitrous oxide capacity of 80 mmol/g/bar (79 mmol/g/atm) or higher at 30°C and the first, second and third adsorbents may optionally be the same material.*"

"*18. A process for removing nitrous oxide from a feed gas stream, comprising passing the feed gas stream over* *an adsorbent having a nitrogen diffusion parameter of 0.15 sec^{-1} or higher and a nitrous oxide capacity of 80 mmol/g/bar (79 mmol/g/atm) or higher at 30°C.*"

"*19. The use of an adsorbent having a nitrogen diffusion parameter of 0.15 sec^{-1} or higher and a nitrous oxide capacity of 80 mmol/g/bar (79 mmol/g/atm) or higher at 30°C for removing nitrous oxide from a feed gas stream.*"

Dependent claims 2-15, and 17 are directed to more specific embodiments of the subject-matter of the independent claims on which they depend.

VII. In response to a communication issued by the Board in preparation for oral proceedings,
- the respondent submitted two further sets of amended claims as auxiliary requests 2 and 3 (letter of 31 December 2013),
and
- the party as of right (non-appealing opponent 1) announced that it would not attend the oral proceedings (see the result of telephone conversation between Mr Christoph Zahn and the Registrar, Ms L. Fernández-Gómez, on 18 December 2013).

VIII. Oral proceedings were held on 17 January 2014 in the absence of the duly summoned party as of right. The debate focussed on the issues of novelty over D2 (main request), on inventive step over D1 as the closest prior art and on sufficiency of the disclosure (main request and auxiliary request 1). At the end of the oral proceedings the decision was announced.

IX. The appellant (opponent 2) requested that the decision under appeal be set aside and that the European patent be revoked.

X. The respondent (patent proprietor) requested that the appeal be dismissed or in the alternative that the patent be maintained on the basis of the claims according to the auxiliary request 1 submitted on 1 September 2011, or on the basis of the claims according to one of the auxiliary requests 2 and 3 submitted with letter of 31 December 2013

XI. The parties' arguments regarding the main and the first auxiliary request of relevance here can be summarised as follows:

Arguments of the appellant

Sufficiency of the disclosure (both requests)

a) The process of claim 1 could not be reproduced, for the following reasons:
   i) The manufacturers of zeolite adsorbents did not know how to prepare adsorbent material on the basis of the claimed parameters, which were not sufficiently described. This was also apparent from the cited documents, which dealt with adsorption capacities and not with diffusion parameters. No document proved that the manufacturers knew what to do in order to obtain the claimed material. In particular P4 and P5 did not show the claimed parameters but other parameters.
   ii) The patent in suit stressed the importance of shape and size of adsorbent particles as well as of size of macropores, without however disclosing the manufacturers of these adsorbents or a known suitable process

therefor, let alone how to screen the product in order to select the best batches, if variations arose. So there was insufficient information on e.g. the claimed nitrogen diffusion parameter.

iii) The patent did not disclose how to carry out the volumetric uptake test, let alone how to fit the uptake rate curve to Fick's law.

iv) P1, acknowledged in the patent in suit as a reference for uptake curves, concerned spherical adsorbents (Claim 1 was open to any shape), disclosed that the concentration (q in formulae 6.1 and 6.2 of P1) of the species to be eliminated played a role (the patent in suit did not disclose at which concentration the diffusion parameter was to be measured), and that at 70% uptake its expressions deviated by 2% from the solution (implying that beyond 70% other deviations would appear).

v) Claim 1 defined a temperature of 30°C for the determination of its parameters but the tests described in the examples were carried out at 25°C. So, the operating conditions for determining the claimed parameter were unknown.

vi) Also, Example 1 of the patent in suit showed that calcium content played a role, but no items of information on the effect of calcium content on the claimed parameters were given.

vii) Summing up, even taking into account the contents of P1, the patent in suit did not provide the necessary information for determining the claimed properties.

viii) Thus, neither the product to be used nor the conditions for the determination of the defined parameters were sufficiently disclosed. As a consequence thereof, the skilled person did not know either whether he was working within the boundaries of process claim 1.

Main Request - Novelty

b) The subject-matter of Claim 1 as granted was not novel over D2, which disclosed a process for eliminating water, carbon dioxide and nitrous oxide, wherein the gas to be treated was passed through a first adsorbent for removing water, then through a second adsorbent for removing carbon dioxide and finally through a third adsorbent for removing nitrous oxide. Example 3 of D2 illustrated as the third adsorbent a CaLSX zeolite commercially available from CECA under the designation Siliporite G586, in form of beads having a size of from 1.6 to 2.5 mm. This zeolite was identical to the zeolite mentioned in the patent in suit as being suitable for the claimed process and having a nitrogen diffusion parameter D/r^2 (sec^{-1}) of 0.14. The argument of the respondent that Example 3 of D2 disclosed a low silica, calcium exchanged zeolite X (CaLSX), which was not Siliporite G586, as shown by P4 and P5, was not credible. In fact, D2 disclosed zeolites X having a Si/Al elemental ratio ranging from 0.9 to 1.3 and containing up 100% calcium ions, which encompassed low silica zeolites with a Si/Al ratio of 0.9 to 1.15 and containing 60 to 100% calcium ions. Hence, the mention in Example 3 of D2 of the label "CaLSX" did not contradict the mention

"Siliporite G586", as both were CaX zeolites. Also, the data mentioned in example 3 of D2 and shown in P5 (commercial designation, shape and size of particles) were identical. Furthermore, neither D2 nor P5 mentioned any Si/Al ratio, which was a further indication that both dealt with a CaX zeolite with Si/Al ratio of 0.9 to 1.3, thus encompassing the CaLSX zeolites. Hence, there were no elements of disclosure casting doubts on the identity of the materials mentioned in the patent in suit and in D2, respectively. The disclosure in the patent in suit, acknowledged in the decision under appeal, that not all commercial zeolites Siliporite G586 in form of beads of 1.6 to 2.5 mm as disclosed in D2 had a nitrogen diffusion parameter of at least 0.12 s^{-1}, was not contested as such. In fact, it was known, both from the patent in suit (paragraph [0027] was referred to) and e.g. D5, that calcium exchange influenced the claimed diffusion parameter. Since the identity of the material designation in P5 and D2 implied a disclosure of the claimed parameter, the process of Claim 1 as granted lacked novelty over D2.

Main request - Inventive step

c) D1 was the closest prior art document, as it disclosed a process as claimed. This was *inter alia* apparent from the mention in paragraph [0042] of D1 that the breakthrough of nitrous oxide and carbon dioxide happened virtually at the same time using the binderless CaX zeolite described.

d) The technical problem addressed in the patent in suit was to prevent breakthrough of nitrous oxide prior to carbon dioxide. D1 had already been acknowledged in the application as filed. The process of D1 as carried out with the three adsorbents illustrated in its example attained a percentage of removal of nitrous oxide as high as 93%, which was comparable with the removal achieved with the CaX 4 UOP Grade VSA 2 material referred to in the patent in suit and having a nitrogen diffusion parameter of 0.11 s^{-1}. It should also be considered that the process illustrated in the Example of D1 used a stream containing 370 ppm carbon dioxide, which was comparable with the 400 ppm content illustrated in the patent in suit, whereas the nitrous oxide content of only 290 ppb was much less than the 5 ppm content illustrated in the patent in suit. Thus, in the process illustrated by D1, the high amount of carbon dioxide contained in the gas would tend to occupy more adsorption sites. Hence, it was more difficult to remove nitrous oxide than in the process illustrated in the patent in suit, in which the gas stream contained much more nitrous oxide, so that nitrous oxide would breakthrough earlier. As regards the allegedly lower temperature used in the process illustrated in D1, also the examples of the patent in suit had been carried out at a temperature (25°C) which was lower than that defined in Claim 1 (30°C). Also, the physical properties of the adsorbent which according to the patent in suit were important had not been defined in Claim 1, nor was it made apparent how to select the adsorbent material, so that Claim 1 was broad also in this respect. Nevertheless, even if there were no ambiguity in the temperatures, the claimed process used a third adsorbent having a nitrogen diffusion parameter being only 0.01 s^{-1} better than that used in the process of D1. The effect of this difference, as apparent from Figure 1 of the patent in suit, was an increase of only about 1.2% in the removal of nitrous oxide, wich was not significant. So the problem solved by the claimed process was the mere provision of a further process, e.g. with an alternative third adsorbent.

e) The claimed process, having regard to the small increase in the removal of nitrous oxide, was obvious over the process of D1, as it was the result of an optimisation, which was an usual activity in the technical field. This was also apparent from the patent in suit (Paragraph [0024] was referred to), which mentioned that the skilled person was able to select the optimal shapes and sizes of the zeolite materials for the intended separation. On the other hand, although in the patent in suit attention was drawn to shape and size of the particles and calcium content, which increased both the adsorption capacity and the transfer zone, none of these were defined in Claim 1, so that any particle size and shape was encompassed by Claim 1.

f) If the claimed solution were not considered to be an obvious result of the optimisation of the process of D1, it should be considered to be the obvious result of the combination of the disclosures of D1 and D2. The process disclosed by D2 was similar to the claimed one and to the process of D1. D2 proposed an effective solution, as apparent from its Example 3, which mentioned that no traces of impurities including nitrous oxide would enter the cryogenic plant. The solution proposed by D2 was based on Siliporite

G586, which was also suitable for the process as claimed in the patent in suit. The variance of the nitrogen diffusion parameter from 0.10 to 0.14 was not a problem for the skilled person, as it was usual to test the batches of material coming from the suppliers to take the best ones, which in the present case were those also having the highest nitrogen diffusion parameters.

g) Thus, the claimed process was obvious over the cited prior art.

Auxiliary request 1 - Inventive step

h) The increased minimum nitrogen diffusion parameter value of 0.15 s^{-1} required by Claim 1 of auxiliary request 1 provided no significant further effect to the claimed process. Adsorbents with nitrogen diffusion parameters up to 0.14 s^{-1} were already known according to the patent in suit, and it was apparent from Figure 1 of the patent in suit (comparing the removal of nitrous oxide obtainable with diffusion parameters of respectively 0.14 and 0.15 $s^{-1)}$ that the increase in nitrous oxide removal was very small, and thus not surprising. Since the skilled person knew that the kinetics were important in adsorption separation, also for zeolite with high calcium content which provided the best results, he would look for adsorbents with high calcium content and good diffusion parameters. Hence, also the process of Claim 1 of auxiliary request 1 was merely the result of an obvious optimisation of known separation processes as described in D1.

i) In any case, if the claimed solution were not considered as being obvious over D1 alone, zeolite materials such as CaX achieving a nitrous oxide removal of 98% were already known, e.g. from E1 (Example 3 was referred to) or from D5 (Examples 4 and 6 were referred to), and could be obviously used by the skilled person.

j) D5, in particular, showed that the skilled person knew that the removal of nitrous oxide was unproblematic. This removal could go for 60 minutes (Example 4, Figure 3, were referred to) and up to 100 minutes (Example 6, Figure 5 were referred to), depending on the content of calcium of the zeolite (86% in Example 6 of D5). Thus, D5 already disclosed a zeolite material exchanged with 86% of calcium, which was equivalent to the material CECA G586 mentioned in D2 and in the patent in suit. Hence, it was not apparent that an improvement had actually been achieved over D5. The skilled person would obviously have used zeolite materials with high calcium content as alternative materials for the process of D1. As regards the kinetics, the claimed parameter was merely one of the ways of indicating it, whereby the skilled person routinely checked the zeolite materials for taking the best ones.

Arguments of the respondent

New items of evidence

k) P4 showed that the material referred to in D2 as CaLSX was a molecular sieve form CECA designated as G5L86, whereas P5 showed that the material mentioned in the patent in suit was not a LSX zeolite, as also apparent from the fact that the parameters of the material of P5 did not match with those of the LSX material of P4. These documents were *prima facie* relevant for establishing novelty, and were filed to further corroborate the finding in the decision under appeal that there was a lack of certainty that the CaLSX material mentioned in D2 and the CaX CECA grade G586 mentioned in Table 3 of the patent in suit were the same material. Hence P4 and P5 should be admitted and considered.

Sufficiency of the disclosure

l) The objections concerning sufficiency of disclosure were too vague and thus not conclusive for the following reasons:
   i) The nitrogen diffusion parameter defined in Claim 1 and the expression $D/r^2(sec^{-1})$ mentioned in the table on Page 4 of the patent in suit referred to one and the same nitrogen diffusion parameter. The value mentioned in Claim 1 thus had to be interpreted as described in the patent.
   ii) The diffusion parameter was important to boost the adsorption kinetic, not to maximize it, and was used as specific parameter not directly related to the adsorption capacity.
   iii) In any case, the parameter values required by Claim 1 could be obtained from the manufacturers of zeolite adsorbent, as they were properties ought to be demanded. So the manufacturers could produce and check themselves whether the claimed parameters were fulfilled. This assertion was in

dispute but the appellant never proved that it was false, although the appellant could have simply asked the manufacturers about this.

iv) The volumetric uptake tests and the fitting of the uptake rate curve (fractional approach to equilibrium versus time) to Fick's Law were disclosed in P1, which was acknowledged in the patent in suit. Hence, the skilled person knew how to produce volumetric uptake curves.

v) Nitrogen had been chosen for technical reasons, namely because nitrogen did not generate much heat of adsorption as well as because it was known e.g. from E1 that CaX molecular sieve materials which strongly adsorbed nitrogen also adsorbed nitrous oxide. The determination of the nitrogen diffusion parameter had of course been carried out on a bed of particles, not on a single spherical particle as alleged.

vi) The results did not depend on picking any particular temperature or pressure. As a case in point, a deviation of only 2% was obtained if the temperature were raised from 25°C, as mentioned in the description, to 30°C, as claimed.

Summing up, for a skilled person there was no difficulty in carrying out the claimed process.

Main request - Novelty

m) The zeolite material mentioned in D2 was a low Si/Al CaX zeolite material and D2 was silent on its nitrogen diffusion parameter. The patent proprietor had evidenced (as stated in the patent in suit) that commercially available Siliporite G586 varied from batch to batch and that not all batches met the nitrogen diffusion parameter as defined in Claim 1. Even leaving aside batch issues, there was no certainty that the material illustrated in D2 was identical to the CaX CECA grade G586 illustrated in the patent in suit, as evidenced by P4 and P5, e.g. by the fact that zeolite G5L86 of P4 was different from the zeolite mentioned in the patent in suit. The argument that the calcium content influenced the nitrogen diffusion parameter was a new argument, which however was one more reason for not having lack of novelty over D2. Finally, zeolites, either in pellets or beads form, were complex products with macropores, micropores and skin resistances, so that batch selection was necessary. Therefore, the claimed subject-matter was novel over D2.

Main request - Inventive step

n) D1 could be considered as the closest prior art.

o) The materials used in the claimed process were *inter alia* characterized by their diffusion parameter, which reflected the process's kinetic.

p) The invention was a significant advance in air pre-purification. The examples of the patent in suit showed that by increasing also the kinetic parameter defined in Claim 1 an improvement was attained, which did not amount to only 1.2%, as argued by the appellant, but to a gain of 1.2% of the remaining possible 6.5% increase (i.e. the available range of improvement from 93.5 of D1 to 100%). This was a substantial proportion, which continuously increased as the nitrogen diffusion parameter itself increased, as apparent from the 97.4% recovery obtained with a nitrogen diffusion parameter of 0.25.

q) Since the open range for the diffusion parameter meant the higher the better, it was actually also attained over the whole breadth of the claim. In this respect, the patent in suit also listed materials other than CaX zeolites, other zeolites, with which the same improvement would also be obtained. This had not been contested by the appellants.

r) The significance of the improvement was also apparent from a comparison of the accepted carbon dioxide breakthrough concentration in the patent in suit (1 ppm) and in D1 (20 ppb), even though the two examples were not strictly comparable. These results implied that if the breakthrough concentration of 1 ppm illustrated in the patent in suit were lowered, less nitrous oxide would escape, i.e there would be a further improvement. Instead, if the Example of D1 were carried out for longer time, the percentage of nitrous oxide removal would be less than 93%. Also, D1 (and E1 too) used a much lower temperature (14 and 12.5°C) than that illustrated in the patent in suit (25°C), which thus favoured adsorption of nitrous oxide. The argument brought-up by the appellant for the first time during the oral proceedings, according to which the adsorption of nitrous oxide increased with higher contents of nitrous oxide in the gas, was contested. As a matter of fact, the better results illustrated in the patent in suit proved that the material of D1 had a worse kinetic.

s) The results shown in the table of paragraph [0032] of the patent in suit showed that:
   i) Optimisation of the process in terms of nitrous oxide capacity only was not the problem solved, as the material CaX3, having the best nitrous oxide capacity, nevertheless attained a nitrous oxide removal of only 88.8%.
   ii) The zeolite CaX2 586 (containing 86% calcium) had the right kinetic parameter and attained a better removal, as also apparent from Figure 1, which correlated the diffusion parameter and the removal.
   iii) The zeolite material CaX4 UOP Grade VSA2 did not have the right kinetic and did not achieve a satisfactory removal.
   iv) Summing up, all these data showed that there was a successful correlation between the nitrogen diffusion parameter and the efficiency of the nitrous oxide removal.

t) Finally, the statement in D2 that no traces passed to the cryiogenic plant concerned a small production, and not a high production plant, the results of which were unknown. Hence, the claimed process was an improvement also over that of D2.

u) The patent in suit disclosed something that had not previously been recognized in the prior art, namely that in order to remove more nitrous oxide the kinetic should be improved. Nobody had wanted to optimise the kinetic, especially in a context where larger particles produced less pressure drop through the bed. Also, there were elements of contradiction in the argument of the appellant, according to which an allegedly unusual parameter would have usually been optimised by the skilled person.

v) If the invoked improvement over D1 were not acknowledged, it had not been shown why it would have been obvious to carry out the required modification of the process of D1, let alone where in D2 the skilled person would find those features of Claim 1 at issue which were not disclosed in D1.

Auxiliary request 1 - Inventive step

w) No arguments based on D1, let alone in combination with E1 or D5, had ever been brought up before. Nevertheless, D1 could still be considered as the closest prior art.

x) The arguments provided for the process of Claim 1 of the main request applied *a fortiori* to the process of Claim 1 of auxiliary request 1, which was more distant from the known processes (D1 and D2). Thus, this process too was not obvious over D1, which disclosed breakthrough of nitrous oxide and carbon dioxide at "virtually the same time", but which permitted a removal of only 93% of the nitrous oxide.

y) Neither D5 nor E1 hinted at addressing the kinetic aspects of the process, so that combining them with D1 such as to arrive at the claimed subject-matter was not obvious.

Reasons for the Decision

1. The appeal is admissible.

Procedural aspects - Admissibility of new items of evidence

2. With its response to the statement setting out the grounds of appeal, the respondent submitted further items of evidence P4 and P5 (*supra*).

2.1 P3 and P4 were submitted in reaction to the statement setting out the grounds of appeal, in order to further corroborate that the adsorbent materials mentioned, respectively, in D2 and in the patent in suit were different, hence that the claimed process was indeed novel over D2.

2.2 Therefore, the board decided to admit documents P4 and P5 into the proceedings despite their late filing (Articles 114(2) EPC and 12(4) RPBA).

Main request (Claims as granted)

Sufficiency of the disclosure

3. The appellant's objections under Article 100(b) EPC maintained in the appeal proceedings can be summarised as follows:

   (a) As concerns the capacity, the skilled person did not know the conditions under which it should be calculated.

   (b) Manufacturers did not know how to make adsorbent material having the required properties.

   (c) The patent in suit mentioned but did not disclose:

      (i) the shape and size of the adsorbent particles;

    - (ii) the effect of the calcium content;
    - (iii) a process for obtaining suitable adsorbent particles.

(d) Claim 1 contained two essential features, i.e. the nitrogen diffusion parameter of at least 0.12 s^{-1} and the nitrous oxide capacity of at least 80 mmol/g/bar which could not be determined without ambiguity because:

    - (i) The patent in suit did not disclose any clear and standard way permitting to determine or calculate these two parameters.
    - (ii) The document acknowledged in the patent in suit (P1) contained several formulae and conditions for calculating the diffusion parameter, but the skilled person did not know which of these formulae and conditions had to be taken. As a case in point, the examples of the patent in suit mentioned a temperature of 25°C, whereas Claim 1 referred to a temperature of 30°C.

3.2 However, as regards the information contained in the patent in suit, a distinction must be drawn between the two parameters defined in Claim 1 as regards their usualness and method of determination, as follows:

(a) The capacity specifically refers to nitrous oxide (as the gas to be removed), is a usual parameter (see e.g. D1, Table 1) and the conditions for its determination are, for the Board, sufficiently dealt with in the patent in suit (paragraphs [0029] to [0032]). This was indicated in the Board's communication in preparation for oral proceedings.

(b) The nitrogen diffusion parameter, however, does not concern a gas to be removed, does not appear to be a usual parameter described or defined in the available prior art (no cited document mentions it) and its determination does not appear to described in full detail in the patent in suit.

3.3 As concerns the capacity parameter, neither in response to the Board's communication, let alone during the oral proceedings, did the appellant reiterate any of the objections previously raised. So the Board sees no reason for deviating from its preliminary opinion that the claiming of this capacity parameter value range is not objectionable under Article 100(b) EPC, or for going into more detail in this respect.

3.4 As regards the nitrogen diffusion parameter, the following elements of information are contained in the patent in suit:

(a) The production of adsorbents with the defined diffusion coefficient lies within the skill of adsorbent manufacturers (see paragraph [0024]).

(b) If necessary, suitable batches of adsorbent material having the required nitrogen diffusion parameter value can be selected from available batches of produced adsorbent (see paragraph [0024]).

(c) The nitrogen uptake rate of each sample was determined (see paragraph [0030]) by performing a volumetric uptake test and fitting the uptake rate curve (fractional approach to equilibrium vs. time) to Fick's law, as described in P1 Chapter 6.

(d) The table of paragraph [0032] mentions commercially available adsorbents (first two adsorbents) possessing the claimed diffusion parameter.

3.5 It is in dispute whether the information "*nitrogen diffusion parameter of 0.12* s^{-1} *or higher*" is sufficient to permit purchasing a suitable commercial adsorbent.

Also, no document showing the process of production of adsorbents with the claimed diffusion parameter is acknowledged or described in the patent in suit.

Nevertheless, this is not decisive, as, in this respect, the appellant did not disprove the assertions in paragraph [0024] of the patent in suit, but merely cast doubts thereon, although it could have sought to provide a statement by any such manufacturer. Therefore, the appellant did not discharge its onus of proof in this respect, i.e. it did not show that the attacked relevant statement in the patent in suit was false.

The same conclusion applies to the possibility of selecting the batches, also mentioned in the patent in suit, also not disproved by the appellant. Instead, during the discussion of inventive step, the appellant brought forward that the adsorbent materials were always screened for their properties before use, so as to assess their suitability for the intended task (*infra*).

3.6 As concerns the question whether the nitrogen diffusion parameter is a known or usual parameter in the field of gas adsorption, and whether it can be determined or calculated, for a given adsorbent, on the basis of the information provided in the patent in suit or P1 mentioned therein, the following is taken into account:

3.6.1 As apparent from e.g. P1 (equations 6.4 and 6.6 to 6.9. for "$D_c/r^2{}_c$", where c stands for zeolite crystal; equations 6.15 to 6.17 for "$D_p/R^2{}_p$", where p stands for pellet; equations 6.35 and 6.36, where both are referred to in the definition of "α" (alpha); equation

6.48, where "α" is defined in terms of *inter alia* "D_p/l^2", where "l" is the bed depth) and from P3 (see e.g. equation 5.16), diffusional parameters are known parameters for modeling mass transfer, namely diffusional time constants which depend on particle shape and size and macropore and micropore diameters. On the other hand, it is apparent, e.g. from the prior art patent documents mentioned in the present case, that diffusion parameters are not normally used in the descriptions or definitions of the relevant inventions.

Summing up, for the Board, P1 confirms that the diffusion parameter (for a given gas) as defined in Claim 1 is a known concept, which however does not appear to be a usual parameter for defining an adsorbent in e.g. patent applications.

3.6.2 The patent in suit expressly indicates (paragraph [0030]) that the nitrogen uptake rate was determined by performing a "*volumetric uptake test*". As mentioned in its communication issued in preparation for oral proceedings, the Board understands this test to be a known technique, wherein the response of the system is the measure of the change of the amount of gas phase molecules in the sorption vessel and by "fitting the uptake rate curve (fractional approach to equilibrium versus time) to Fick's law", as disclosed in P1, Chapter 6). This was not contradicted in writing or during the oral proceedings,

3.6.3 According to P1 (page 166, first paragraph, penultimate sentence), uptake methods are known "*simple method of studying adsorption kinetics*". In particular, equations such as equation 6.4 are "*familiar*" (which evidently also applies to D_c/r^2_c). Furthermore, P1 (pages 166 to 167, section 6.1) discloses that particle size and heat of sorption play a role, i.e should be taken into account. Finally, still according to P1 (paragraph 6.5, first sentence), "in the measurement of micropore diffusivities by the uptake rate method it is generally necessary to use an assemblage of microparticles (such as zeolite crystals) rather than a single isolated particle", hence to use a bed of zeolite particles.

3.7 Therefore, on the one hand, the Board has no reason to object to the choices made by the respondent (volumetric uptake rate, nitrogen, reduction of heat generation to approach isothermal conditions, bed of particles instead of single particle) when applying said measuring method to obtain a parameter value range defining its invention, as they appear to be sensible choices, fully justified in light of P1, and appropriate in the context of the technical field of the present invention.

3.8 Moreover, on the other hand, in opposition proceedings the burden to show that the claimed subject-matter is not disclosed in a manner sufficiently clear and complete for it to be carried out by a person skilled in the art normally lies on the opponent (Article 100(b) EPC).

3.8.1 However, considering the lack of evidence supporting the objections raised, it is not apparent where in particular the difficulty invoked by the appellant actually lies.

3.8.2 Also, the appellants did not indicate why the findings in the decision under appeal in relation to sufficiency of the disclosure are not correct.

3.8.3 Thus, the appellants' objections were not sufficiently substantiated to discharge the onus of proof of an opponent alleging insufficiency.

3.9 Summing up, the doubts cast by the appellant are not substantiated by verifiable facts, so that, in the board's judgement, the patent as granted is not objectionable under Article 100(b) EPC.

Novelty - Claim 1

4. Lack of novelty was only invoked having regard to the disclosure of D2, in particular of its Example 3.

4.1 It is not disputed that D2 does not expressly disclose the nitrogen diffusion parameter and the nitrous oxide capacity of the adsorbents described therein.

4.2 The appellant's objection is based on the assumption that the adsorbent used according to example 3 of D2, for removing *inter alia* N_2O from a feed gas stream, i.e. a "**CaLSX** type zeolite adsorbent, available as **Siliporite G586**, bead size **1.6 to 2.5** mm, from CECA France" (emphasis added), inherently has a nitrogen diffusion parameter and a nitrous oxide capacity within the respective ranges according to Claim 1 at issue.

4.3 The appellant argued that this assumption was justified in view of the data contained in the patent in suit (table of paragraph [0032]) where an adsorbent designated as "**Siliporite G586** from Ceca in **1.6 x 2.5** mm beads (EP-1092465)" is reported to have a nitrogen diffusion coefficient of 0.14 s^{-1} and a nitrous oxide capacity of 254 mmol/g/bar.

5. However, the respondent contested, also relying on documents P4 and P5, that said adsorbents mentioned in D2 and in the patent in suit are necessarily the same.

5.1 In this respect, the following must also be taken into account:

(a) P4, on the one hand, refers to a "G5**L**86 type" "Ca**L**SX zeolite" from Ceca, in form of 1.2/2.0 mm beads, i.e. a **l**ow silicon CaX zeolite.

(b) P5, on the other hand, describes a "Siliporite G586" from Ceca, in form of 1.6-2.5 mm beads, and discloses that this material is used in air pre-purification upstream air separation units. P5 does **not** mention that Siliporite G 586 is a Ca**L**SX zeolite, i.e. a low silicon CaX zeolite.

(c) None of P4 and P5 discloses the calcium content of the described zeolites, which thus is not necessarily the same in both materials.

(d) Hence, if only for these reasons, there is no evidence convincingly showing that the low silicon ("CaLSX") zeolite "Siliporite G586" mentioned in Example 3 of D2 and the "Siliporite G586" zeolite mentioned in the patent in suit are zeolite materials which are necessarily identical, let alone in terms of the properties of relevance here.

5.2 Also, the statement in the patent in suit (last sentence of paragraph [0034]) that the Siliporite G586 from Ceca in 1.6-2.5 mm beads form had a nitrogen diffusion parameter which **varied in the range of from 0.10 to 0.14 s^{-1}**, is not disproven by evidence. Hence, the mentioning of a "S*iliporite G586*" does not necessarily imply a nitrogen diffusion parameter value within the claimed range.

6. Hence, in the Board's judgement, D2 does not, expressly or inherently, directly and unambiguously disclose the use of an adsorbent with all of the properties of the third adsorbent as defined in claim 1 at issue. The claimed subject-matter is thus novel (Articles 52(1) and 54(1)(2) EPC).

Inventive step - Claim 1

7. The invention

7.1 The invention concerns *inter alia* (see claim 1 and paragraph [0001] of the patent in suit) a process for removing nitrous oxide from a feed gas stream, more particularly in the pre-purification of air, making use of a suitable nitrous oxide adsorbent.

7.2 Nitrous oxide is a minor component of air, which however presents potential operation problems resulting from solids formation in the separation apparatus column and heat exchangers (paragraph [0002] of the patent in suit). Nitrous oxide also presents a safety hazard since it enhances the combustion of organic materials) and since it is shock sensitive (paragraph [0003] of the patent in suit).

8. Closest prior art

8.1 For the Board, D1 constitutes the closest prior art, considering that it discloses a process of the type defined in Claim 1 and that it addresses issues related to early breakthrough of nitrous oxide in pre-purification of air (D1: paragraph [0042]). Also, the process of D1 has more features in common with that of Claim 1 than the process of D2, initially invoked by the appellant as the closest prior art document and dealt with in the decision under appeal. This was also common ground between the parties at the oral proceedings.

8.2 More particularly, D1 (Claim 1) discloses a process for removing water, carbon dioxide and nitrous oxide from a feed air stream prior to cryogenic distillation of the air stream to separate a nitrogen rich stream and/or an oxygen rich stream, comprising passing said feed air stream containing water, carbon dioxide and nitrous oxide through a first adsorbent to adsorb said water, through a second adsorbent which optionally may be the same as the first adsorbent to remove carbon dioxide and through a third adsorbent to remove said nitrous oxide from said air stream.

8.2.1 The third adsorbent can preferably be a calcium exchanged X zeolite (Claim 8), most preferably binderless Ca exchanged X zeolite (paragraph [0022]).

8.2.2 In the example of D1 (paragraph [0044]), the three-layer bed concept was tested in a pilot unit (about 15 cm in diameter by 122 cm long). The bed was filled with 30 cm of potassium carbonate impregnated alumina, followed by 60 cm of 13X zeolite and a final layer of 30 cm of binderless CaX. Feed air at 61 kPa feed pressure, 14°C, with 370 ppm CO_2, 1 ppm acetylene, 1 ppm ethylene and 290 ppb N_2O was passed through the bed. The CO_2 breakthrough concentration was 20 ppb. The three-layer bed gave (table 2) 100% acetylene removal, 100% ethylene removal and 93% nitrous oxide removal.

8.2.3 The examples shows that the process of D1 with the said three-layer bed brings about a higher nitrous oxide removal compared to earlier prior art making use of a two-layer bed without CaX as third adsorbent (see D1, paragraph [0045] and the results in Table 2).

8.2.4 D1 discloses as critical the Henry's Law constants for carbon dioxide and nitrous oxide, i.e. the capacity of the adsorbent with respect to these gases. According to D1, the Henry's Law constant for nitrous oxide adsorption of the third adsorbent is preferably at least 79 mmole/g/atm (Claim 7), more preferably at least 500 mmole/g/atm, and still more preferably at least 1000 mmole/g/atm (paragraph [0021]).

9. The technical problem according to the respondent

At the oral proceedings, the respondent, referring to the contents of the patent in suit (paragraphs [0001], [0012], [0027] and [0028]), submitted that the problem to be solved also in the light of the closest prior art D1 was to provide an improved process for the removal of water, carbon dioxide and nitrous oxide from a feed gas stream.

10. The solution

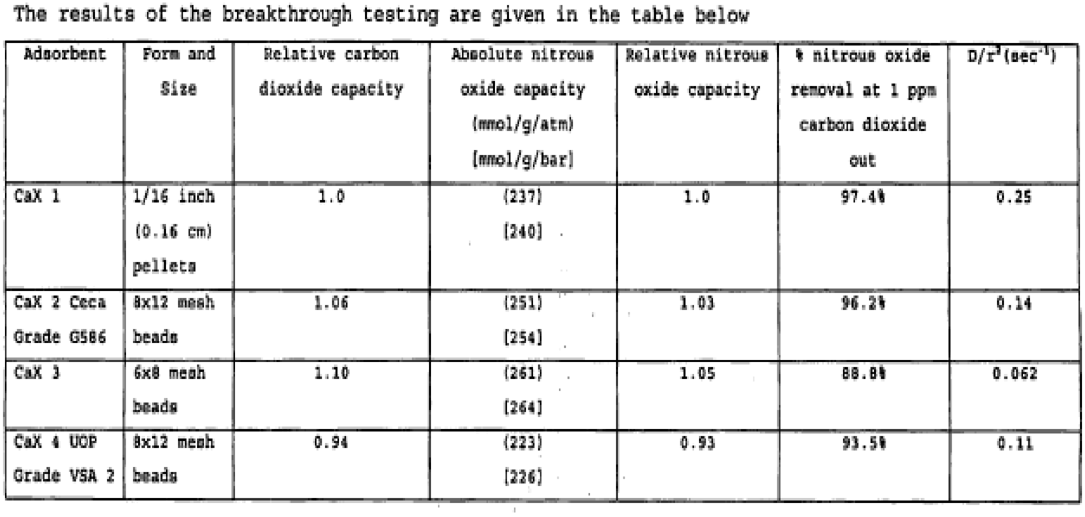
As a solution to this technical problem, the patent in suit proposes a process for removing water, carbon dioxide and nitrous oxide from a feed gas stream according to Claim 1 as granted, which is characterised in particular in that it "the third adsorbent has **both a "*nitrogen diffusion parameter of 0.12* s^{-1} *or higher*" and a "*nitrous oxide capacity of 80 mmol/g/bar or higher at 30°C*".**

11. The alleged success of the claimed solution

11.1 The respondent held that the adsorbent as defined in claim 1, due to its high capacity and a short (narrow) mass transfer zone for nitrous oxide, prevented early breakthrough of the gases.

11.2 According to the patent in suit (paragraph [0034]), the Siliporite G586 of D2 in form of beads of 1.6x2.5 mm has a nitrogen diffusion parameter which may range from 0.1 to 0.14 s^{-1}, i.e. which might or might not fulfil the required nitrogen diffusion parameter of Claim 1.

11.3 It can be gathered from Example 2 of the patent in suit, in particular from the table of paragraph [0032], reproduced herebelow,



The results of the breakthrough testing are given in the table below

Adsorbent	Form and Size	Relative carbon dioxide capacity	Absolute nitrous oxide capacity (mmol/g/atm) [mmol/g/bar]	Relative nitrous oxide capacity	% nitrous oxide removal at 1 ppm carbon dioxide out	D/r^2 (sec^{-1})
CaX 1	1/16 inch (0.16 cm) pellets	1.0	(237) [240]	1.0	97.4%	0.25
CaX 2 Ceca Grade G586	8x12 mesh beads	1.06	(251) [254]	1.03	96.2%	0.14
CaX 3	6x8 mesh beads	1.10	(261) [264]	1.05	88.8%	0.062
CaX 4 UOP Grade VSA 2	8x12 mesh beads	0.94	(223) [226]	0.93	93.5%	0.11

that the choice of CaX zeolites having a nitrogen diffusion parameter of respectively 0.11, 0.14 and 0.25 permits to attain a percentage of nitrous oxide removal, at 1 ppm carbon dioxide out, of respectively 88.8%, 96%2 and 97.4%. Hence, these results appear to show that the higher the nitrogen diffusion parameter, the better the removal of nitrous oxide.

11.4 Still from the said table, it can be gathered that:
  (a) The said trend is not, or not only, dependent on the absolute nitrous oxide capacity, which does not necessarily increase as does the nitrogen diffusion parameter.
  (b) For the CaX zeolite materials in form of 8x12 mesh beads (2.38x1.68 mm), it appears that an increased nitrogen diffusion parameter leads to an increased nitrous oxide removal (93.5% versus 96.2%). According to the patent in suit (paragraph [0035], last sentence), this is because zeolite CaX 4 has a smaller macropore diameter (hence, higher resistance to mass transfer) than CaX 2.
  (c) A CaX in form of pellets and having a smaller particle size (1/16 inch = 1.6 mm) gives the best result (97% nitrous oxide removal).
  (d) Hence, shape and size of particles as well as size of macropores influence the nitrogen diffusion parameter, thus the overall rate of adsorption.

11.5 It follows from the foregoing that the data in the patent in suit invoked by the respondent show that the kinetic aspects of the zeolite material, such as the nitrogen diffusion parameter, are not arbitrary and permit to attain better removal.

11.6 However, proof that a solution is not made up of arbitrary features does not necessarily prove that an improvement is effectively achieved across the whole breadth of claim 1 over the closest prior art, i.e. the process according to D1.

11.7 In fact, D1 is acknowledged in the patent in suit which does not, however, contain any statement that the claimed process is improved over that according to D1. From paragraph [0035] of the patent in suit, it can only be gathered that the adsorbents disclosed in D1 are defined by material and size only, i.e. are not (sufficiently) well defined to determine their nitrogen diffusion parameter. Hence, no comparison over D1 in terms of nitrogen diffusion parameter is made in the patent in suit, so that the question arises whether an improvement over D1 can be acknowledged on the basis of other facts.

11.8 During the oral proceedings, the respondent argued that since the CaX 4 UOP Grade VSA 2 in form of beads of 8x12 mesh mentioned in the said table of the patent in suit, which, with a nitrogen diffusion parameter of 0.11 s^{-1}, attains a nitrous dioxide removal of 93.5%, corresponds to that disclosed in D1, an improvement over the process of D1 in terms of percentage of nitrous oxide removal had been demonstrated.

11.9 However, the CaX 4 UOP Grade VSA 2 in form of beads of 8x12 mesh mentioned in the said table of the patent in suit has an absolute nitrous oxide capacity of only 226 mmole/g/bar, which is about one third of that illustrated in D1 for the binderless CaX, and about half that still illustrated in D1 for the CaX material. Hence, the CaX 4 UOP Grade VSA 2 material is not directly comparable with the zeolite materials illustrated in D1.

11.10 Also, the operating conditions (such as the size of the pilot unit, the pressure and temperature, the flow rates, the (relative) content of e.g. carbon dioxide and nitrous oxide in the feed gas stream) described in Example 2 of the patent in suit and in the example of D1 are very different, thus not comparable. In this respect, it must be taken into account that Claim 1 is entirely open as to the operating conditions of the claimed process.

11.11 Furthermore, the materials mentioned in said table of the patent in suit are not fully characterised, e.g. in terms of their macro and micropores, or calcium content, which according to D5 (page 8, last paragraph) greatly influences the adsorption capacity of nitrous oxide.

Therefore, it is not apparent
- that the alleged correlation or trend appearing from the data shown in that table indeed depends only on kinetic factors and particle size and shape, and not also to some extent on other factors such as calcium content, macro- and micropore system and/or operating conditions,
- let alone that the alleged results are achieved across the whole breadth of Claim 1 at issue.

11.12 The Board thus concludes that no improvement whatsoever over the process of D1, directly attributable to some features of claimed process, in particular to the nitrogen diffusion parameter value, across the whole breadth of Claim 1, was conclusively made plausible or demonstrated by evidence, let alone across the full breadth of claim 1 at issue.

12. Reformulated technical problem

12.1 Since the technical problem cannot be seen in providing a process that is improved in comparison to the process according to the closest prior art D1, it has to be reformulated in a less ambitious manner.

12.2 Starting from the process described in the example of D1, and also considering the data contained in the table of paragraph [0032] and in Figure 1 of the patent in suit, the technical problem solved can plausibly be seen in the provision of a further process for removing water, carbon dioxide and nitrous oxide from a feed gas stream, whereby an almost complete removal of nitrous oxide can be achieved whilst preventing early breakthrough of the gases, in particular breakthrough of nitrous oxide prior to carbon dioxide.

13. Success of the claimed solution

As apparent from the data reported in the patent in suit (see table [0032] and figure 1) this less ambitious problem is indeed solved by the process according to claim 1 (see also points 11.2 to 11.5 *supra*). This is not in dispute.

14. Obviousness

14.1 It remains to be decided whether starting from the closest prior art D1 the claimed solution was obvious for the skilled person in the light of common general knowledge and the prior art invoked by the appellant.

14.2 Document D1 taken alone

14.2.1 As regards D1, it can be gathered from its Figure 3 (and the relevant description thereof on page 5, lines 16-17) that the use of binderless CaX zeolite as adsorbent permits obtaining the simultaneous breakthrough of nitrous oxide and carbon dioxide, i.e. preventing nitrous oxide from breaking through before carbon dioxide. This appears to be the very same prevention of early breakthrough sought-for according to paragraph [0042] of the patent in suit.

14.2.2 Still D1 (paragraph [0024], table 1) illustrates the Henry's Law constants for nitrous oxide and carbon dioxide of a number of adsorbents, as well as the Henry's Law selectivity (ratio of Henry's Law constants), as follows:

Adsorbent	(mmole/g/atm) K_HCO_2	(mmole/g/atm) K_HN_2O	S N_2O/CO_2
Alcan AA-300 alumina	5.6	0.45	0.08
UOP 13X	162	63	0.39
UOP 5A	145	54	0.37
Binderless CaX	1031	1035	1.00
Na-mordenite	366	185	0.51
Ca-mordenite	374	113	0.30
CaX	1020	503	0.49
BaX	155	79	0.51

14.2.3 According to D1 (paragraphs [0022] and [0025]), calcium exchanged X zeolite (CaX) is a preferred third adsorbent, which satisfies the requirement of D1 of a minimum adsorption capacity for nitrous oxide of at least 500 mmole/g/atm.

14.2.4 The zeolite material disclosed by D1 such as CaX zeolite is used in a three-layer bed, through which the feed gas stream flows (this is apparent from Figure 1 of D1).

14.2.5 D1 itself is silent as regards the CaX particle size and shape or their macro- and micropore system, so that nothing is disclosed as regards the kinetic aspects.

14.2.6 However, it appears to be generally known in the field of adsorption processes that:

(a) Adsorption kinetics need consideration, as the overall rate of adsorption is controlled by e.g.

mass transfer (P1, page 166, "Kinetics of sorption in batch systems", first full paragraph).

(b) Micro- and macropore resistances are important and depend on the ratio of diffusional time constants, which widely depend on system and conditions (P1, page 167, first paragraph; page 173, Point 6.3).

(c) The diffusional time constant depends on the ratio $D_c/r^2{}_c$, or $D_p/R^2{}_p$, which in the patent in suit is defined as diffusion parameter (P1, page 168, last paragraph, last line; page 174, lines 15-18).

14.2.7 This also appears to be generally known in the field of pressure swing gas adsorption (e.g. P3, Point 5.1.3, first paragraph, last sentence, and equations 5.15 and 5.16), according to which kinetic selectivity is important and, in the widely used models for full simulation of PSA systems, the diffusion parameters ("$D_p/R^2{}_p$" or "$D_c/r^2{}_c$") play a role.

14.2.8 Nevertheless, even considering this common general knowledge on adsorption's kinetic aspects, there is no hint in D1 to choose a calcium exchanged zeolite X material with a particular diffusional time constant, i.e. a nitrogen diffusional parameter value of 0.12 s^{-1} or higher.

14.3 Combination of document D1 with document D2

14.3.1 Calcium exchanged X zeolites suitable for removal of nitrous oxide were known at the effective filing date of the patent in suit, *inter alia* from D2 (*supra*), which fact is not in dispute.

14.3.2 The Siliporite CaX zeolite adsorbents used in the process illustrated in Example 3 of D2, which prevents breakthrough of nitrous oxide prior to carbon dioxide and attains complete removal of nitrous oxide, is a suitable CaX third adsorbent for the process of D1.

14.3.3 It is not in dispute, but expressly stated in the patent in suit (paragraph [0034]) that the Siliporite G586 of D2 in form of beads of 1.6x2.5 mm mentioned in D2 has a nitrogen diffusion parameter which may range from 0.10 to 0.14 s^{-1}.

14.3.4 Also not in dispute is the fact that the zeolite adsorbent materials coming from the manufacturers present variations, so that the batches are usually screened before they are packed into beds. This is apparent, on the one hand, from the assertion in the patent in suit (paragraph [0024], last sentence) and, on the other hand, from the arguments of the appellant e.g. during the oral proceedings.

14.3.5 By carrying out the routine batch screening of CaX zeolite material such as Siliporite G586 mentioned in D2, the skilled person would identify the zeolite material, which, as prescribed by D1, has the highest nitrous oxide capacity, and which, since it is intended to be packed in a bed, also offers the appropriate kinetic selectivity. Proceeding in this manner, the skilled person would obviously choose the batch with the particle size and shape offering the highest diffusion parameter, which within the variation range 0.10-0.14 s^{-1} corresponds to a nitrogen diffusion parameter being higher than the minimum of 0.12 s-1 defined in Claim 1, which lies in the middle of the variation range.

14.3.6 By replacing the material used according to D1 by the adsorbent material identified as explained hereinbefore, the skilled person would arrive at a process falling within the terms of claim 1 without inventive ingenuity.

14.4 Hence, the process according to claim 1 as granted does not involve an inventive step (Articles 52(1) and 56 defined in Article 56 EPC).

15. The respondent's main request is thus not allowable.

Auxiliary request 1

Amendments

16. Independent Claims 1, 16, 18 and 19 at issue differ from independent claims 1, 17, 19 and 20 in terms of the minimum value of the numerical range specified for the "*nitrogen diffusion parameter*". In the independent claims at issue, the range "***0.15*** *sec*$^{-1}$ *or higher*" replaces the range "*0.12 sec*$^{-1}$ *or higher*" of the granted claims.

16.1 No formal objections were raised against the set of claims according to the request at issue.

16.2 Said amendment finds a basis on on page 4, lines 6 to 7, and in claim 3 of the application as filed. The incorporation of a higher lower limit for said parameter restricts the scope of the independent claims.

Hence the amendments made meet the requiremnts of Article 123, paragraphs (2)and (3), EPC:

16.3 The amendments were made to overcome grounds of opposition under Article 100(a) EPC and thus fulfil also the requirements of Rule 80 EPC.

16.4 Finally, no issues under Article 84 EPC arise from the amendments.

Sufficiency of the disclosure

17. The further amendments made to the claims have no bearing on the considerations and conclusions of the board concerning sufficiency of disclosure with regard to the claims as granted (points 3 to 3.8.3 *supra*). No additional argument was put forward by the Appellant in this respect.

Hence, in the board's judgement, the invention as defined in the claims according to the auxiliary request 1 is not objectionable under Article 100(b) EPC either.

Novelty

18. Novelty of the subject-matter of the claims according to auxiliary request 1 was not called into question by the appellant.

Considering that none of the documents relied upon by the appellant discloses an adsorbent for removing nitrous oxide with a nitrogen diffusion parameter of 0.15 sec^{-1} or more, let alone in combination with a nitrous oxide capacity of 80 mmol/g/bar or more, the board is also satisfied that the subject-matter of claims 1 to 19 is novel (Article 52(1) and 54(1)(2) EPC).

Inventive step

18.1 The invention is claimed in terms of process, apparatus and use claims, all of which comprise as a feature the presence of a nitrous oxide adsorbent with a nitrogen diffusion parameter of 0.1**5** sec^{-1} or higher and a nitrous oxide capacity of 80 mmol/g/bar (79 mmol/g/atm) or higher at 30°C.

18.2 The process according to Claim 1

18.2.1 Claim 1 at issue is directed, like claim 1 according to the main request, to a process for removing water carbon dioxide and nitrous oxide from a feed gas stream wherein said adsorbent is used as third adsorbent to adsorb nitrous oxide.

18.2.2 Considering the amendments made, D1 can thus still be considered to constitute the closest prior art. This was not in dispute.

18.2.3 Considering that there is also no evidence on file which convincingly demonstrates that the process according to amended claim 1 at issue is somehow improved over the process described in D1, there is no reason for changing the technical problem as formulated when assessing claim 1 according to the main request (point 12.2 *supra*).

18.2.4 As already acknowledged (Points 11.2 to 11.5, *supra*), higher nitrogen diffusion parameter values were shown to lead to increased nitrous oxide removal efficiency in combination with a nitrous oxide capacity as specified in Claim 1 (see in particular paragraph [0032] and figure 1 of the patent in suit).

Considering that the lower limit for said parameter according to claim 1 at issue is even higher than the one according to claim 1 of the main request, the board concludes that the technical problem posed must also be solved by the process according to claim 1 at issue.

18.2.5 As regards obviousness, the board notes that there is no evidence on file convincingly showing that one of the adsorbents described in the prior art invoked by the appellant, including D1 and D2, has a nitrogen diffusion parameter of at least 0.15 s^{-1}, let alone in combination with a nitrous oxide capacity of 80 mmol/g/bar or more. Nor is it apparent from the evidence on file that one of the known CaX zeolite type materials was ever produced and made available, with a diffusion parameter value lying within the range according to claim 1 at issue, as a result of usual variations between commercially produced batches.

18.2.6 Moreover, the following information must also be taken into account:

(a) E1 mentions (paragraph [0017]) that CaX zeolite materials not only have a reduced adsorption capacity for carbon dioxide but also worse kinetics than zeolite 13X, because of the increased mass transfer front (zone), which greatly reduces the operating time. However, to overcome these drawbacks, E1 (paragraphs [0008], [0009] and [0013]; Example 3, paragraphs [0024] to [0027]) discloses a process for removing *inter alia* nitrous oxide from air, by adsorption on CaX molecular sieve adsorbent, extensively exchanged (e.g. 86%) with calcium, and achieving a nitrous oxide removal of 98%.

(b) D5 too concerns a process for removing nitrous oxide from air streams and stresses that calcium exchange greatly influences the adsorption capacity (page 6, lines 21-23; page 8, lines 26-30) for nitrous oxide. D5 illustrates that by increasing the calcium exchange percentage of CaX zeolites in form of spheres of 2 mm from 60 to 86% (Examples 4 and 6; Figures 3 and 5) nitrous oxide does not breakthrough before carbon dioxide, in any case not before 60 minutes. Although D5 mentions spherical zeolites having a size of 2 mm (which in any case is bigger than the 1.6 mm mentioned in the patent in suit), nothing is said as regards their kinetic characteristics, let alone in terms of their nitrogen diffusion parameter.

18.2.7 Consequently, there is no evidence on file that the use of an adsorbent having a diffusion parameter and a nitrous oxide capacity according to Claim 1 at issue was, at the effective filing date of the patent in suit, a known option available to the skilled person, or something that might have become available upon carrying out usual screening tests on CaX zeolite materials of the type disclosed in any of D1, D2, D5 or E1.

18.2.8 Indeed, as apparent from the examples in the patent in suit, to obtain adsorbents with diffusion parameters of at least 0.15 s^{-1}, it is necessary to specifically produce adsorbent materials with tailored size and shape and appropriate micro- and macropores. This is, however, something the skilled person - unaware of the present invention an absent any hints to this end in the prior art - would not obviously consider when trying to solve the posed technical problem. Since the prior art relied upon by the appellant does not, however, contain hints inducing the skilled person to consider in particular the kinetic characteristics of the adsorbents implied by a nitrogen diffusion parameter as claimed, the use of such an adsorbent in a process as described in D1 was not a solution that the skilled person would obviously consider.

18.2.9 Therefore, in the board's judgement, the process defined in Claim 1 according to auxiliary request 1 and, consequently, the processes defined in claims 2 to 15 dependent thereon, involve an inventive step (Articles 52(1) and 56 EPC.

18.3 Claims 16 to 19

18.3.1 Apparatus claims 16 and 17, process Claim 18 and use Claim 19 all require, as a mandatory feature, the presence and involvement of an an adsorbent for removing nitrous oxide as defined in claim 1, i.e. "*having a nitrogen diffusion parameter of 0.15* sec^{-1} *or higher and a nitrous oxide capacity of 80 mmol/g/bar (79 mmol/g/atm) or higher at 30°C*".

18.3.2 Hence, for analogous reasons, and absent any argumentation to the contrary, besides the one developed with regard to the process of Claim 1, the Board concludes that the subject-matters of these claims is not obvious in the light of the prior art invoked by the appellant.

18.3.3 Their subject-matters thus also involve an inventive step (Article 52(1) and 56 EPC).

Order

For these reasons it is decided that:

The case is remitted to the Opposition Division with the order to maintain the patent on the basis of the claims according to the auxiliary request 1 submitted on 1 September 2011 and a description to be adapted thereto where appropriate and figures as granted.

The Registrar: The Chairman:

Beschwerdekammern des Europäischen Patentamts
Boards of Appeal of the European Patent Office
Chambres de recours de l'Office européen des brevets

D. Magliano B. Czech

Decision electronically authenticated