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Datasheet for the decision of 5 February 2009

Case Number: T 1556/05 - 3.3.07

Application Number: 97109191.3

Publication Number: 0811423

IPC: B01J 20/28

Language of the proceedings: EN

Title of invention:
A material with microporous crystalline walls defining a narrow sized distribution of mesopores, and process for preparing same

Applicant:
Intevep SA

Headword:
-

Relevant legal provisions:
EPC Art. 123(2)

Relevant legal provisions (EPC 1973):
EPC Art. 111

Keyword:
"Amendments not allowable (main request)"
"Fresh case (auxiliary request) - remittal"

Decisions cited:
-

Catchword:
-

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Case Number: T 1556/05 - 3.3.07

D E C I S I O N
of the Technical Board of Appeal 3.3.07
of 5 February 2009

Appellant: Intevep SA
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Decision under appeal: **Decision of the Examining Division of the European Patent Office posted 13 June 2005 refusing European application No. 97109191.3 pursuant to Article 97(1) EPC.**

Composition of the Board:

Chairman: S. Perryman
Members: F. Rousseau
G. Santavicca

Summary of Facts and Submissions

I. The appeal lies from the decision of the examining division refusing European application No. 97109191.3, published under No. 0 811 423. The application as filed comprised seventeen claims, claims 1 and 11 reading as follows:

"1. A composition of matter, comprising a crystalline material having at least one microporous crystalline phase having a micropore volume of at least about 0.15 cc/g distributed in channels between about 3 to about 15 Å in average diameter, and having a mesopore volume of at least about 0.1 cc/g distributed in channels between about 20 to about 100 Å in average diameter, whereby said mesopore volume renders said micropore volume accessible.

11. A process for preparing a crystalline molecular sieve material, comprising the steps of:
providing a suspension of nuclei of a crystalline molecular sieve material in an aqueous media;
mixing the suspension with a water soluble tensoactive (sic) organic compound to provide a mixture of the nuclei and micelles of the tensoactive (sic) organic compound in solution;
inducing nucleation of the nuclei so as to provide a solid material having the organic compound dispersed therein;
extracting the organic compound from the solid material to provide a crystalline molecular sieve material having a micropore volume greater than or equal to about 0.15 cc/g distributed in channels about 3 to about 15 Å in average diameter, and having a mesopore

volume of at least about 0.1 cc/g distributed in channels about 20 Å to about 100 Å in average diameter."

II. The contested decision was based on four sets of amended claims submitted at oral proceedings held on 07 April 2005. The application was refused as the amended claims either (i) were not based on the application as filed in contravention of Article 123(2) EPC (main, first and second auxiliary requests), or (ii) did not meet the requirements of Article 84 EPC as they did not contain all the essential features necessary to carry out the invention (first and third auxiliary requests), or (iii) lacked an inventive step over document (1) (EP-A-0 206 871), because they encompassed embodiments which did not solve the problem addressed by the application (third auxiliary request).

III. With the statement setting out the grounds of appeal filed on 13 October 2005, the appellants submitted six sets of amended claims as their main and first to fifth auxiliary requests respectively.

IV. In a communication by the Board accompanying the summons to attend oral proceedings to be held on 5 February 2009, the subject-matter of the claims then on file was, in view of the numerous parametric definitions contained therein, objected to as to lacking clarity, adequate support in the description and sufficiency of disclosure. Document (3) (US-A-5 250 282) was cited in the Board's communication and introduced into the proceedings as being relevant for assessing novelty and inventive step.

V. In reply to the Board's communication, all requests then on file were replaced by a main and sixteen auxiliary requests, which in turn in the course of the oral proceedings before the Board held on 5 February 2009 were replaced by a new main request and a new auxiliary request both consisting of one claim, reading as follows:

Main request

"1. A process for preparing a crystalline molecular sieve material, comprising the steps of:

providing a suspension of nuclei of a crystalline molecular sieve material in an aqueous media;

mixing the suspension with a water soluble tensoactive (sic) organic compound to provide a mixture of the nuclei and micelles of the tensoactive (sic)organic compound in solution;

inducing aggregation of the nuclei so as to provide a solid material having the organic compound dispersed therein;

extracting the organic compound from the solid material to provide a crystalline molecular sieve material having a micropore volume greater than or equal to 0,21 cc/g, determined from equilibrium N2 adsorption capacity at different N2 partial pressures, according to ASTM standard method D 4365-85,
the micropore volume distributed in channels between 3 to 15 ANGSTROM in average diameter, as measured by x-ray diffraction, the x-ray diffraction pattern

exhibiting at least two lines at d-spacings of less than 15 ANGSTROM,

the crystalline molecular sieve material having a mesopore volume of at least 0,12 cc/g, determined according to ASTM standard method D 4641-93, the mesopore volume being distributed in channels of 50 ANGSTROM in average diameter, as determined from equilibrium N2 desorption isotherm, according to ASTM standard method D 4641-93,

wherein the nuclei of the microporous crystalline molecular sieve material comprise faujasite type zeolite nuclei,

wherein the zeolite nuclei are mixed with a basic solution containing 1 cationic surfactant,

wherein this mixture is then aged at temperatures of 80 DEG C for 24 hours,

wherein a resulting solid is then washed in water and dried,

and wherein the tensoactive (sic) organic compound is then extracted by calcination in air."

Auxiliary request

"1. A process for preparing a crystalline molecular sieve material, comprising the steps of:

- preparing a Suspension A by vigorously stirring a mixture of 20.5 g of sodium silicate (29% wt SiO2; 9.3% wt Na2O; 61.7% wt H2O) and 6.8 mL of a 13.4 M NaOH solution,
- preparing a Solution B by dissolving 1.38 g of sodium aluminate (49.1% wt Al2O3; 27.2% Na2O, 23.7% H2O) into 8.2 mL of a 5.9 M solution of NaOH,
- adding Suspension A over Solution B, while stirring the latter,
- after the addition is complete, continuing the stirring for one more hour, resulting in a clear suspension of nuclei having the following composition: 1 SiO2 : 0.066 Al2O3 : 1.06 Na2O : 15.1 H2O
- aging this suspension at room temperature for 2.7 hours, or for 24 hours,
- thereafter adding a solution of 7.98 g of cetyltrimethylammonium bromide (CTAB) in 10 g of water, resulting in a second suspension having the following composition: 1 SiO2 : 0.066 Al2O3 : 1.06 Na2O : 0.22 CTAB : 21 H2O
- aging this second suspension for 24 hours at a temperature of 80 DEG C,
- washing the resulting solid,
- drying the solid at 100 DEG C for 4 hours,
- then calcining the solid at 500 DEG C for 6 hours in a flow of air,

the calcined product showing an x-ray diffraction pattern having the main strong signals as set forth

below in Table 1, or in Table 2, respectively:

TABLE 1

d-spacing (Å)	Relative intensity (%)
14.5	100
8.9	18
7.6	14
5.8	38
4.8	17
4.4	25
4.0	14
3.8	71
3.6	9
3.3	65
3.1	19
3.0	18
2.9	45
2.8	16
2.8	11
2.7	14
2.6	5
2.4	9

TABLE 2

d-spacing (Å)	Relative intensity (%)
14.5	100
8.9	21
7.6	13
5.8	32
4.8	16
4.4	20
4.0	12
3.8	69
3.6	7
3.3	66
3.1	16
3.0	17
2.9	49
2.8	19
2.8	12
2.7	15
2.4	6

."

VI. The appellants argued that Claim 1 of the main request was essentially based on claim 11 as filed, wherein the values for the micropore volume, the mesopore volume, the size of the channels, the temperature and the

duration of the aging of the solution containing the faujasite type zeolite nuclei and the cationic surfactant originate from examples 1 and 2 as originally filed. Claim 1 of the auxiliary request was based on the examples 1 and 2 as filed.

VII. The appellants requested that the decision under appeal be set aside and that a patent be granted on the basis of the claim of the new main request or of the claim of the new auxiliary request, both submitted at oral proceedings on 5 February 2009.

VIII. At the end of the oral proceedings the Board's decision was pronounced.

Reasons for the Decision

1. The appeal is admissible.

Main request

2. Amendments

The process of claim 1 is partly based on claim 11 as originally filed, wherein the process is defined as to lead to a crystalline molecular sieve material which exhibits *inter alia* a micropore volume greater than or equal to 0,21 cc/g, the micropore volume being distributed in channels between 3 to 15 Å and a mesopore volume of at least 0,12 cc/g, the mesopore volume being distributed in channels of 50 Å in average diameter. The appellants referred to examples 1 and 2 of the application as filed as forming the basis for

supplementing claim 11 as originally filed with the features that the crystalline molecular sieve material obtained should have the above mentioned microporous and mesoporous properties. No other passage disclosing a micropore volume of 0,21 cc/g or a mesopore volume of 0,12 cc/g can be found in the application as filed. Thus, it has to be established whether or not the particular processes disclosed in examples 1 and 2 form a proper basis for the amendments proposed.

Example 1 and 2 in the application as filed disclose both the preparation of a molecular sieve material where in a first step a suspension of nuclei of composition 1 SiO_2 : 0.066 Al_2O_3 : 1.06 Na_2O : 0.22 cetyltrimethylammonium bromide (CTAB): 21 H_2O is obtained, from which after aging a solid is extracted, washed, dried at 100 °C for 4 hours and calcined at 500 °C for 6 hours in a flow of air, leading to a calcined product showing an x-ray diffraction pattern having the specific main strong signals as set forth in Table 1 for the molecular sieve material of example 1 or in Table 2 for the molecular sieve material of example 2. The molecular sieve material of example 1 is described to have a micropore volume of 0.25 cc/g and a mesopore volume of 0.12 cc/g distributed in pores having diameters comprised between 40 and 60 Å in diameter. The molecular sieve material obtained in example 2 has a micropore volume of 0.21 cc/g and a mesopore volume of 0.15 cc/g distributed in pores having diameters comprised between 50 and 70 Å in diameter.

In the Board's judgement, the skilled person derives from each of these examples nothing more than the bare disclosure of the specific characteristics of these

processes, namely the combination of particular nuclei (SiO_2 : 0.066 Al_2O_3 : 1.06 Na_2O : 0.22 cetyltrimethylammonium bromide (CTAB): 21 H_2O), with a particular thermal treatment leading to a specific crystalline structure (as shown in Tables 1 and 2) and to a particular mesopores distribution (diameters of the pores comprised either between 40 and 60 Å or 50 and 70 Å). The application as originally filed does not however provide any basis for a lower limit for the micropore volume of 0.21 cc/g in combination with any type of faujasite zeolite nuclei. It also does not provide any basis for a mesopore volume of at least 0.12 cc/g, the mesopore volume being distributed in channels of 50Å on average (regardless of the distribution) and in combination with any type of faujasite zeolite nuclei.

As a result of the omission in present claim 1 among others of the definition of the constituents of the nuclei in the suspension, of the x-ray structure obtained and the specific distribution of mesopore diameters, which are disclosed in examples 1 and 2 in combination with either a micropore volume of 0,21 cc/g or a mesopore volume of 0,12 cc/g, claim 1 represents an undue generalisation of the particular embodiments of examples 1 and 2 which extends beyond the content of the application as filed.

Hence, the main request must be rejected pursuant to Article 123(2) EPC.

Auxiliary request

3. Claim 1 of the auxiliary request defines two processes, which exactly correspond to those shown in examples 1 and 2 of the application as filed. The macro- and mesoporosity of the product is not explicitly defined, but however considered to automatically result from the process steps listed in claim 1. Claim 1 thus meets the requirements of article 123(2) EPC. The objection under Article 84 EPC 1973 that the claimed subject-matter did not contain all the essential features in order to obtain products meeting the parametric definition of former claim 1 in relation to their micro- and mesoporosity, has been also removed by restricting claim 1 to the specific processes of examples 1 and 2.

4. The decision under appeal did not consider claim 1 in the form of the present auxiliary request, as such request was submitted only on appeal in response to the objections raised by the Board. Thus, claim 1 of the auxiliary request represents a fresh case not yet examined by the first instance. It is also noted that the search did not reveal prior art such as document (3) (US-A-5 250 282), which relates to the synthesis of mesoporous crystalline oxides comprising the formation of surfactant micelles in aqueous solutions, the micelles serving as template for the formation of the mesopores, which is therefore more relevant for assessing inventive step of the claimed invention than the documents cited in the search report. Under these circumstances the Board considers it appropriate to exercise the power conferred by Article 111(1), second sentence, EPC 1973 to remit the present fresh case to the examining division for further prosecution, in

particular in view of document (3) and further relevant prior art, if any.

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 new auxiliary request submitted at oral proceedings on 5 February 2009.

The Registrar: The Chairman:

C. Eickhoff S. Perryman