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D E C I S I O N
of 26 September 1995

Case Number: T 0953/92 - 3.3.2

Application Number: 84870180.1

Publication Number: 0146525

IPC: C01B 33/18

Language of the proceedings: EN

Title of invention:
Process for preparing crystalline silicas

Patentee:
FINA TECHNOLOGY, INC.

Opponent:
Mobil Oil Corporation

Headword:
Crystalline silicas II/FINA

Relevant legal provisions:
EPC Art. 56, 123(2)

Keyword:
"Inventive step (no)"
"Limitation of range (allowable)"

Decisions cited:
T 0201/83

Catchword:
-

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Case Number: T 0953/92 - 3.3.2

D E C I S I O N
of the Technical Board of Appeal 3.3.2
of 26 September 1995

Appellant:
(Proprietor of the patent)

FINA TECHNOLOGY, INC.
8350 North Central Expressway
Dallas, Texas 75206 (US)

Representative:

Detrait, Jean-Claude
c/o Fina Research S.A.
Patent Department
Zone Industrielle C
B-7181 Feluy (BE)

Respondent:
(Opponent 01)

Mobil Oil Corporation
150 East 42nd Street
New York, N.Y. 10017 (US)

Representative:

Cooper, John Anthony
CARPMAELS & RANSFORD
43, Bloomsbury Square
London WC1A 2RA (GB)

Decision under appeal:

Decision of the Opposition Division of the European Patent Office posted 17 August 1992 revoking European patent No. 0 146 525 pursuant to Article 102(1) EPC.

Composition of the Board:

Chairman: P. A. M. Lançon
Members: G. J. Wassenaar
J. Van Moer

Summary of Facts and Submissions

I. European patent No. 0 146 525 was granted in response to European patent application No. 84 870 180.1.

II. A notice of opposition was filed by the Respondent (Opponent). Revocation of the patent in its entirety was requested on the grounds of lack of novelty, lack of inventive step and insufficient disclosure (Articles 52, 54, 56, 83, 100(a) and 100(b) EPC).

Amongst others, the following documents were cited:

(1) Nature, 296, 530-533 (1982)
(3) The Journal of Physical Chemistry, 83, 2777-2781 (1979)
(8) US-A-4 061 724
(11) Chemistry Letters, 1013-1016 (1981)

III. The Opposition Division revoked the patent. The decision was taken on the basis of the claims as granted as main request and five new sets of claims as auxiliary requests 1 to 5.

They considered that claim 1 as granted (main request) and claim 1 of auxiliary request 4 lacked novelty over (1), that claim 1 according to auxiliary request 1 extended beyond the content of the application as filed (Article 123(2) EPC), that auxiliary requests 2 and 3 lacked clarity (Article 84 EPC) and that claim 1 of auxiliary request 5 lacked an inventive step over (1) in combination with (11).

IV. The Appellant (Patentee) lodged an appeal against this decision.

With the statement of grounds, the Appellant filed six sets of claims as first to sixth auxiliary request. With reference to decisions T 324/89 and T 301/87, it was argued that it was not possible to reject the auxiliary request under Article 84 because the deficiency, if any, did not arise out of the amendments themselves.

With respect to the amendment in the second, fourth and sixth auxiliary request ("with the proviso that SiO_2/Al_2O_3 molar ratio and Si/Al atomic ratio are both at least 80"), it had been pointed out that it was accepted under Article 123(2) EPC by the Opposition Division and thus no longer needed to be discussed.

V. The Respondent disagreed with the Appellant's submissions and maintained that the new auxiliary requests 1 to 4 violated Article 123(2) EPC, that auxiliary requests 2, 4, 5 and 6 lacked clarity within the meaning of Article 84 EPC, that auxiliary request 4 lacked novelty, that auxiliary request 6 lacked an inventive step and that, because of the ambiguous nature of the expression "silica to alumina atomic ratio", the invention was insufficiently disclosed. Amongst others, reference was made to the following document mentioned for the first time in the proceedings:

(14) Journal of Catalysis 61, 390-396 (1980)

VI. Oral proceedings were held on 26 September 1995. With the consent of the parties they took place one day earlier than the scheduled date. During oral proceedings the Appellant abandoned the claims as granted and made the set of claims filed as first auxiliary request with the statement of grounds his main request and renumbered the other auxiliary requests 1 to 5.

In the novelty and inventive step discussion special attention was drawn to documents (1), (3), (11) and (14).

The independent claims of the requests on file read as follows:

Main request:

A process for preparing crystalline silicas having the monoclinic symmetry which comprises the step of calcining in air for 3 hours to 10 hours, at a temperature of at least 500°C a crystalline silica of the silicalite type having a silica to alumina atomic ratio of at least 80.

Auxiliary request 1:

A process for preparing crystalline silicas having the monoclinic symmetry which comprises the step of calcining in air for 3 hours to 10 hours, at a temperature of at least 500°C a crystalline silica of the silicalite type with the proviso that SiO_2/Al_2O_3 molar ratio and Si/Al atomic ratio are both at least 80.

Auxiliary request 2:

A process for preparing crystalline silicas having the monoclinic symmetry which comprises the step of calcining in air for 3 hours to 10 hours, at a temperature of 500°C to 650°C a crystalline silica of the silicalite type having a silica to alumina atomic ratio of at least 80.

Auxiliary request 3:

A process for preparing crystalline silicas having the monoclinic symmetry which comprises the step of calcining in air for 3 hours to 10 hours, at a temperature of 500°C to 650°C a crystalline silica of the silicalite type with the proviso that SiO_2/Al_2O_3 molar ratio and Si/Al atomic ratio are both at least 80.

Auxiliary request 4:

A process for preparing crystalline silicas having the monoclinic symmetry which comprises the step of calcining in air for 3 hours at a temperature of at least 500°C a crystalline silica of the silicalite type having a silica to alumina atomic ratio of at least 80.

Auxiliary request 5

A process for preparing crystalline silicas having the monoclinic symmetry which comprises the step of calcining in air for 3 hours, at a temperature of at least 500°C a crystalline silica of the silicalite type with the proviso that SiO_2/Al_2O_3 molar ratio and Si/Al atomic ratio are both at least 80.

VII. The Appellant requested that the decision under appeal be set aside and that the patent be maintained on the basis of a set of claims according to one of the six auxiliary requests filed on 16 December 1992, with the statement of grounds, renumbered main request and auxiliary requests 1 to 5.

The Respondent requested that the appeal be dismissed.

Reasons for the Decision

1. The appeal is admissible.

2. *Main request*

2.1 General remarks

Claim 1 contains an obvious mistake since silica and alumina relate to molar species and not to atomic species. If the expression "silica to alumina atomic ratio" is to be understood as silica to alumina molar ratio, then the silicon to aluminium atomic ratio is only half its value. For the issue of novelty and inventive step the ambiguity becomes irrelevant if it can be shown that the claim even in its most narrow interpretation, i.e. with a silicon to aluminium atomic ratio of at least 80, lacks novelty or an inventive step. Under this provision, clarity is not at issue in the present case.

2.2 Allowability of amendments under Article 123(2) EPC

Present claim 1 differs from claim 1 as granted in the introduction of an upper limit of 10 hours for the calcination time. The figure of 10 hours is based on Example 1 of the application as originally filed, disclosing a calcination time of 10 hours at 600°C. The Opposition Division did not allow this amendment because of lack of support for the upper time limit of 10 hours in connection with the claimed temperature range; point II, 3.2 of the contested decision.

In the Board's opinion, in agreement with the case law on this point (T 201/83, OJ EPO, 1984, 481), it is generally allowable to take a figure from an Example to define a more limited range, provided the skilled person

could have readily recognised this figure as not so closely associated with the other features of the example that it determines the effect of that embodiment of the invention in a unique manner. Since no special effect is associated with the calcination condition of 10 hours at 600°C and in view of the originally disclosed preferred calcination time range of 5 to 72 hours and preferred temperature range of 500°C to 650°C (claims 3 and 5 as originally filed), the skilled person would expect about the same result for a calcination time of 10 hours at temperatures between 500°C and 600°C and above 600°C.

The new range also satisfies the novelty test with respect to the application as filed since Example 1 would clearly destroy the novelty of present claim 1.

Thus the Board is of the opinion that present claim 1 does not contain subject matter which extends beyond the content of the application as filed so that it satisfies the requirements of Article 123(2) EPC.

2.3 Novelty

2.3.1 Document (1), page 531, right column, discloses the structure determination of calcined silicalite by magic-angle spinning nuclear magnetic resonance (MASNMR). The sample of silicalite of very high purity and crystallinity was prepared according to Example 4 of ref. 2, which is the basic silicalite patent referred to as document (8) in these proceedings, followed by calcination in air at 500°C for 16 hours. The X-ray powder diffraction pattern of the same sample as given in Fig. 4 reveals the typical pattern for monoclinic symmetry, i.e. the peak doublets at 24.4° and 29.2°. The Si/Al atomic ratio of the sample was at least 1000 (page 533, left column). This document confirmed the

close structural relationship between silicalite and ZSM-5 as earlier observed by Wu et al.(3) and Olsen et al.(14), both referred to in (1).

2.3.2 Present claim 1 differs from the process disclosed in (1) in that the calcination time is limited to 3 to 10 hours.

Document (14), relating to the chemical and physical properties of the ZSM-5 substitutional series, discloses that "silicalite" appears to be a member of the ZSM-5 substitutional series. It discloses experiments performed with ZSM-5 members with a silica to alumina molar ratio of 1600. It also discloses that the ammonium form of the ZSM-5 members was converted to the hydrogen form by calcination in air for 10 hours at 538°C (pages 394 and 395). Thus according to (14), high silica ZSM-5 has ion-exchange properties. This is contrary to the disclosure in (8), which is generally considered as the basic document on silicalite, and in which it is stated in column 3, lines 3 to 7 that silicalite exhibits no ion-exchange properties and that this is highly advantageous for use in aqueous environments. In view of this, the Board has still some doubts whether the said ammonium form of ZSM-5 should be considered to be of the "silicalite type" within the meaning of the patent in suit. Present claim 1 is therefore also considered to be novel over (14).

Since none of the other citations explicitly discloses a calcination time within the range of present claim 1, its subject matter is considered novel.

2.4 Inventive step

2.4.1 The closest prior art is (1), which, as indicated above, discloses the transformation of freshly prepared (orthorhombic) silicalite into monoclinic silicalite by calcination in air at 500°C for 16 hours.

The Appellant has not indicated any advantage of the process of present claim 1 over the process as disclosed in (1).

The technical problem underlying the invention is therefore merely to be seen in providing an alternative process for preparing monoclinic silicas.

According to claim 1, this problem is solved by reducing the calcination time to 3 to 10 hours.

From Examples 1 and 3, disclosing monoclinic transformation at calcination times of 10 and 3 hours respectively, it is credible that the process of claim 1 actually solves the said problem.

2.4.2 It remains therefore to be decided if it was obvious to modify the process of (1) in the way indicated in present claim 1.

From documents (1), (3) and (14), the skilled person knew that "silicalite" is structurally very closely related, if not identical, to ZSM-5 with a low alumina content. The skilled person, trying to find alternative processes for obtaining monoclinic silicas will therefore certainly take into account the known process conditions for obtaining monoclinic ZSM-5 with a low alumina content.

The transformation of orthorhombic ZSM-5 to monoclinic ZSM-5 has been disclosed in (3) and (11).

Document (3), relating to factors affecting the crystal symmetry of ZSM-5-Type materials, discloses that for ZSM-5 with a silica to alumina molar ratio of about 1600 and a low sodium content a transformation already takes place after 1 hour at 600°C (page 2779, right column).

Document (11), relating to crystal symmetry change of ZSM-5 by various treatments, discloses the monoclinic phase transformation for ZSM-5 with silica to alumina molar ratios of from 169 at a calcination temperature of 540°C (Table 1). The calcination time is not disclosed, which indicates that the calcination time is not a very critical parameter.

Knowing that the phase transformation for ZSM-5 with a low alumina content can already take place one hour after calcination at 600°C, the skilled person would expect that the "silicalite" phase transformation disclosed in (1) was probably also obtainable at calcination times below 16 hours, and certainly if calcination temperatures higher than 500°C were used.

Thus, contrary to the submissions of the appellant, the Board is convinced that the skilled person, looking for an alternative process for the phase transformation of silicalite, would contemplate a shorter calcination time than in (1). The optimum calcination time, somewhere between 1 and 16 hours, could be easily determined by the skilled person. Therefore, the present choice of 3 to 10 hours must be considered obvious to the skilled person.

Claim 1 of the main request, therefore, lacks an inventive step in the meaning of Article 56 EPC.

3. *Auxiliary requests*

3.1 Claim 1 of auxiliary request 1 has the same scope as claim 1 of the main request in its most limited understanding; i.e. with a Si/Al atomic ratio of at least 80. The inventive step objection, therefore , equally applies to claim 1 of auxiliary request 1.

3.2 Claim 1 of auxiliary request 2 differs from claim 1 of the main request only in the introduction of an upper limit of 650°C for the calcination time. Since the documents relied on in the inventive step analysis under point 2, all disclose calcination times of between 500°C and 650°C, the inventive step objection equally applies to claim 1 of the second auxiliary request.

3.3 Claim 1 of auxiliary request 3 has the same scope as claim 1 of auxiliary request 2 in its most limited understanding. The inventive step objection with regard to auxiliary request 2, therefore, equally applies to claim 1 of auxiliary request 3.

3.4 Claim 1 of auxiliary request 4 differs from claim 1 of the main request in a limitation of the calcination time to 3 hours. The reasoning given above with respect to the inventive step objection of claim 1 of the main request does not only apply to the range of 3 to 10 hours but also to each individual member within this range. In the absence of a surprising technical effect any figure within the range of 3 to 10 hours must be considered to be the result of routine experimentation of the skilled person. A process limited to a calcination time of 3 hours, therefore, equally lacks an inventive step.

3.5 Claim 1 of auxiliary request 5 has the same scope as claim 1 of auxiliary request 4 in its most limited understanding. The inventive step objection with regard to auxiliary request 4, therefore, equally applies to claim 1 of auxiliary request 5.

4. *Objections under Article 83 EPC*

Since none of the requests are acceptable under Article 56 EPC it was not necessary to decide on this point.

5. Since none of the appellant's requests can be allowed, the appeal must be dismissed.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar: The Chairman:

P. Martorana P.A.M. Lançon