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DECISION
of 10 May 1994

Case Number: T 0311/91 - 3.3.2

Application Number: 88200703.2

Publication Number: 0292030

IPC: B01J 29/08

Language of the proceedings: EN

Title of invention:
Process for the preparation of modified zeolites

Applicant:
Shell Internationale Research Maatschappij B.V.

Opponent:
-

Headword:
Modified zeolites/SHELL

Relevant legal norms:
EPC Art. 54(2)

Keyword:
"Novelty (yes, after amendment of the claims)"

Decisions cited:
-

Headnote/Catchword:

Case Number: T 0311/91 - 3.3.2

DECISION
of the Technical Board of Appeal 3.3.2
of 10 May 1994

Appellant:

Shell Internationale Research
Maatschappij B.V.
Carel van Bylandtlaan 30
NL-2596 HR Den Haag

Representative: -

Decision under appeal: **Decision of the Examining Division of the European Patent Office dated 4 December 1990 refusing European patent application No. 88 200 703.2 pursuant to Article 97(1) EPC.**

Composition of the Board:

Chairman: P.A.M. Lançon
Members: M.M. Eberhard
E.M.C. Holtz

Summary of Facts and Submissions

I. European patent application No. 88 200 703.2 (publication No. 0 292 030) was refused by a decision of the Examining Division. The decision was based on the set of amended claims filed on 21 May 1990.

II. The ground for the refusal was that the subject-matter of Claims 1 to 9 lacked novelty with respect to the disclosure of document US-A-3 714 029 (hereinafter D1), in particular example 5 thereof. According to the decision, this document described the modification of a Y-faujasite zeolite by subjecting it to a treatment with a zinc solution and subsequently to a calcination at 800°C. The Examining Division interpreted the terms "Y-faujasite zeolite" used in D1 in view of the reference document D5, i.e. Molecular Sieve Catalysts, P. Michiels and O.C.E De Herdt, 1987, pages 39 to 42, and came to the conclusion that this zeolite had an alkali oxide/aluminium oxide molar ratio between 0.13 and 1. It was further pointed out that the process of D1 also comprised a treatment with a NH_4NO_3 solution but that the wording of Claim 1 did not exclude the inclusion of other process steps.

III. The Appellant lodged an appeal against this decision. In a communication of the Board pursuant to Article 11(2) RPBD, the Appellant's attention was drawn to example 1 of D1 and to further documents cited in the search report, namely US-A-3 322 690 (D2) and FR-A-2 030 800 (D4). The Appellant was informed that it was questionable whether the process of Claim 1 filed on 21 May 1990 was novel over the process disclosed in example 1 of D1, or in the examples 11 or 12 of D2, or in the examples 2, 4, 8 or 10 of D4. In reply to this communication the Appellant submitted an amended set of claims on 28 April 1994 and contended that in none of the examples of the cited documents a US-Y zeolite had been used as starting material.

IV. Oral proceedings were held on 10 May 1994. At these oral proceedings the Board raised the question whether sodium oxide contents as high as 13.5%wt were compatible with the fact that the starting material was an ultra-stable Y-zeolite (US-Y). In connection with the novelty issue, the Board made reference to document US-A-3 830 725 (hereinafter D3) cited in the search report. It was pointed out in this context that D3 described a process involving a treatment of a type Z-14 US zeolite, such as disclosed in US-A-3 449 070, with a solution containing a mixture of soluble nickel and magnesium salts and a calcination at a temperature of preferably about 1400°F (cf. column 2, lines 18 to 39) and that the stabilized zeolite of document US-A-3 449 070 referred to in D3 could contain sodium oxide in an amount of greater than 1 to about 4%wt. In reply thereto the Appellant submitted a new set of amended claims as the single request in the course of the oral proceedings.

Claim 1 reads as follows:

"1. Process for the preparation of a modified zeolite by treatment with a solution of one or more metal salts and a calcination which comprises treating an US-Y having a sodium oxide content of at least 2.2%wt and an alkali oxide/aluminium oxide molar ratio between 0.13 and 1 with a solution of a salt of a multi-valent metal-ion of Group 3a and converting the thus treated zeolite by calcination at a temperature in the range of from 350 to 850°C into a product having a unit cell size between 24.21 and 24.40 Å."

The dependent Claims 2 and 3 relate to preferred embodiments of the process according to Claim 1.

The Appellant argued that the process of Claim 1 was novel over the process disclosed in D3 since the latter did not involve the treatment of the US-Y zeolite with a salt of a multi-valent metal ion of Group 3a. As regards D2 the Appellant contended that the stabilized faujasite zeolite of example 1, which included MgO in its framework, was not considered by the Appellant's experts as being an ultra-stable Y zeolite since after calcination it could not exhibit the reduced unit cell size characterising the US-Y zeolite in view of the relatively high amount of MgO present in the framework (5.85%wt).

V. The Appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of Claims 1 to 3 as submitted in the oral proceedings.

Reasons for the Decision

1. The appeal is admissible.

2. There are no formal objections under Article 123(2) EPC to the amended set of claims submitted at the oral proceedings. The subject-matter of Claim 1 is supported by the process Claims 1, 6 and 7 as originally filed and by the following passages of the original description: page 3, lines 12 to 14 as regards the metal ions of Group 3a, and page 2, lines 9 to 13 and 25 to 26 for the use of an ultra-stable Y zeolite (US-Y) as starting material. In connection with the sodium oxide content of the US-Y, i.e. "at least 2.2%wt", the Board observes that a sodium oxide content "between 2.2 and 13.5%wt, in

particular between 2.5 and 13%wt" is disclosed at page 2, lines 25 to 30. The Appellant pointed out at the oral proceedings that the term "between" used in this context was intended to include also the lower limits 2.2 or 2.5%wt of these ranges. Taking into account that the starting material of the examples is a US-Y containing 2.5%wt of sodium oxide, the Board accepts that the terms "between 2.2%wt and ..." or "between 2.5%wt and ..." are to be interpreted as including the limit values. Therefore the mention of a sodium oxide content of "at least 2.2%wt" in the amended Claim 1 does not contravene Article 123(2) EPC. The dependent Claims 2 and 3 find a support in Claims 4 and 5 as originally filed and at page 4, lines 15 to 19, of the original application.

3. The process according to Claim 1 is novel with respect to the prior art cited in the search report.

D1 discloses the treatment of a Y-zeolite with a solution of an aluminium salt, i.e. a salt of a metal-ion of Group 3a, and the subsequent calcination of the treated zeolite at a temperature of 550°C or 815°C. However the starting material is not an ultra-stable Y zeolite containing at least 2.2%wt of sodium oxide.

In the examples 11 and 12 of D2 the starting faujasite zeolite was not treated with a salt of a metal-ion of Group 3a but with an ammoniacal palladium chloride complex solution or with a $MgCl_2$ solution before calcination at a temperature of 1000°F or 850°F respectively. Moreover, the Board notes that although the starting material of example 12 is a stabilised synthetic faujasite zeolite having a SiO_2/Al_2O_3 molar ratio of about 6.2 (cf. the composition of the stabilised zeolite reported in example 1), the Appellant's experts consider this stabilised zeolite as not being a US-Y taking into account that after calcination it cannot exhibit the typical decreased unit cell size of a US-Y because of the high content of MgO in its framework (5.85%wt). In the absence of evidence to the contrary the Board can follow these arguments.

The process of D3 involves the treatment of the Z-14 US zeolite with a solution containing a mixture of soluble nickel and magnesium salts. The treatment with a solution of a salt of a multi-valent metal-ion of Group 3a is not disclosed.

D4 relates to the thermal stabilisation of faujasite-type zeolites having a silica to alumina ratio of at least 3.2 by ion-exchange and calcination (cf. page 1, lines 1 to 4 and 26 to 37, page 2, lines 1 to 10). In example 9, which was discussed at the oral proceedings, the stabilised faujasite-type zeolite is ion-exchanged with a solution containing an aluminium salt (cf. table E, sample 4). The zeolite is then dried and calcined at 815°C. As the stabilised zeolite used as starting material contains only 0.14%wt of sodium oxide, the claimed process differs from that of D4 at least by the higher sodium content of the starting material.

D5, which was cited by the Examining Division to illustrate the composition of the Y-zeolite, does not disclose the process steps recited in Claim 1.

The remaining document GB-A-2 176 128 cited in the search report describes a process including the treatment of a stabilised Y zeolite with a solution of lanthanum nitrate and calcination at 500°C (cf. example 3, catalyst B). However the stabilised zeolite has a sodium content lower than 0.15%wt, i.e. lower than the lower limit of 2.2%wt stated in Claim 1, and this document does not disclose the use of a salt of a multivalent metal-ion of Group 3a instead of the lanthanum nitrate.

4. Only novelty was at issue in this appeal. The question whether or not a process as defined in Claim 1 involves an inventive step has not been examined by the Examining Division. In particular, it is still not clear what technical problem has been solved by the process of Claim 1 with respect to the closest prior art. In these circumstances, the Board finds it appropriate, in accordance with Article 111(1) EPC, to remit the case to the Examining Division for further prosecution on the basis of the process claims, in order to avoid loss of one instance.

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.

The Registrar: The Chairman:

P. Martorana P.A.M. Lançon