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DECISION
of 8 February 2006

Case Number: T 0533/03 - 3.3.07

Application Number: 95921830.6

Publication Number: 0762935

IPC: B01J 37/02

Language of the proceedings: EN

Title of invention:
FCC catalyst comprising coated zeolite particles

Patentee:
Akzo Nobel N.V.

Opponent:
W.R. Grace & Co.-Conn.

Headword:
-

Relevant legal provisions:
EPC Art. 54, 56, 83

Keyword:
"Novelty - yes"
"Inventive step - yes"
"Sufficiency of disclosure - yes"

Decisions cited:
T 0219/83, T 0131/03

Catchword:
-

Case Number: T 0533/03 - 3.3.07

DECISION
of the Technical Board of Appeal 3.3.07
of 8 February 2006

Appellant: (Opponent)	W.R. Grace & Co.-Conn. Grace Plaza, 1114 Avenue of the Americas New York, N.Y. 10036 (US)
Representative:	van Heesch, Helmut Werner Uexküll & Stolberg Patentanwälte Beselerstrasse 4 D-22607 Hamburg (DE)
Respondent: (Patent Proprietor)	Akzo Nobel N.V. Velperweg 76 NL-6824 BM Arnhem (NL)
Representative:	Rasser, Jacobus Cornelis Howrey LLP Rembrandt Tower Amstelplein 1, 31st Floor NL-1096 HA Amsterdam (NL)
Decision under appeal:	**Decision of the Opposition Division of the European Patent Office posted 24 March 2003 rejecting the opposition filed against European patent No. 0762935 pursuant to Article 102(2) EPC.**

Composition of the Board:

Chairman:	S. Perryman
Members:	B. Struif
	G. Santavicca

Summary of Facts and Submissions

I. The mention of the grant of European patent No. 0 762 935 in respect of European patent application No. 95 921 830.6, originating from international application PCT/EP95/02131 having a filing date of 2 June 1995 and claiming priority of 3 June 1994 (EP 94201592) was published on 15 April 1998. The granted patent comprised six claims. Independent claims 1 and 6 read as follows:

"1. FCC catalyst comprising coated zeolite particles and a matrix material wherein at least 50% of the outer surface of the zeolite particles is coated with a layer of pre-formed inorganic oxide, which layer has a thickness in the range of 10 nm to 5 µm, wherein the ratio between the particle size of the oxide and the mean particle size of the zeolite particles is in the range of 0.001:1 to 0.5:1."

"6. A process for fluidized catalytic cracking of hydrocarbon feeds in which a hydrocarbon feed is contacted under cracking conditions with a catalyst according to any one of claims 1 to 5."

II. On 15 January 1999, a notice of opposition was filed against the granted patent, in which revocation of the patent in its entirety was requested on the grounds of lack of novelty, lack of an inventive step and insufficient disclosure (Article 100, paragraphs (a) and (b), EPC). The opposition was *inter alia* supported by the following document:

D1: US-A-4 308 129

III. In a decision posted on 24 March 2003, the opposition was rejected.

According to the reasons of that decision:

(a) As regards sufficiency of disclosure, coating by hypercoagulation was generally disclosed in the patent in suit and a coating procedure was illustrated by examples. Hence, inorganic oxides other than the exemplified oxide materials could be used as well. The opponent had not shown that the invention could not be extended to other inorganic oxides. Thus, the description provided sufficient general teaching, how to coat the zeolite particles with pre-formed oxide particles and Article 83 EPC was met.

(b) As regards novelty, D1 disclosed a process for the preparation of a composite particle product (catalyst M), in which a zeolite was first combined with silica-alumina hydrogel and the resulting washed and dried product was combined with a slurry of alumina, colloid milled and then washed and dried. However, the alumina used had not been peptized before contacting it with the slurry of zeolite. The opponent had not shown that when such a catalyst was prepared the skilled person would inevitably arrive at a catalyst falling within the terms of claim 1, even if it was assumed that the zeolite particles were coated with the oxide particles. In particular, it had not been shown that the oxide layer thickness was in a range as specified in the claim, that at least 50% of the surface of zeolite was coated and that the ratio of zeolite to oxide particle sizes was within the claimed range and that these three structural features were met in combination.

As to the source of public prior use, the evidence in support thereof lay within the power and the knowledge of the opponent, who had to prove his case up to the hilt. However, the documents submitted in respect of prior use did not establish "a chain of commercial transactions" as required by case law.

Hence, the claimed subject-matter was novel over D1 and the alleged public prior use.

(c) As regards inventive step, the problem addressed in the patent in suit was to provide coated zeolite particles suitable for fluidized beds which were protected against abrasion and extremes of pH and deposits of heavy metals. D1 disclosed a composition comprising discrete particles of zeolite and discrete particles of an oxide in a matrix. When preparing catalyst M of D1, no details of the starting particle sizes or of the condition of colloiding were given nor was it stated that particles of alumina smaller than those of zeolite should be used. The Japanese document cited in the patent in suit (now D15b) taught the in-situ deposition of oxides on zeolite particles by precipitation from aqueous solutions. Since the oxide coating on the claimed catalyst had the purpose to protect the zeolite against physical and chemical degradation similar to that mentioned in the Japanese document it was a more appropriate starting point than D1 for evaluating inventive step. Nevertheless, no calcination was mentioned in D15b. Hence, the claimed solution of using preformed particles to provide a more adherent coating was not made obvious by the cited prior art.

As regards the objection of the opponent that not all of the oxides formed a coating by hypercoagulation and that a coating of only 50% of the outer surface did not provide the advantages as sought, no convincing evidence had been provided. Therefore, the claimed subject-matter was not obvious over the whole breadth of claim 1, and thus involved an inventive step.

IV. On 13 May 2003, the opponent (appellant) filed a notice of appeal against the above decision, the prescribed fee being paid on the same day. With the statement setting out the grounds of appeal, filed on 4 August 2003, the appellant submitted the following documents:

D8: Experimental report 1 relating to the preparation of catalyst M, Example 9 of US-A-4 308 129 (D1)

D9: US-A-4 086 187

D10: Experimental report 2 relating to the preparation of catalyst according to Example 4 of US-A-4 086 187 (D9)

D11: US-A-4 206 085

D12: Experimental report 3 relating to the preparation of catalyst J according to Example 7 of US-A-4 206 085 (D11)

D13: Experimental report 4 relating to the preparation of catalysts according to Examples 1 and 3 of the patent in suit

V. On 22 June 2004, the respondent submitted an amended set of claims 1 to 4 as auxiliary request.

VI. In a communication of 11 November 2005 the board addressed the points to be discussed during the oral proceedings.

VII. By letter of 3 February 2006, the appellant submitted the following document:

D15b: verified English translation of JP-A-S58-112 051

VIII. Oral proceedings were held on 8 February 2006. At the oral proceedings the respondent withdrew the auxiliary request then on file and submitted a set of amended claims 1 to 5 as the sole auxiliary request. Also, the original certified translation of document D15b was submitted.

IX. The appellant argued in substance as follows:

(a) As regards the terms used in the definition of granted claim 1, they mainly referred to the starting materials and to the process condition "pre-formed" for the preparation of the FCC catalyst. The term "preformed" in the claimed context did not necessarily mean that the oxide was formed separately at an earlier stage and did not as process feature contribute to the final definition of the catalyst. Also the other features of the starting materials could not be detected in the final catalyst product and thus were not significant for the definition of a product. Furthermore, the coating material could be the same as the matrix material, such as alumina, which could not be distinguished in the final product. Thus, it was not possible to ascertain whether a catalyst was within the scope of the claim.

(b) As regards lack of sufficiency of disclosure, the minimum degree of coating of the outer surface of the zeolite (50%) could only be determined at single particles, since the electron-microscopic detection method by TEM should be applied to several samples to provide a statistically significant result which method was very expensive. However, the detection on single particles did not allow the conclusion whether that feature was fulfilled by all particles of the catalyst. Similar arguments applied to the thickness of the layer. Thus, it was impossible to determine any parameters of the catalyst with the required degree of accuracy.

(c) As to novelty, since the terms used in claim 1 did not allow one to distinguish the claimed subject-matter from D1, it was necessary to rely on experimental report 1 (D8), describing the reproduction of catalyst M according to example 9 of D1. Example 9 disclosed co-milling a zeolite and a boehmite alumina prior to adding the mixture to a co-gel. Commercial alumina particles stick together in the form of agglomerates having a relative low density. By milling, the agglomerated alumina particles more easily broke up to smaller particles than the zeolite particles which were already present as crystallites. Thus, only the size of the alumina would be reduced. After milling, a sufficient amount of particles was present so that heterocoagulation inevitably took place to form a coating of the pre-formed inorganic oxide particles on the surface of the zeolite. In fact, the test report showed that all the features of claim 1 were met, when reproducing catalyst M as described in example 9 of D1. On the basis of that proof, the patentee could not simply claim the benefit of the doubt that the product obtained according to the teaching of document D1 did not exhibit the claimed parameters (see decision T 0131/03 dated 22 December 2004, not published in OJ EPO).

(d) Concerning the further documents cited, the wording of claim 1 did not distinguish the claimed subject-matter from D15b either. Furthermore, test reports D10 and D12 showed that the reworking of examples of D9 and D11, respectively, resulted in catalyst products which exhibited all the claimed features.

(e) As regards inventive step, D15b was considered to be the closest state of the art. D15b addressed the same technical effects as those mentioned in the patent in suit. The claimed subject-matter might differ from D15b only by the process feature "pre-formed". However, that difference could not distinguish the final products from one another. In that respect the starting alumina according to the patent in suit could exist in different forms dependent on the calcining conditions. According to D15b a calcination above 500°C should be avoided. The product according to the patent in suit also could be uncalcined as defined in claim 4. In the patent in suit no comparison in catalytic performance was made with respect to D15b but only a comparison with coated and uncoated zeolite particles. In the patent in suit the resistance to nickel and vanadium poisoning did not show any advantageous effects compared to results shown in D15b. Thus, the objective problem was to provide an alternative catalyst over that of D15b. A solution of that problem was made obvious by D15b. Thus, the claimed subject-matter lacked an inventive step.

X. The arguments of the respondent can be summarized as follows:

(a) In claim 1 the pre-formed inorganic oxide referred to particles separately formed from the zeolite which then were coated as a layer on the zeolite. The particles sizes referred to the starting materials of the zeolite and the inorganic oxide. The particle size of the oxide related to an average value as specified in the patent in suit. The mean particle size of the zeolite might be based on the weight or the volume. It was common to use weight or volume means interchangeably because the density was the intrinsic density of the particles.

(b) As regards sufficiency of disclosure, the arguments of the appellant were not convincing. The thickness of the coating on the zeolite particle and its percentage of the covering could be measured as specified in the patent in suit and as confirmed by the appellant's test reports. If some particles were investigated by suitable electron microscopic method, this would allow a conclusion whether or not the claimed features were fulfilled over the entire population of the particles. The cost was no reason that the determination was impossible. No experiments had been made which showed that the skilled could not reproduce the examples of the patent in suit nor the catalysts within the whole ambit of the claims.

(c) Regarding novelty, the purpose of D1 was to provide discrete alumina and discrete zeolite particles dispersed within a matrix material, which was different from providing a coating of inorganic oxide particles onto the zeolite according to the patent in suit. According to D1, the preparation of catalyst M required that a ball milled alumina was added to a slurry of a ball milled zeolite and that the resulting slurry was colloid milled. In claim 1 as granted the particle size of the alumina was always at least 50% less than that of the zeolite particles. The experiments carried out by the appellant deviated from those of D1. In the appellant's experiments the size of the alumina particles was higher than that of the zeolite particles. Furthermore, according to the appellant's experiments the significant reduction of the particle size after colloid milling implied a severe milling, which was not in line with colloid milling of D1.

(d) Furthermore, the appellant's choice of the alumina and that of the zeolite in the experiments was not appropriate. Whilst in D1 a silica alumina amorphous gel was mentioned, in the reworked example a silica gel was used. Furthermore, the silica alumina filter cake for preparing catalyst M contained more water than the filter cake according to the test report D1. The sulfate present in the impure filter cake would preferentially go to the surface of the zeolite to increase its charge so that heterocoagulation was stimulated. Moreover, the test report used deionized water and a two hour ball milling, which conditions were not mentioned in D1. Furthermore, it had not been shown that the claimed ratio of particles size of alumina to zeolite was met. The TEM graphs of the spray dried catalyst did not show discrete particles of ultra stable Y-type zeolite and discrete particles of alumina, which particles should be dispersed in a porous oxide matrix as required by D1.

The reworking of D9 was not proper either, since the experimental report used Ludox AS-40 instead of a polysilicalite. The appellant's reworking of example 4 of D9 also used a different zeolite. Furthermore, rare earth salts different from those of D9 were used. Moreover, the time of contacting between the zeolite and the peptized alumina was not disclosed in D9. Finally, the pH of the diluted silica sol was different from the stabilized ammonium polysilicate of D9.

The disclosure of D11 was similar to that of D9. The reworking of example 7 of D11 was deficient for reasons similar to those mentioned above for D9, except for the polysilicate. In particular, the zeolite, the salts for ion exchange and the contact time used in the experimental report were different from those specified in D11.

(e) As to inventive step, in D15b an amorphous form of alumina was coated onto the zeolite which was different from pre-formed particles. The pre-formed particles allowed to choose the crystallinity of the coating. Furthermore, according to D15b no individual particles were formed on the zeolite nor was their size measured. The quality of the coating obtained by the process on D15b was not sufficient, since it did not firmly adhere on the zeolite particles. Furthermore, the zeolite coated according to the patent in suit was not exposed to pH changes, whilst in D15b during the in-situ formation of the coating high pH values were used. The claimed subject-matter allowed a proper control of the alumina formed. Furthermore, when using the catalyst a reduced coke formation was observed as shown by Table 4 of the patent in suit. Hence, the claimed subject-matter was not made obvious by the cited prior art and involved an inventive step.

XI. The appellant requested that the decision under appeal be set aside and that the European patent be revoked.

XII. The respondent requested as main request that the appeal be dismissed and as auxiliary request that the decision under appeal be set aside and that the patent be maintained, on the basis of the auxiliary request submitted on 8 February 2006 during the oral proceedings.

Reasons for the Decision

1. The appeal is admissible.

Interpretation of the claimed subject-matter

2. According to the patent in suit, the inorganic oxide material to be coated on the zeolite has been formed before it is contacted with the zeolite particles (page 4, lines 25, 26 and 33). The particle size of the zeolite is a mean particle size and refers to the particle size at which 50 wt.% of the particles has a greater particle size while the other 50 wt.% of the particles has a smaller particle size (page 4, lines 40 to 43). The particle size of the oxide (to be) coated onto the zeolite particles is defined as the value of Z_{avg} determined by quasi-elastic light scattering (page 4, lines 49 to 51 and page 7, lines 2 to 11).

2.1 As regards the coated zeolite particles, at least 50% of the outer surface of the zeolite is coated with a layer of the preformed inorganic oxide (particles), which layer has a thickness in the range of 10 nm to 5 µm (page 3, lines 19 to 22). That coating of the zeolite particles can be analysed with electron-microscopic techniques, such as TEM and STEM/EDX, before the particles are incorporated into the FCC catalyst or it can be visualized by scanning electron microscopy (SEM) (page 4, lines 1 to 3). The features of the coating can be determined before the coated zeolite particles are incorporated into the matrix material (see example 4 of the patent in suit, page 8, lines 57) and can also be detected in the final FCC catalyst (page 9, lines 23 to 25).

2.2 From the above it follows that the particle sizes of the zeolite and the inorganic oxide according to claim 1 refer to specific average particle sizes of the starting materials for preparing the catalyst and also relate to the coated zeolite product formed thereof. Hence, the ratio between the particle size of the oxide and the mean particle size of the zeolite according to claim 1 has a concrete and specific meaning. Furthermore, the term "pre-formed inorganic oxide" refers to inorganic oxide particles defined by its particle size Z_{avg} before they are contacted with the zeolite particles. Hence, the claimed FCC catalyst is defined in terms of structural features of their starting materials (zeolite and oxide particles), which features are detectable also in the coated zeolite particles. The coating thickness of the oxide particles and the coverage of the outer surface of the zeolite particles can be detected in the final structure of the FCC catalyst product.

Sufficiency of disclosure (Article 100(b) EPC)

3. According to granted claim 1 "at least 50% of the outer surface of the zeolite particles is coated with a layer of preformed inorganic oxide which layer have a thickness of 10 nm to 5 µm". The essence of the appellant's argument regarding lack of disclosure is that the claimed catalyst could not be reproduced, because the coverage on the outer surface of the zeolite particles and the thickness of the layer could not be determined with the required degree of accuracy.

3.1 The question arises whether or not the patent in suit provides sufficient information which enables the skilled person when taking into account common general knowledge to reproduce a FCC catalyst comprising a coated zeolite particles having the features as defined in claim 1.

3.1.1 According to the patent in suit the process of coating the zeolite particles with pre-formed oxide particles is based on heterocoagulation. That is, use is made of the attracting force between the coating particles and the zeolite particles to be coated, which attracting force is caused by the charge difference resulting from the differences in isoelectric point of the various components. This process results in a high quality coating (page 3, lines 37 to 41). TEM and STEM/EDX can be used to study whether or not an FCC catalyst comprises zeolite particles with a coating as defined above. The thickness of said coating can also be determined with these techniques (page 3, lines 55 to 56).

3.1.2 The presence of a complete coating cannot only be ascertained by these electron microscopical techniques, but also via an electrokinetic route. The isoelectric point (IEP) of the uncoated zeolite particles and that of the coating oxide can both be determined by electrokinetic techniques such as acoustophoresis. The determination of the surface coverage of the zeolite particles by alumina and titania is illustrated (page 4, lines 3 to 12).

3.1.3 Furthermore, according to Example 1, the method of coating Y zeolite particles with 5 wt% alumina particles is described. The zeolite particles are specified by their content of Na_2O and R_2O_3, the specific surface and the mean particle size. The alumina particles are commercially available and specified by their content of Al_2O_3, Na_2O, and SO_4, their specific surface, their particle size Z_{avg} and their polydispersity (see Tables 1 and 2). The preparation method is described in detail, including the heterocoagulation, drying and the calcination of the coated particles.

3.1.4 Example 3 of the patent in suit discloses a FCC catalyst comprising alumina coated zeolite according to Example 1. The catalyst obtained comprises 26.25 wt.% of coated zeolite, and the balance, a matrix material comprising clay and silica. The catalyst particles are obtained by spray-drying. After removing particles having a diameter of less than 20 µm, the remaining catalyst particles are washed to remove salts (page 8, lines 46 to 52).

3.1.5 Furthermore, in Example 4 the alumina coated zeolite particles prepared in Example 1 are studied with TEM, STEM/EDX, and SEM. The catalysts prepared according to Example 3 are studied with TEM and STEM/EDX.

3.2 Consequently, the patent in suit exemplifies in detail how to coat the zeolite particles, as well as how the thickness and degree of coverage can be determined by suitable measuring methods. Thus, the patent in suit provides a sufficient disclosure how the claimed FCC catalyst can be reproduced.

3.3 The appellant however argued that the STEM/EDX tests were very expensive and could only be made on single particles, which did not allow a conclusion whether or not the 50% coverage of the zeolite particle was met for the entire population of the particles.

3.3.1 According to the patent in suit, it is preferred that the outer surface of the zeolite is coated to the fullest extent, up to 100% (compare also page 3, lines 23 and 24). Indeed according to the examples, the outer surface of all the zeolite particles according to the exemplified product is coated and no uncoated zeolite edges are visible (page 9, lines 4 to 8). Hence, the 50% coverage of the zeolite particles should be seen as a minimum requirement that at least half of the surface of the zeolite should be covered and that this coverage applies not only to a single particle but to the whole population of the particles. Since a complete coverage of the zeolite is aimed at, the specification focusses on that goal. Normally, no problems with that feature should arise.

3.3.2 In fact, the appellant has reproduced alumina coated zeolite particles according to Example 1 and FCC catalysts according to Example 3 of the patent in suit in experimental report D13. According to SEM pictures, the reproduced particles are fully coated. Furthermore, TEM analysis confirms that more than 50% of the surface of the zeolite particles are coated and that the coating thickness is 10 to 45 nm (see D13, test results, table on page 5 and corresponding graphs as indicated). Consequently, the appellant's own experimental report shows that the skilled person was able to reproduce the claimed catalyst and to determine whether or not the claimed parameters are met.

3.3.3 Although the determination of the above features, in particular of the thickness of the layer and its coverage on the zeolite is expensive, the costs are no reasons that the skilled person is unable to determine the required parameter, even if the determination had to be carried out on more than one particle. On the other hand, the appellant has not carried out any experiments which show that coated zeolite particles having the claimed surface coverage and thickness of the layer could not be reproduced by the skilled person without undue burden. The onus of proof in that respect lies however with the appellant (opponent), which he has failed to discharge (T 219/83, OJ EPO 1986, 211).

3.3.4 From the above it follows that a case of insufficient disclosure (Article 100(b) EPC) has not been established.

Novelty

4. D1 discloses a process for the catalytic cracking of a hydrocarbon feedstock which comprises contacting said feedstock under catalytic cracking conditions with a cracking catalyst composition comprising discrete particles of ultra-stable Y zeolite and discrete particles of alumina, which particles are dispersed in a porous oxide matrix to produce a catalyst containing 5-40 wt.% ultra-stable Y zeolite, 5-40 wt.% alumina and 40-90 wt.% of porous oxide matrix (claim 1).

4.1 Example 9 of D1 *inter alia* describes the preparation of catalyst M, which contains discrete particles of ultra-stable Y zeolite and discrete particles of alumina which are dispersed in an amorphous silica-alumina gel matrix to give a catalyst containing 20 wt.% ultra-stable Y zeolite, 20 wt.% alumina and 60 wt.% amorphous silica-alumina gel (column 13, lines 17 to 22).

4.1.1 Catalyst M is prepared by the admixture of 30 lbs. of water with 93 lbs. (equivalent to 9.0 lbs. dry catalytic solids) of the impure silica-alumina filter cake as described in the preparation of catalyst I. In a separate vessel, 20 lbs. of water are admixed with 3.74 lbs. (equivalent to 3.0 lbs. dry basis) of the ball milled and uncalcined ultra-stable Y zeolite designated as LZ-Y82. Thereafter, 4.08 lbs. (equivalent to 3.0 lbs. dry basis) of the ball milled and uncalcined alumina are added to the slurry containing the ultra-stable Y zeolite designated as LZ-Y82. The resultant slurry is colloid milled and combined with the impure silica-alumina slurry to form a composite which is colloid milled, spray dried and washed free of extraneous soluble salts with ammonium sulfate solution at a pH of 7.8 to 8.3 (see preparation of catalyst I, column 12). It is calcined at about 1000°F for six hours (column 13, lines 17 to 51). The alumina is commercial grade Boehmite type alumina stabilized with 2.5 wt.% SiO_2 and having a surface area of 523 m^2/g to which nitric acid is added under stirring and thereafter diluted with water to provide a peptized alumina slurry (see column 12, lines 41 to 49 and 60 to 61).

4.1.2 It follows from the above that catalyst M is prepared by using a peptized alumina at a low pH value which is then brought together with a zeolite. The co-milling of the zeolite with peptized alumina is effected prior to the addition of the co-gel matrix. However, D1 does not aim at obtaining a layer of inorganic oxyde on the zeolite. In fact, neither the general disclosure of D1 nor the specific process conditions for the preparation of catalyst M of D1 address the particle sizes of the inorganic oxide and zeolite, a surface layer, the thickness of the layer and the 50% coverage of the surface of the zeolite particles. Thus, D1 does not directly and unambiguously disclose the claimed features.

4.1.3 The appellant argued that the reproduction of catalyst M as described in example 9 of D1 inevitably results in a catalyst having all the claimed features as shown by experimental report D8.

4.1.4 According to experimental report D8, the alumina has a SiO_2 content of 2.73% and a surface area of 474 m^2/g compared to 2.5 wt% and 523 m^2/g, respectively, as used in D1. The ultra stable zeolite in D8 is produced by appellant. The LZ-Y 82 zeolite product of D1 which is available on the market has not been used. Furthermore, in experimental report, D8 27261g of the cogel filter cake (TV=85%) is slurried with 28577g deionized water so that the cogel slurry has a solid content of about 50%. According to catalyst M of D1, 30 lbs water is admixed with 93 lbs (equivalent to 9.0 lbs dry catalytic solid) so that the solid content of the slurry is less than 10% which is significantly lower than that used in D8. As regards the milling steps, D1 does not disclose like D8, that the dry alumina and zeolite are ball milled separately for two hour at room temperature. An excessive ball milling reduces the alumina size as shown by the size analysis of D8. The mean particle size d(0.5) of the alumina after a first milling step is about 9.1 µm and after a second milling step about 3.3 µm. The mean particle size d(0.5) of the zeolite is about 3.9 µm. Thus, there are differences between the disclosure in D1 and the reproduction according to D8.

4.1.5 Furthermore, according to granted Claim 1, the ratio between the particle size of the oxide (Z_{avg} see Reasons, point 2) and the mean particle size of the zeolite is at most 0.5:1. If the average value (d(0.5)) of the alumina particles after the second milling step is taken as Z_{avg} value, the ratio between the particle size of the oxide and the mean particle size of the zeolite is at most 3.3 : 3.9 = 0.85 outside the claimed range. That 4.52 wt.% of alumina particles after the second milling step have a particle size of less than 1 µm would meet the requirement of claim 1, as argued by the appellant, is not in line with the definition according to claim 1 as interpreted in the context of the description (Reasons, point 2).

4.1.6 In any case, D1 aims at a catalyst composition comprising **discrete** particles of ultra-stable zeolite and **discrete** particles of alumina, which particles are dispersed in a porous oxide matrix (claim 1, emphasis by the board). Thus, the purpose of D1 is the provision of discrete particles within a matrix material but not a coating of preformed oxide particles on the zeolite particles. Thus, the experiments should aim at producing such discrete particles dispersed in a matrix. If the test results showed coated zeolite particles rather than the expected discrete particles dispersed in the matrix, these results would rather confirm that the teaching of D1 has not been followed.

4.1.7 From the above it follows that process conditions, starting materials and particle sizes of experimental report D8 cannot directly and unambiguously be derived from D1. Thus, the test report D8 cannot be a proper reproduction of D1 nor can it show that the catalyst obtained is the inevitable result of a reproduction of catalyst M as described in example 9 of D1. In addition, in D8 it is not shown that the critical ratio between the particle size of the oxide (Z_{avg}) and the mean particle size of the zeolite is met.

4.2 The appellant referred to decision T 0131/03 (*supra*) concerning an opposition case in which the issue in dispute was the geometry of the pigment particles which was defined in terms of the largest and smallest values of the major and minor axes of the particles, respectively (Reasons, point 2.3). The opponent had filed experimental results based on a prior document which showed that this ratio was inherently disclosed in that prior art document. Since the proprietor had chosen to define the invention by way of unusual parameters he could not simply claim the benefit of the doubt. The burden of proving that the product obtained from the teaching of the prior art document did not exhibit the claimed parameters had switched to his side (Reasons, point 2.7).

The clear teaching in D1 that the catalyst composition comprises **discrete** particles of zeolite and **discrete** particles of alumina in the matrix (claim 1, emphasis by the board) is in contrast to the provision of a coating of alumina on the zeolite particles according to the patent in suit. Thus, the skilled person has no reason to infer that the experiments of D1 would result in a coating. Furthermore, the experimental report D8 does not show that the ratio between the particle size of the oxide (z_{avg}) and the mean particle size of the zeolite is fulfilled. Hence, the present case is not comparable with that of T 0131/03 and there is no need for the proprietor to prove anything to the contrary.

4.3 The appellant had furthermore relied on Example 4 of D9 and catalyst J according to Example 7 of D11 on which experimental reports D10 and D12, respectively, were based.

4.3.1 According to Example 4 of D9, a catalyst is formed having the following composition:
19% by weight of a zeolite exchanged with rare earth sulfate solution is combined with 18% by weight of pseudoboehmite and with 16% by weight of SiO_2 in the

form of ammonia polysilicate containing 5.5% by weight of silica expressed as SiO_2, the ammonium silicate solution having a specific gravity of 1.045 grams per cubic centimeter. 16% of the ammonium polysilicate and 42% of ball clay, all percentages by weight on a volatile-free basis are added.

405 grams, on a volatile-free basis, of the above pseudoboehmite are blended with 3.568 liters of water containing 38 ml of formic acid for about 30 minutes under mild agitation. To this slurry, 427 grams, on a volatile-free basis, of a zeolite product are blended with the above peptized pseudoboehmite under mild agitation. To the resultant slurry is added a solution containing 360 grams of ammonium polysilicate calculated on a volatile-free basis, and the mixture is stirred for about 10 minutes under vigorous agitation. Following this agitation, 1,058 grams on a volatile-free basis of ball clay are added and mixed about 10 minutes with vigorous agitation; and the slurry is passed promptly to the spray drier. The resultant catalyst produced by spray drying has an average particle size of 70 micron diameter (column 12, lines 23 to 44).

4.3.2 The zeolite employed in example 4 of D9 is a faujasite-type zeolite produced by silication of clays having the components: Na_2O - 4.5% by weight, ReO - 12.0% by weight, Al_2O_3 - 26% by weight, remainder silica (SiO_2) (Example 1 and column 5, lines 63 to 67). According to experimental report D10 a socalled Y-zeolite is used that contains: Na_2O - 1.55% by weight, rare earth oxide - 13.0% by weight, Al_2O_3 - 20% by weight, remainder silica (SiO_2). Thus, the zeolite material used in experimental report D10 is different from the exemplified zeolite of D9. Moreover, D9 uses a stabilized ammonium polysilicate solution which contains NH_4^+ cations and polysilicate anions of low molecular weight with a major portion of SiO_2 in the form of said anion to distinguish it from a colloidal sol (column 9, lines 16 to 22). In contrast thereto, experimental report D10 uses diluted Ludox AS 40 which is a colloidal silica sol (D10, page 2, points 2. and 3.) and not a stabilized ammonium silicalite solution. According to D10, the mean particle size of the zeolite is about 3.8 µm and the mean particle size of alumina before mixing is about 5.7 µm (annex 2B) and of the mixed slurry is about 3.2 µm (annex 2C). In any case the ratio between the particle size of the alumina (Z_{avg}) and the mean particle size of the zeolite is at most 3.2 : 3.8 = 0.84 outside the claimed range.

4.4 For the preparation of catalyst J of example 7 of D11 30 wt% Type A and 20 wt% Type B-3 aluminas are prepared by dispersing the aluminas in deionized water. Type A alumina is unmilled and acidified with formic acid. Sample B-3 of Type B alumina is an acidified portion of the alumina milled by passage through a colloid milled until the temperature started at ambient temperature rises to 15°F. Catalyst J is formulated from the above alumina types, employing 25 wt% ball clay as the clay and 25% zeolite (Table 6). The mixture of pseudoboehmite, clay and the zeolite are then spray dried.

4.4.1 The teaching of D11 is similar to that of D9 described above (Reasons, point 4.3 above) except for the omission of the polysilicalite. According to test report D12 an ultrastable Y zeolite having a Na_2O content of 0.55 wt.% is used, whilst D11 employs a classic faujasite zeolite having a Na_2O content of 2.20 wt.% (Example 3). Furthermore, in D12 chloride salts instead of sulphate salts are used to form exchanged zeolite. According to D12, the mean particle size of the zeolite is about 3.9 µm and the mean particle size of Type A alumina is about 5.5 µm (annex 3B) and the mean particle size of Type B3 alumina is about 2.0 µm (annex 3C) before mixing. In any case the mean particle size of the mixture of both alumina types which has not been measured is much higher than 2 µm. Thus, the ratio between the particle size of the alumina (Z_{avg}) and of the mean particle size of the zeolite is outside the claimed range, even if the percentage of alumina particles having a size of below 1 µm in both alumina type fractions is about 32 to 33 wt.% (D12, table page 5).

4.4.2 From the above it follows that the starting materials and process conditions used in experimental reports 10 and D12 cannot directly and unambiguously be derived from the disclosure of D9 and D11, respectively. Furthermore, in D10 no ammonium polysilicalite has been used. Thus, the test reports D10 and D12 cannot be appropriate to show that the catalyst obtained is the inevitable result of a reproduction of example 4 of D9 and catalyst J of D11, respectively. In addition, according to D10 and D12 the claimed ratio between the average particle size of the alumina (Z_{avg})and the mean particle size of the zeolite is not met.

4.5 D15b discloses a catalyst composition for fluid catalytic cracking of hydrocarbons having fine zeolite dispersed on a siliceous matrix, in which the zeolite is dispersed on the matrix in a form covered with a single metallic oxide (claim 1). The metallic oxide makes up from 3 to 20% by weight of the zeolite and the zeolite covered with a single metallic oxide makes up from 5 to 40% by weight of the composition (claims 2 and 3).

4.5.1 For the preparation of zeolite covered with alumina, aluminum sulfate is mixed with zeolite that has been processed in a colloid mill. The pH level of that mixture is from 3 to 4. Aqueous ammonia is added to the mixture under stirring; when the pH rises to about 9, aluminum hydroxide precipitates onto the surface of the zeolite and is cured for 2 hours at 80°C. Then, the zeolite covered with the aluminum hydroxide is separated from the fluid, washed with water and dried for about 12 hours at 120°C. If the covered zeolite dried in that way is calcined in air at a temperature above 500°C alumina is separated from the surface of the zeolite during the manufacture of the catalyst composition which leads to undesirable results (paragraph bridging pages 2 and 3).

4.5.2 Hence, in D15b no pre-formed inorganic oxide particles have been used and none of the other claimed features is specified therein. In particular, the ratio of the particle size and the thickness of the layer cannot directly and unambiguously be derived from the disclosure of D15b. Whilst the catalysts according to D15b can be calcined in air only up to 500°C without damaging the layer (see page 3, second complete paragraph), the calcination according to the patent in suit can be as high as 800°C (see page 5, line 26) so that the structure of the layers must be different. In addition, the appellant has not submitted any evidence which shows that when following the teaching of D15b a FCC catalyst as claimed is obtained as an inevitable result of the known process.

4.6 Consequently, the opponent has not shown that the claimed subject-matter lacks novelty over the cited prior art documents.

Inventive step

Problem and solution

5. The patent in suit concerns a FCC catalyst comprising coated zeolite particles. Such compositions are known from D15b, which is acknowledged in the patent in suit, by both parties and by the opposition division as the closest prior art document. The board has no reason to deviate from that starting point.

5.1 D15b concerns the protection of zeolite particles by coating them with an oxide before they are dispersed on the matrix of the catalyst. FCC catalysts are impaired when pores of the zeolite are sealed by the silica component of the matrix. When the sealing occurs, the function as catalyst is reduced markedly (D15b, page 2, first full paragraph). Hence, D15b aims at a FCC catalyst composition with superior cracking activity, gasoline selectivity' hydrothermal stability and resistance to metal poisoning (page 2, second full paragraph).

5.2 The quality of the in-situ formed coating on the zeolite particles by means of the process of D15b has been studied by electron-microscopic techniques and compared with those obtained by the patent in suit (examples 1 to 4). In example 1 a coating of zeolite particles with pre-formed alumina is prepared by using a peptised alumina slurry and a zeolite slurry. The mixture of the sluries has a final pH of 4.9. The properties of the zeolite and of the pre-formed alumina including the mean particle size and the Z_{avg} value respectively, are stated in Tables 1 and 2. In comparison, an alumina coating of zeolite particles is prepared according to D15b by using the zeolite employed in Example 1, aluminum sulfate and ammonia. The precipitation of the coating occurs at a pH of 9.3 (example 2). In Examples 1 and 2 the coated zeolite contains the same amount of alumina calculated on the weight of the zeolite (5 wt.-%).

The coated zeolite particles of Examples 1 and 2 are incorporated into a matrix comprising silica and clay to provide an FCC catalyst (Example 3). The catalysts particles are spray dried and washed to remove salts.

Thus, the coated zeolite particles according to Examples 1 and 2 and the FCC catalysts obtained according to Example 3 have identical compositions and their preparation differs essentially only in that a pre-formed alumina instead of an in-situ formation is used. The products obtained according to Examples 1 to 3 are then studied with TEM, STEM/EDX, and SEM (Example 4).

5.3 The results in Example 4 show that all zeolite particles coated according to Example 1 (invention) are separately covered with a thin dense alumina layer in which separate alumina particles can be distinguished and no uncoated zeolite-edges are visible (page 9, lines 2 to 4). Substantially all the non-zeolitic alumina present in the sample is in the form of a coating on the zeolite particles (page 9, lines 6 and 7). The coating of the zeolite particles has remained substantially intact during the preparation of the catalyst (page 9, lines 27 and 28).

5.4 In the zeolite particles prepared according to D15b (Example 2), the alumina layer is thicker, and less dense than the zeolite coated according to Example 1, and the coating adheres quite loosely to the zeolite particles (page 9, lines 9 to 11). The zeolite particles are partially coated with alumina and large parts of the zeolite surface are bare (page 9, lines 14 and 15). The FCC catalyst containing zeolite particles coated according to D15b (example 2) are not resistant to the catalyst preparation conditions, since the zeolite particles in that catalyst comprise hardly any coating, even where the zeolite is not in direct contact with the matrix in the finished catalyst particle (page 9, lines 30 to 33).

5.5 From the above results it follows that by the use of pre-formed particles compared to zeolite particles obtained by in-situ coating according to D15b structural differences in the FCC catalyst and an improved coating adherence are achieved. In particular, the coated layer is more resistant to severe conditions under which the FCC catalysts are prepared (see also page 2, lines 50 to 53). Furthermore, the coating density and the degree of coverage on the zeolite particles can be better controlled.

5.6 Moreover, the precipitation process in D15b takes place at high pH values, for example [pH of] about 9 (page 2, line 3 from bottom). The coating process according to the patent in suit is carried out at a pH below 7 (page 3, line 36). Since zeolites are susceptible to high pH values, the properties of the zeolite, particularly the crystallinity, may be adversely affected during the coating process of D15b (page 3, lines 3 to 6). Furthermore, the protons, or proton precursors such as ammonium ions, in the zeolite can be, and often are, exchanged during the coating process with the cations of which the oxides are to precipitate on the zeolite. This process is uncontrollable, and may lead to undesirable alteration of the zeolite properties (page 3, lines 6 to 10).

5.7 The respondent furthermore argued that the claimed FCC catalyst provided higher conversion, less hydrogen and less coke formation when subjected to deactivation in the presence of 1 000 ppm nickel and 5 000 ppm vanadium as shown by Table 4 of the patent in suit.

5.7.1 The comparison made in Table 4 is based on a catalyst A prepared according to Example 3 of the patent in suit comprising zeolite particles coated with 6% alumina based on the weight of the finished catalyst composition. Comparison catalyst B comprises uncoated zeolite, in which 6% of alumina is incorporated in the catalyst matrix to compensate for the absence of the coating alumina (Example 5, page 9, lines 42 to 48). In catalyst C calcined alumina instead of uncalcined alumina in catalyst B has been used (page 10, lines 56 to 58). Consequently, comparative catalysts B and C do not comprise zeolite particles coated according to D15b and thus cannot be used for a comparison whether or not an improvement over the closest state of the art has been achieved in the activity and selectivity of the catalyst.

5.7.2 Furthermore, a comparison of the data presented in the patent in suit (Table 4) with those indicated in D15b (Tables 1 to 4) is not possible, since the catalyst used in the patent in suit have a zeolite content of 28 wt.-% (page 10, table 3) whilst the catalysts used in D15b have a zeolite content of 3 wt-% (Table 1, page 5).

5.7.3 Thus, the catalyst performance indicated under point 5.7. to 5.7.2 above cannot be taken into consideration when formulating the problem underlying the invention (Case Law, *supra*, I.D.4.4).

5.8 From the above it follows that the patent in suit justifies the formulation of a problem which may be seen in providing FFC catalysts comprising zeolite particles with a coated layer of inorganic oxide, wherein the adherence of the coating on the zeolite particles is improved and the coating density and the degree of coverage on the zeolite particles withstands the manufacture, without disadvantageously affecting properties of the zeolite particles' in line with the patent in suit (page 3, lines 24 to 30).

5.9 In the light of Examples 1 to 4 of the patent in suit, the above-defined technical problem is effectively solved.

Obviousness

6. It remains to be decided whether the claimed subject-matter is obvious having regard to the documents on file.

6.1 Although D15b provides a coating of the metallic oxide on the zeolite particles by in-situ formation, it aims at preventing the blocking of the zeolite pores by a component of the matrix (page 2, complete first and second paragraph). In contrast thereto, the patent in suit aims at a better adherence and resistance of the coated layer on the zeolite particles. Thus, D15b does not give any incentive for a layer of pre-formed inorganic oxide particles and no hint to use a specific ratio between the particle size of the inorganic oxide (Zavg) and the mean article size of the zeolite particles to better control said layer properties. Hence, the claimed subject-matter is not made obvious by D15b alone.

6.2 D1 addresses the problem of cracking higher boiling point feedstocks with a catalyst comprising discrete particles of zeolite and alumina to produce high octane gasoline fractions, in which alumina is an active catalytic agent (column 3, lines 19 to 23). Quite to the contrary, the oxide in the claimed catalyst had a different purpose, namely to protect the zeolite against physical and chemical degradation. Furthermore, since D1 provided discrete particles of the zeolite and discrete particles of alumina dispersed in the porous oxide matrix, there is no incentive to modify the teaching of D15b in the direction as claimed.

6.3 D9 discloses a water slurry consisting essentially of a mixture of a zeolite of the faujasite type having a Na content, expressed as Na_2O_3, of less than about 5% by weight of the zeolite, a kaolin clay, alumina in the form of pseudoboehmite and ammonium polysilicate (claim 1). D9 aims at a catalyst having improved attrition resistance (column 5, lines 1 to 5) and thus has some similarity with the problem of the patent in suit. That problem is solved by properly selecting the ammonium silicate (column 5, lines 24 to 29) and by using pseudoboehmite which preferably has a high peptizability and it substantially free of sodium (column 8, lines 3 to 9). However, ammonium silicate does not play any role in the claimed catalyst. Furthermore, in D9 there is no hint to prepare any layer of pseudoboehmite particles onto the zeolite particles having the claimed thickness and surface coverage by using a specific ratio between the particle size of the oxide (Z_{avg}) and the mean particle size of the zeolite. Thus, the skilled person gets no incentive from D9 to modify the teaching of D15b in the direction as claimed.

6.4 D11 discloses a hydrocarbon conversion catalyst comprising a zeolite and a matrix, said matrix including alumina derived from pseudoboehmite, said pseudoboehmite composed of Type A and milled Type B aluminas, and SiO_2 derived from ammonium polysilicate or silica sol having particles of average particle size of less than 5 millimicrons (claim 1). Type A pseudoboemite is for example Catapol alumina (see column 1, lines 64 to 67). In Type B pseudoboehmite substantially all the particles (94%) have equivalent diameters less than 3 µm and 19% of all the particles are 0.2 µm or less (column 3, lines 23 and 27 to 30). According to D11, Type B alumina decreases the bulk density of the spray dried micro particles and provides an acceptable attrition index when combined with a Type A alumina without adversely affecting the catalytic activity (column 4, lines 23 to 28). If Type B alumina is used alone the attrition resistance of the catalyst is deteriorated (table 5, column 9, lines 63 to 68). In addition, the attrition index may be improved by the addition of ammonium polysilicate (table 6, sample G compared to sample J). Hence, the attrition resistance in D11 can be improved if coarser pseudoboehmite particles of the Type A and ammonium polysilicate are used as well.

However, the above teaching of D11 is different from the claimed subject-matter in which use is made of a specific ratio of oxide particle size (Z_{avg}) and the mean particle size of the zeolite in order to provide a coating of the oxide particles on the zeolite particles having a defined thickness and surface coverage. Thus, the skilled person gets no incentive from D11 to modify the teaching of D15b in the direction as claimed.

6.5 The other documents cited during the proceedings are even more remote than the documents already discussed, and no combination of one or more of these documents with D15b would lead the skilled person to arrive at the claimed subject-matter either.

6.6 Therefore, the claimed subject-matter of the granted patent involves an inventive step.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar	The Chairman
C. Eickhoff	S. Perryman