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**D E C I S I O N**  
**of 6 September 2002**

**Case Number:** T 0289/98 - 3.3.6

**Application Number:** 91104569.8

**Publication Number:** 0449144

**IPC:** C10G 45/12

**Language of the proceedings:** EN

**Title of invention:**

Catalyst composition for hydrotreating of hydrocarbons and hydrotreating process using the same

**Patentee:**

COSMO OIL COMPANY, LTD, et al

**Opponent:**

Akzo Nobel N.V.

**Headword:**

Hydrotreatment Catalyst/COSMO OIL

**Relevant legal provisions:**

EPC Art. 114(2), 123(2), 56

**Keyword:**

"Late-filed documents: not admitted"

"Added subject-matter (no): basis for amendment to be found in the examples"

"Inventive step (yes)"

**Decisions cited:**

T 0715/95, T 1002/92, T 0201/83

**Catchword:**

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Case Number: T 0289/98 - 3.3.6

**D E C I S I O N**  
of the Technical Board of Appeal 3.3.6  
of 6 September 2002

**Appellant:**  
(Opponent)

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**Representative:**

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**Respondents:**  
(Proprietors of the patent)

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and

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**Representative:**

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**Decision under appeal:**

Interlocutory decision of the Opposition Division  
of the European Patent Office posted 27 January  
1998 concerning maintenance of European patent  
No. 0 449 144 in amended form.

**Composition of the Board:**

**Chairman:** P. Krasa  
**Members:** L. Li Voti  
C. Holtz

## Summary of Facts and Submissions

- I. The present appeal is from the interlocutory decision of the Opposition Division concerning the maintenance in amended form of the European patent no. 0 449 144 relating to a catalyst composition for the hydrotreatment of hydrocarbons.

Claim 1 of the set of claims found to comply with the requirements of the EPC had the following wording:

"1. A catalyst composition for the hydrotreatment of hydrocarbon oils comprising at least one metal component having hydrogenating activity selected from each of the metals belonging to Group VIB and Group VIII of the Periodic Table carried on a carrier comprising 2-25% by weight of Y zeolite and 98-75% by weight of alumina or an alumina-containing substance, and wherein, (A) said alumina or alumina-containing substance (1) has a mean pore diameter of 7 to 10 nm (70-100 angstrom) and (2) contains the pore volume of which the diameter falls within  $\pm 1$  nm ( $\pm 10$  angstrom) of the mean pore diameter of 85-98% of the total pore volume, (B) said Y zeolite (3) has an average particle size of 2.5 to 6  $\mu\text{m}$  and (4) contains particles of which the diameter is 6  $\mu\text{m}$  or smaller of 70-98% of all zeolite particles, and (C) said catalyst contains at least one metal belonging to Group VIB of the Periodic Table in an amount of 2-30% by weight, in terms of an oxide, and at least one metal belonging to Group VIII of the Periodic Table in an amount of 0.5-20% by weight, in terms of an oxide."

Independent claim 16 related to a process for the hydrotreatment of a hydrocarbon oil by contacting said hydrocarbon oil in the presence of hydrogen with a catalyst composition as in claim 1.

Dependent claims 2 to 15 and 17 to 19 related to particular embodiments of the claimed catalyst or process.

II. In its notice of opposition the Appellant (Opponent) sought revocation of the patent on the grounds of Article 100(a) EPC, in particular because of an alleged lack of inventive step of the claimed subject-matter and cited inter alia the following document:

(1) = JP-A-8184639 (English translation).

Moreover, the wording of claim 1 filed with the letter of 31 October 1997 (see paragraph I above), maintained unamended in the set of claims further modified during the oral proceedings held before the first instance, was objected under Article 123(2) EPC.

III. In its decision, the Opposition Division found that

- the amended claims complied with the requirements of Article 123(2) EPC and in particular the lower value of average particle size (B)(3) of the zeolite Y in claim 1 found support in the original description;
- the selection of the catalyst features enabled to hydrotreat and particularly to desulphurise the hydrocarbon feed whilst providing a substantial cracking as shown in the examples of the patent in suit and in the comparative examples filed with the letter of 31 October 1997;

- the prior art and in particular document (1) did not suggest to operate this selection in order to solve this technical problem;
- the claimed invention and the patent in suit as amended fulfilled therefore the patentability requirements of the EPC.

IV. An appeal was filed against this decision and eight new documents (6) to (13) were cited by the Appellant in support of the arguments put forward in its statement of the grounds of appeal.

The Respondents and Patent Proprietors filed two auxiliary requests with a letter dated 15 October 1998.

V. The Appellant's arguments in regard to the admissibility of the additionally cited documents, submitted in writing and during the oral proceedings held before the Board on 6 September 2002, can be summarized as follows:

- the filing of additional documents had been rendered necessary by the maintenance of the patent in suit on the basis of amended claims filed only one month before oral proceedings and further modified during such oral proceedings;
- these documents were necessary for illustrating that
  1. it was known in the prior art that catalyst parameters such as the pore size distribution, the presence of zeolite Y as a cracking component and its particle size and amount in the catalyst, had an influence on the catalyst performance;

2. the selection of such catalyst parameters for optimizing the simultaneous hydrodesulphurisation and hydrocracking of heavy hydrocarbon feedstocks was part of the common general knowledge of the skilled person;
- in particular, document (6) to (12) showed that the selection of the catalyst parameters and their optimization were conventional in this technical field, whilst document (13) showed that it was advantageous to select a zeolite Y with an average particle size of 2 to 7  $\mu\text{m}$  as a cracking catalyst.

The Appellant argued, furthermore, that the incorporation into the wording of claim 1 of the value of 2.5  $\mu\text{m}$  as a lower limit for the range of average particle size of zeolite Y contravened the requirements of Article 123(2) EPC, since a range of particle size of 2.5 to 6  $\mu\text{m}$  had not been originally disclosed and the value of 2.5  $\mu\text{m}$  could be derived only from the specific examples of catalyst compositions of the application as filed. Moreover, this new range of particle size made a technical contribution to the claimed subject-matter and gave an unwarranted advantage to the Respondents by distancing further the claimed subject-matter from the prior art.

As regards the inventiveness of the claimed subject-matter it argued that

- document (1) disclosed a catalyst for the hydrotreatment of heavy hydrocarbon feedstocks comprising zeolite Y and alumina and metals of the groups VIB and VIII of the periodical table and having parameters overlapping with those of the patent in suit;

- the technical problem solved by the claimed invention amounted only to the provision of an alternative hydrotreatment catalyst;
- it was therefore obvious for the skilled person to select from the teaching of document (1) catalyst parameters falling within the ranges claimed in the patent in suit;
- moreover, it was known from the prior art how to select the catalyst features in order to optimize a simultaneous desulphurisation and cracking;
- the experimental results provided in the patent in suit and with the Respondents' letter of 31 October 1997 did not show any unexpected improvement obtained by the selection of the claimed catalyst parameters.

VI. The Respondents argued in writing and during the oral proceedings that:

- the additional references (6) to (13) were late filed and no more relevant than those cited before the first instance and had thus not to be admitted;
- the claims complied with the requirements of Article 123(2) EPC since the value selected as lower average particle size of zeolite Y had been used in at least 15 examples of the application as filed and was thus a value which the skilled person would have understood to be suitable for the claimed invention;

- as shown in the comparative tests of the patent in suit and in those filed with letter of 31 October 1997, the selected catalyst features led surprisingly to an improved simultaneous desulphurisation and cracking; this result was not suggested in document (1) or in the other documents of the prior art.

Therefore the claimed subject-matter involved an inventive step.

VII. The Appellant requested that the decision of the first instance be set aside and the patent be revoked.

The Respondents requested that the appeal be rejected or the patent be maintained on the basis of the first or second auxiliary request, both filed with the letter of 15 October 1998.

VIII. At the end of the oral proceedings, the chairman announced the decision of the Board.

## Reasons for the decision

### Main Request

#### 1. *Admissibility of the additionally cited evidence*

1.1 The Appellant has cited for the first time in the statement of the grounds of appeal eight new documents (6) to (13).

The filing of these documents was rendered necessary in the Appellant's view by the maintenance of the patent in suit on the basis of claims substantially modified only one month before oral proceedings and further modified during such oral proceedings, i.e. claims



containing features for which an additional search had not been possible before the first instance.

However, the Board finds that the only amendment to the claims not based on features which were already subject-matter of the granted claims was the introduction during the first instance proceedings of a lower limit of 2.5  $\mu\text{m}$  for the range of average particle size of zeolite Y. Therefore, the Appellant had ample time to file documents addressing at least the features of the granted claims before the appeal proceedings were started.

- 1.2 Documents (6) to (11) deal with the selection of catalyst parameters such as the pore size distribution of a hydrotreatment catalyst and the concentration of zeolite Y in the catalyst and concern therefore features which were already contained in the claims as granted.

Documents (12) and (13) address the particle size of a zeolite Y catalyst. However, they just confirm that there existed cracking catalysts based upon zeolite Y with an average particle size as required by the amended range of claim 1. The Respondents did not dispute during oral proceedings that such catalysts were known at the priority date of the patent in suit and in fact the claimed invention can make use of commercially available zeolite Y having a particle size within the range claimed, as explained in the patent in suit (see page 5, line 27). Therefore these documents address a point which has been acknowledged as known from the Respondents.

In the present case there were therefore no circumstances which could excuse the delay in producing the evidence in question and the new documents (6) to (13) must therefore in the Board's view be considered as late filed (see e.g. T 0715/95, not published in the OJ EPO, point 3 of the reasons).

- 1.3 It is established case law that late filed evidence should only be admitted at the appeal stage, if it can be considered at first sight to be more relevant than the evidence relied on at first instance and to be prejudicial to the maintenance of the patent (see, e.g. T 1002/92 OJ EPO 1995, 605, point 3.4 of the reasons).

The Board finds that from the Appellant's written statement read in combination with the therein indicated specific passages of the cited evidence, this newly cited evidence is no more relevant than that already on file.

For example, document (1), cited before the first instance, already illustrated, similarly to documents (6) to (11), that catalyst parameters such as the pore size distribution, the presence of a cracking component such as zeolite Y and its amount in the catalyst, have an influence on the catalyst performance (see paragraph 4.1 hereinafter); moreover, documents (12) and (13) relate to a zeolite Y catalyst and not to a mixed catalyst as in the patent in suit.

Therefore the Board concludes that the new cited documents (6) to (13) should not be admitted into the proceedings.

2. Article 123(2) EPC

2.1 The wording of Claim 1 differs from claim 1 as granted insofar as:

- the upper limit of the concentration of zeolite Y in the catalyst has been reduced to 25 wt% and the lower limit of the concentration of alumina or alumina-containing substance has been correspondingly increased to 75 wt%;
- the mean pore diameter range for the alumina or alumina-containing substance has been limited to 7 to 10 nm and the pore size distribution has been so limited that 85 to 98% of the total pore volume is comprised within  $\pm 1$ nm from the mean pore diameter;
- the zeolite is a Y zeolite;
- the range of average particle size for the zeolite Y has a lower limit of 2.5  $\mu$ m.

2.2 As agreed by the Respondents, the above mentioned lower limit of 2.5  $\mu$ m finds support only in the examples of the application as filed. In fact this value is used in 18 out of 21 examples relating to a catalyst comprising a zeolite. Since this value of particle size is used in the greatest part of the illustrative examples of the patent in suit and also in some comparative ones, the Board finds that the skilled person, by reading the application as filed, would have understood this value of particle size as one which could be used for performing the claimed invention independently from the selection of the other catalyst parameters. This value

is thus to be regarded as being not so closely associated with the other features of the examples as to determine the effect of those specific embodiments (see T 0201/83, OJ EPO 1984, 481, point 2 of the reasons).

Moreover, since the selected range was already **implicitly disclosed** in the application as filed, the Appellant's argument that the Respondents have achieved therewith an unwarranted advantage and that therefore the amendment contravenes Article 123(2) EPC cannot apply to the present case.

Therefore the Board concludes that the Respondents' selection of this value as a lower limit for the range of particle size of claim 1 complies with the requirements of Article 123(2) EPC.

- 2.3 The Board is also satisfied that all other amendments to claim 1 are based on the application as filed and comply with the requirements of Article 123(2) EPC. This was not contested by the Appellant and no further comment on this matter is necessary.

3. *Novelty*

The Appellant did not contest the novelty of the claimed subject-matter and the Board is also satisfied that the claims comply with the requirements of novelty.

4. *Inventive step*

- 4.1 The Board accepts document (1) as the most suitable starting point for discussing inventive step as agreed by both parties.

This document deals with a catalyst for hydrotreating hydrocarbon feedstock such as heavy oils containing asphaltenes, comprising zeolite Y and alumina and metals of the group VIB such as chromium or molybdenum and VIII of the Periodic Table (page 4, lines 7 to 24). Such a catalyst has a long life (see passage bridging pages 4 and 5).

The catalyst features disclosed in this document are the following:

- the average pore diameter of the mixture alumina/zeolite Y measured by mercury porosimeter is of 80 to 120 angstroms and the pore size distribution is such that at least 60% of the pore volume is within 20 angstroms from the average pore diameter (see page 5, lines 15 to 19);
- the amount of zeolite Y based on the weight of zeolite Y and alumina is preferably from 20 to 50% by weight (page 7, lines 3 to 5);
- the amount of metals of the VIB group is preferably of 7 to 16 wt% and that of metals of the VIII group is preferably of 1.5 to 5 wt% (see page 8, lines 24 to 29).

Since the pore diameter and the pore volume of the alumina is calculated in the patent in suit on the mixture zeolite Y/alumina, thus assuming that the very small pores of the zeolite Y would not affect the measurement (see page 4, line 51 to page 5, line 5 of the patent in suit), the ranges of mean pore diameters and pore size distribution disclosed in document (1) and indicated hereinabove, even considering the possible difference derived from the method of measurement used in document (1), can be compared with those of the patent in suit.

The average pore diameter disclosed in document (1) therefore overlaps with that of the patent in suit of 70 to 100 angstroms.

Moreover document (1) includes a range of pore size distribution of the catalyst which is much broader than that required in the patent in suit, wherein the pore diameter of 85-98% of the total pore volume must fall within  $\pm 1$  nm of the mean pore diameter. The very narrow pore size distribution disclosed in example 1 of document (1) (page 10, lines 21 to 22) can furthermore be disregarded, since it refers to the alumina before mixing with the zeolite Y, whilst the value measured after mixing with the zeolite (i.e. the value as calculated in the patent in suit), corresponds to a much broader distribution outside the range of disputed claim 1 (see table 1 on page 13: catalyst A).

Furthermore, the range of concentrations for zeolite Y and alumina overlap with those of the patent in suit, which are, respectively, 2 to 25 wt% and 98 to 75 wt%. The concentrations of the other metals correspond with those of the patent in suit.

Document (1), however, does not contain any disclosure of the average particle size of zeolite Y, which according to claim 1 of the patent in suit must be within the range of 2.5 to 6  $\mu\text{m}$ , and of its particle size distribution, whilst the disputed claim 1 requires that 70-98% of all zeolite Y particles have a diameter of 6  $\mu\text{m}$  or smaller.

Therefore document (1) does not disclose all the feature of the claimed catalyst in combination.

4.2 The technical problem underlying the claimed invention as defined in the text of the patent in suit was the provision of an alternative hydrotreatment catalyst which has high desulphurization activity and at the same time hydrocracking or denitrification activities and has the capability of maintaining its activity for a long period of time (page 2, lines 21 to 22 and 32 to 34 as well as page 9, lines 35 to 38).

As explained in the patent in suit, since both the hydrodesulphurization and the hydrocracking proceed in competition at the same active site, it is difficult to increase both reaction yields simultaneously and in particular to increase cracking whilst maintaining a high desulphurisation (page 2, lines 36 to 40). This is especially difficult in the hydrogenation of heavy hydrocarbon feedstock such as residual oils containing asphaltenes (see page 2, lines 34 to 35).

The patent in suit claims to have solved this problem by means of the specific catalyst features of claim 1, i.e. the combination of particular concentrations of the Y zeolite and of the alumina or alumina-containing substance, the pore diameter and the pore size distribution of the alumina and the particle size and the particle size distribution of the zeolite Y (see page 4, lines 39 to 41 and page 9, lines 6 to 23).

The comparative tests contained in the patent in suit show in table 4 on page 14 that a catalyst (Q) not containing zeolite Y or a catalyst (R) having too great an amount of zeolite Y and too low an amount of alumina or a catalyst (S) having a particle size of zeolite Y above the required range, provide inferior desulphurization and cracking in respect to the similar catalysts (A), (B), (D) and (E) of the patent in suit (see tables 2 and 3). Moreover, table 6 shows that

catalysts (T), (U) and (V), having a pore size diameter or a pore size distribution for the alumina outside the ranges defined for the invention, are inferior in desulphurization than the similar catalysts (O) and (P) according to the invention. Table 8 shows, moreover, that a catalyst (A) according to the invention has a long life.

4.4 The tests filed with the Respondents' letter of 31 October 1997 (considered by the Appellant as not showing any unexpected improvement throughout the whole range of selected catalyst parameters) compare two catalysts according to the disputed claim 1, i.e. a catalyst (A) having an average particle size of zeolite Y of 2.5  $\mu\text{m}$  and a catalyst (D) having a zeolite average particle size of 3.9  $\mu\text{m}$ , with catalysts (C), (X), (Y) and (Z) having a zeolite average particle size below 2.0  $\mu\text{m}$  (thus falling outside the claimed range of particle size); such comparisons are, however, not relevant for the assessment of inventive step since the catalysts (C), (X), (Y) and (Z) do not belong to the prior art. Therefore, these tests have to be disregarded.

4.5 However, as explained above (see point 4.3), the tests contained in the patent in suit prove convincingly that catalyst compositions according to the claimed invention solve the above mentioned technical problem.

The Board consequently finds that the technical problem underlying the invention is not just the provision of an alternative hydrotreatment catalyst, as argued by the Appellant, but that set out in the patent in suit as mentioned hereinabove (see point 4.2).



4.6 The question to be answered in the present case for the assessment of an inventive step is therefore whether a skilled person, making use of his common general knowledge, would have modified the catalysts specifically disclosed in document (1) by selecting catalyst parameters according to the patent in suit in order to solve the underlying technical problem.

Document (1) is silent about the importance of selecting a zeolite Y concentration and a pore size and pore size distribution of the catalyst as well as a particle size for the zeolite Y and a particle size distribution for the zeolite Y within the limits of the patent in suit, in order to obtain simultaneously a high desulphurisation and substantial hydrocracking of heavy oil feedstocks as convincingly shown in the comparative tests of the patent in suit. In fact it does not disclose in any of the illustrative examples a concentration of zeolite Y or an average pore size or a pore size distribution of the mixed catalyst alumina/zeolite Y in accordance with claim 1 of the patent in suit.

Therefore the Board finds that this document regards catalyst parameters falling within the ranges of the patent in suit at the best as being equivalent to those used in the illustrative examples.

The Board further finds that a skilled person, even knowing at the priority date of the patent in suit that the selection of catalyst parameters would affect the catalyst performance (as also suggested by the teaching of document (1)), would not have found in the prior art any suggestion for selecting and combining the

parameters of a catalyst mixture zeolite Y/alumina, as done in the patent in suit, in order to achieve a simultaneous high desulphurisation and cracking of heavy oil feedstocks.

Consequently, the subject-matter of claim 1 involves an inventive step.

4.4 Since the subject-matter of claim 1 involves an inventive step, the subject-matter of claim 16 (a process of hydrotreatment by means of a catalyst possessing all the features of that of claim 1) similarly involves an inventive step.  
The same applies to the dependent claims.

Since the claims according to the main request have been found to comply with the requirements of the EPC there is no need to deal with the auxiliary requests.

### Order

**For these reasons it is decided that:**

The appeal is dismissed.

The Registrar:

  
G. Rauh

The Chairman:

  
P. Krasa