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**D E C I S I O N**  
**of 13 June 2005**

**Case Number:** T 0596/04 - 3.3.6

**Application Number:** 99201148.6

**Publication Number:** 0950702

**IPC:** C10G 47/20

**Language of the proceedings:** EN

**Title of invention:**

Hydrocracking catalyst and hydrocracking method for hydrocarbon oils

**Applicant:**

Nippon Mitsubishi Oil Corporation

**Opponent:**

-

**Headword:**

Hydrocracking catalyst/NIPPON

**Relevant legal provisions:**

EPC Art. 83

**Keyword:**

"Sufficiency of disclosure (yes)"

**Decisions cited:**

T 0721/89, T 0728/98, T 0019/90, T 0484/92

**Catchword:**

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Case Number: T 0596/04 - 3.3.6

**D E C I S I O N**  
of the Technical Board of Appeal 3.3.6  
of 13 June 2005

**Appellant:** Nippon Mitsubishi Oil Corporation  
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**Representative:** Schnappauf, Georg, Dr.  
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**Decision under appeal:** Decision of the Examining Division of the  
European Patent Office posted 1 December 2003  
refusing European application No. 99201148.6  
pursuant to Article 97(1) EPC.

**Composition of the Board:**

**Chairman:** P. Krasa  
**Members:** G. Dischinger-Höppler  
A. Pignatelli

## Summary of Facts and Submissions

I. This appeal is from the decision of the Examining Division to refuse for insufficiency of disclosure the European patent application No. 99 201 148.6 relating to a hydrocracking catalyst and hydrocracking method for hydrocarbon oils. The decision under appeal was based on an amended set of 14 claims with independent Claim 1 reading:

"1. A hydrocracking catalyst for hydrocarbon oil, comprising:

- (i) a complex oxide selected from the group consisting of silicon-zirconium, silicon-aluminium-titanium, silicon-zirconium-aluminium, aluminium-boron, silicon-aluminium-boron,
- (ii) zeolite having a solid Al-NMR spectrum wherein the ratio A/B of the peak area A in a chemical shift of -30 to 18 ppm to the peak area B in a chemical shift of 20-100 ppm is 0.01-0.39 and whose surface area of pores of diameter 10 angstrom or smaller constitutes 10-85% of the total surface area, and
- (iii) at least one metal selected from Group 6a and Group 8 of the Periodic Table."

The claim set contains a further independent claim (Claim 6) relating to a hydrocracking method wherein the above defined hydrocracking catalyst is used.

II. During the examining proceedings, the Appellant filed the following documents:

- D4 A.G. Ashton et al., "Acidity in Zeolites" in "Catalysis by Acids and Bases", proceedings of an International Symposium organized by the Institut de Recherches sur la Catalyse - CNRS - Villeurbanne (Lyon), September 25-27, 1984, Elsevier 1985, pages 101 to 109;
- D5 L.D. Fernandes et al., "The effect of cyclic dealumination of mordenite on its physiochemical and catalytic properties" in "Zeolites", 1994, Vol. 14, pages 533 to 540; and
- D6 J. Klinowski et al., "Monitoring of structural changes accompanying ultrastabilization of faujasitic zeolite catalysts" in "Nature", Vol. 296, 1982, pages 533 to 536

with the intention to show that the zeolite of Claim 1 could be achieved by conventional methods known to those skilled in the art.

III. In its decision, the Examining division found that the application in suit did not meet the requirements of Article 83 EPC since the disclosure of the zeolite mentioned in part (ii) of Claim 1 was "inadequate". In particular, it was held that

- the application in suit did not give any instructions how a zeolite might be obtained having a structure as defined by the particular A/B ratio in combination with the requirement that the surface area of pores of  $\leq 10 \text{ \AA}$  in diameter constitute 10-85% of the total surface area (hereinafter F-value);

- no evidence was available to allow the Examining Division to decide how credible was the Applicant's contention that the skilled person would be able to carry out the invention on the basis of the information given in the application in suit and the common general knowledge in the art;
- D4 to D6 did not support the Applicant's view that the skilled person could work the invention; and
- the disclosure of the application in suit was insufficient to allow the skilled person to prepare compositions over the whole range of values of both parameters in a way that is reproducible and without undue burden or need of inventive skill.

IV. This decision was appealed by the Applicant (hereinafter Appellant) who filed a new request based on an amended set of 14 claims under cover of a letter dated 30 March 2005. The independent Claim 1 of this request differs from Claim 1 considered by the Examining Division (see I above) in that

- the term "which is an oxide of a combination of elements" has been introduced in item (i) between "a complex oxide" and "selected from ..." and
- "a" has been inserted in item (ii) before "zeolite".

The Appellant further filed experimental data under cover of a letter dated 7 September 2005.

V. The Appellant submitted the following arguments:

- According to the application in suit, zeolites suitable in the claimed catalyst were, for instance, ultrastable Y-type zeolites (USY).
- D4 to D6 showed that a person skilled in the art knows how to produce USY, how to modify the A/B ratio and the F-value in a zeolite and how to identify the A/B ratio by solid Al-NMR and the F-value by BET determination and t-plot analysis.
- Given the common general knowledge illustrated in D4 to D6 a person skilled in the art was, therefore, in a position to put the invention into practise without undue burden. This was supported by the experimental data.

VI. The Appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of the set of 14 claims filed under cover of a letter dated 30 March 2005.

## **Reasons for the Decision**

### **1. *Amendments (Article 123(2) EPC)***

The Board is satisfied that the claims as amended comply with the requirements of Article 123(2) EPC since their wording is supported by the application as originally filed (see original claims in combination with the description page 5, lines 7 to 14, page 7, lines 1 to 4, page 7, line 19 to page 8, line 3 and

page 9, lines 17 to 22 or, respectively paragraphs [0015], [0022], [0025], [0026] and [0031] of the A2 publication).

2. *Sufficiency of disclosure (Article 83 EPC)*

2.1 Article 83 EPC requires that the invention has to be disclosed in a manner sufficiently clear and complete for it to be carried out by a person skilled in the art.

2.2 In the present case, the claimed subject-matter relates to a hydrocracking catalyst comprising three different components, i.e.

(i) a particular complex oxide,

(ii) a particular zeolite and

(iii) a particular metal,

wherein component (ii) is defined by two parameters, namely the ratio A/B of peak area A to peak area B and the F-value as an indication of a particular pore size distribution.

2.3 The Examining Division's reasons on which the refusal of the application in suit is based concern only component (ii) and the question whether or not it is credible that a person skilled in the art, on the basis of its common general knowledge and the information given in the application in suit, would be able to reproducibly manufacture this component over the whole claimed range without undue burden or need of inventive skill (see III above).

2.4 In accordance with the established Case Law of the Boards of Appeal, it is in fact dependent on the common general knowledge of a person skilled in the art to which extent the steps required to achieve the claimed subject-matter have to be disclosed in an application in order to be sufficient for a skilled person to carry out the invention (see e.g. T 721/89, not published in the OJ EPO, reasons No. 3.5). Thus, it is essential for the assessment of sufficiency of disclosure to establish the disclosure of the application in suit in the light of the common general knowledge of the relevant technical field.

2.5.1 In the application in suit it is stated that a zeolite suitable for the purposes of the invention, i.e. for the purpose of providing a hydrocracking catalyst, is any normal zeolite provided it contains tetra- and hexa-coordinated Al atoms in particular amounts and has a particular pore size distribution. Particularly preferred is a Y-type zeolite, such as ultrastable Y-type (USY) zeolite.

It is further stated that the ratio between the amounts of tetra-coordinated and hexa-coordinated Al can be determined via solid Al-NMR spectroscopy. In particular, it is stated that the amount of hexa-coordinated Al is represented by the area of peak B present in the solid Al-NMR spectrum in the range of 20 to 100 ppm and the amount of tetra-coordinated Al is represented by the area of peak A present in the range of -30 to 18 ppm. For the purposes of the invention it is required that the A/B ratio ranges from 0.01 to 0.39 (column 3, line 26 to column 4, line 12).



2.5.2 As correctly indicated by the Examining Division, it is apparent from D4 to D6 that upon proper interpretation of the Al-NMR spectra, peaks appearing at a chemical shift of about 0 ppm correspond to hexa-coordinated Al whereas peaks at a chemical shift of about 50 ppm indicates tetra-coordinated Al. The above reverse attribution of the peaks according to the application in suit (peak A to tetra-coordinated Al and peak B to hexa-coordinated Al) is, therefore obviously wrong. However, since those skilled in the art would readily identify this obvious mistake and know how to correct it, it is not detrimental to the sufficiency of disclosure.

2.5.3 The Board has no doubts that a person skilled in the art knows how to determine the relative size of the peak areas in an Al-NMR spectrum but notes that the result might possibly depend on specific conditions, e.g. for carrying out spectroscopy. However, this would be a matter of clarity concerning the question whether the claims define the subject-matter for which protection is sought in a way ensuring that the public is not left in any doubt as to which subject-matter is covered by the claims and which is not (see e.g. T 728/98, OJ EPO, 2001, 319, reasons No. 3.1) which issue has to be addressed under the provisions of Article 84 EPC.

2.5.4 In accordance with the application in suit, the other requirement, the pore size distribution, is fulfilled if the F-value ranges between 10 and 85%. The F-value is defined as the surface area of pores having a diameter of 10 Å or less, expressed as the percentage

based on the total BET surface area of the zeolite. F is expressed by the equation

$$F = (S_a - S_b) / S_a \times 100$$

with  $S_a$  being the BET surface area and  $S_b$  being the surface area of pores having a diameter of more than 10 Å as determined by the t-plot method (column 4, lines 18 to 33).

- 2.6 The Board concludes, therefore, that the application in suit not only identifies the zeolites to be used and the essential parameters A/B and F but also gives instructions of how to determine their numerical values. It has not been disputed by the Examining Division that the methods indicated in the application in suit for measuring the parameters, i.e. Al-NMR spectroscopy, BET measurement and T-plot analysis are known in the art.
- 2.7 It is true, as stated by the Examining Division, that the application in suit does not disclose how the zeolite having an A/B ratio and F-value as defined in Claim 1 under (ii) is produced. However, the Appellant has filed D4 to D6 as evidence for the common general knowledge concerning zeolite treatment in relation with the parameters in question.
- 2.8 In the Examining Division's opinion it was still unlikely that a skilled person would achieve the claimed subject-matter by the application of trial-and-error in a way that is repeatable since there was no teaching in D4 to D6 concerning the extent of dealumination, i.e. the A/B ratio, and the treatment conditions necessary for obtaining the selected F-value.

Instead it was apparent from D4 that optimum steaming depended on the structure and composition of the parent zeolite (page 108, last sentence) and a variety of different treating methods was disclosed in D6, namely calcination, steaming and acid leaching. A further burden for the skilled person consisted in the fact that both parameters depended on the treatment conditions and had to be obtained in combination. Thus, the Examining Division concluded that a person skilled in the art would not know how to prepare zeolites having the required A/B ratio and F-value, let alone over the whole ranges of values given.

2.9 However, all three documents cited by the Appellant relate to hydrothermal treatment of zeolite, its consequences on the formation of octahedral (= hexacoordinated or extra-framework) aluminium by dislodgement of framework aluminium (= tetrahedral or tetracoordinated aluminium), and teach that  $^{27}\text{Al}$  magic angle spinning NMR spectroscopy (i.e.  $^{27}\text{Al}$  MASNMR), which gives solid state Al-NMR spectra, is the method for measuring these changes.

2.9.1 Thus, it can be seen from the relative intensities of peaks occurring in the solid Al-NMR spectra of D4 at chemical shifts of around 0 ppm and 50 ppm that dealumination of H-ZSM-5 increases with increasing steam pressure during hydrothermal treatment at 600°C for 2.5 hours (see experimental part on page 102 in combination with Figure 4 and page 105, second and third full paragraphs). This was also acknowledged by the Examining Division.

Likewise, the spectra given in D5 show that dealumination of mordenite occurs upon hydrothermal treatment at 823 K for two hours possibly followed by prolonged acid leaching and the spectra in D6 show that dealumination of faujasitic zeolite (NH<sub>4</sub>-Na-Y zeolite) occurred already upon calcination at 400°C for two hours but was higher upon steaming at 700°C for one hour and still higher when steaming was followed by acid leaching under reflux for two hours, thereby producing USY (see in D5, page 534, left-hand column, third full paragraph and last paragraph in combination with Figure 1 and page 535, right-hand column to page 537, left-hand column, sixth full paragraph; in D6, page 534, right-hand column, last full paragraph, left-hand column, first full paragraph to page 536, right-hand column, first full paragraph, in combination with Figure 3).

2.9.2 The Board observes that the claimed requirement for the A/B ratio to be within the broad range of 0.01 to 0.39 is most probably fulfilled in all those instances where the Al-NMR spectra illustrated in D4 to D6 show a strong peak at a chemical shift of around 50 ppm and a clearly weaker peak at a chemical shift of around 0 ppm (see in D4, Figure 4, spectra obtained at a steam pressure of 100 mm Hg and more; in D5, Figure 1, spectra for HM, H201 and H212; in D6, Figure 3).

2.9.3 Further, the Board wishes to note in this respect that it is not relevant for the claimed subject-matter to be sufficiently disclosed that - as noted by the Examining Division - D4 identifies a third species of Al, namely low symmetry polymeric Al, which appears in the Al-NMR spectrum near the tetra-coordinated framework Al

(page 105, third full paragraph) since Claim 1 is not limited in this respect but covers within the peak area B ranging from 20 to 100 ppm both species.

2.9.4 According to D5 and D6, steaming and acid leaching also change the porosity of the zeolite by creating a secondary mesopore system in the original zeolite. Thus, it is indicated in D6 that a secondary mesopore system with pore radii in the range of 15 to 19 Å is created by the treatment in addition to the original small pores of the faujasite with pore radii varying from 7 to 13 Å (see page 533, left-hand column, last sentence, in combination with page 536, right-hand column, second full paragraph). The same occurs with mordenite, a large-pore zeolite investigated in D5. Here it is stated that "t-plot analysis revealed formation of mesopores in the dealuminated samples" of the mordenite (page 533, right-hand column, last paragraph). Table 1 shows the extent of mesopore creation in relation to the treatment conditions, viz. hydrothermal treatment temperature, application of acid treatment or not and number of treatment cycles (see also page 534, "Sample preparation" and page 538, "Textural properties"). In Table 1, specific values for the BET surface area and the surface area of the mesopores as determined by t-plot analysis are given for a number of differently treated and untreated samples.

2.10 The Board concludes therefore, that a person skilled in the art knows, e.g. from D4 to D6, that hydrothermal treatment and acid leaching are suitable for modifying the A/B ratio and F-value in a given zeolite, that temperature, steam pressure and time of the treatment as well as the number of treatment cycles are the main

variables for different results and how to identify in an obtained product the respective A/B ratio via solid state Al-NMR and the F-value via BET measurement and t-plot analysis.

On the basis of this knowledge, it is in the Board's judgment only a matter of empirical investigation which does not impose excessive burden on those skilled in the art to arrive at the claimed subject-matter by varying for a given parent zeolite, e.g. a Y-type zeolite as preferred in the application in suit, the treatment conditions during steaming and acid leaching as suggested in D4 to D6. This is corroborated by the Appellant's experiments showing that the required catalysts can be obtained by working under treatment conditions similar to those disclosed in D4 to D6.

- 2.11 In contrast, the Examining Division's reasoning (see 2.8 above) is based on a suspicion rather than on evidence that those skilled in the art might be unable to arrive without undue burden, i.e. upon a reasonable number of trials, at a suitable zeolite. The Examining Division has not provided a single piece of evidence from which it would be plausible that - on the basis of the application in suit and the common general knowledge in the respective technical field - a skilled person would fail to produce with reasonable effort zeolites according to Claim 1. Likewise is the Examining Division's belief that the invention cannot be carried out within the whole claimed range and in a way that is repeatable, not based on verifiable facts but merely on the ground that the application in suit was lacking corresponding examples in support. This latter ground is not alone sufficient for an objection

under Article 83 EPC if in the light of the common general knowledge of the skilled person a detailed description is superfluous (see also T 19/90, OJ EPO, 1990, 476, reasons No. 3.2 and 3.3; T 484/92, not published in the OJ EPO, reasons No. 4; T 721/89, not published in the OJ EPO, reasons No. 3.6).

2.12 The Board concludes, therefore, that the Examining Division's line of argument is untenable since, as stated above, there is no reason to doubt that it was sufficient for a person skilled in the art to know which parent zeolite should be used and how to modify and identify therein the A/B ratio and the F-value to be able to put the invention into practice throughout the whole claimed scope in a reproducible manner and without undue burden or need of inventive skill.

For these reasons, the application in suit is deemed to comply with the requirements of Article 83 EPC.

### 3. *Remittal*

The Examining Division has refused the application in suit for insufficiency of disclosure without considering novelty and inventive step or any other requirements of the EPC. The Board, therefore, considers it appropriate to exercise its discretion under Article 111(1) EPC to remit the case to the first instance for further prosecution on the basis of Claims 1 to 14 submitted under cover of the letter dated 30 March 2005.

**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 with the order of further prosecution of the examining proceedings on the basis of Claims 1 to 14 submitted under cover of the letter dated 30 March 2005.

The Registrar:

The Chairman:

G. Rauh

P. Krasa