

Internal distribution code:

- (A) [] Publication in OJ
(B) [] To Chairmen and Members
(C) [X] To Chairmen
(D) [] No distribution

D E C I S I O N
of 6 June 2001

Case Number: T 0299/97 - 3.3.6

Application Number: 90100067.9

Publication Number: 0436049

IPC: C10G 47/16

Language of the proceedings: EN

Title of invention:

Middle distillate hydrocracking catalyst employing low acidity
Y zeolite

Patentee:

UOP

Opponent:

Akzo Nobel N.V.

Headword:

Hydrocracking catalyst/UOP

Relevant legal provisions:

EPC Art. 83, 52(1), 54(1)(2)

Keyword:

"Sufficiency - yes (measured value independent of particular
measuring conditions)

"Novelty - no (evidence provided by respondent shifts burden
of proof to appellant who submits insufficient counter-
evidence)"

Decisions cited:

-

Catchword:

-



Case Number: T 0299/97 - 3.3.6

D E C I S I O N
of the Technical Board of Appeal 3.3.6
of 6 June 2001

Appellant: UOP
(Proprietor of the patent) 25 East Algonquin Road
Des Plaines
Illinois 60017-5017 (US)

Representative: Weber, Dieter, Dr.
Weber, Dieter, Dr.
Seiffert, Klaus, Dipl.-Phys.
Lieke, Winfried, Dr.
Postfach 61 45
D-65051 Wiesbaden (DE)

Respondent: Akzo Nobel N.V.
(Opponent) P.O. Box 9300
6800 SB Arnhem (NL)

Decision under appeal: Decision of the Opposition Division of the
European Patent Office posted 14 January 1997
revoking European patent No. 0 436 049 pursuant
to Article 102(1) EPC.

Composition of the Board:

Chairman: P. Krasa
Members: G. Dischinger-Höppler
C. Rennie-Smith

Summary of Facts and Submissions

I. The appeal is from the decision of the Opposition Division to revoke European patent No. 0 436 049. The decision was based on the claims as granted as a main request and on amended claims according to an auxiliary request. Claim 1 as granted reads:

"1. A hydrocracking process for the production of middle distillates having boiling points of 149-371 °C in which a hydrocarbon feed stream is passed into contact with a catalyst composition comprising a hydrogenation component, a refractory inorganic oxide matrix and a crystalline hydrogen form Y-type zeolite, with the zeolite having a unit cell size between 2,420 and 2,440 nm (between 24,20 and 24,40 Å), at hydrocracking conditions and a product stream is recovered, **characterized in** that the zeolite has an ion exchange capacity greater than 0,07, and a NH₃-TPD acidity strength value of less than 2,00."

Independent Claim 8 refers to a hydrocracking catalyst composition as defined in Claim 1.

II. The opposition was filed by the Respondent (Opponent) against the patent in its entirety on the Grounds of Articles 100(a) EPC (in particular Article 54 EPC) and 100(b) EPC (Article 83 EPC). The opposition was based *inter alia* on the following documents:

(2) US-A-3 853 742;

(10) US-A-4 401 556 and

on experimental data concerning properties of a zeolite obtained by reworking Example 7 of document (2).

- III. The Opposition Division held that the claimed subject-matter was sufficiently disclosed but that the NH_3 -TPD acidity strength value was an unreliable parameter and therefore not to be considered as a distinguishing feature. Based on the Opponent's experiments concerning Example 7 of document (2), they found that the remaining features of Claim 1 of both the main and the auxiliary request were all in combination present in document (2).
- IV. With its statement of grounds of appeal, the Appellant (Proprietor) filed data obtained from several attempts to reproduce Example 7 of document (2).
- V. Amended sets of claims, submitted by the Appellant with its letter dated 25 May 2001 in auxiliary requests I and II, were withdrawn during the oral proceedings held before the Board on 6 June 2001.
- VI. The Appellant's arguments submitted in writing and orally can be summarized as follows:
- Without being explicitly mentioned, it was evident from the contents of the patent in suit that measuring a zeolites acidity by NH_3 -TPD (temperature programmed desorption of ammonia) must be done on a pre-dried zeolite sample. The disclosure given in the patent in suit was, therefore, not only sufficient within the meaning of Article 83 EPC but also reliable with respect to the NH_3 -TPD parameter. (This argument, made during the oral proceedings before the Board,

meant the Appellant abandoned its prior line of argument according to which "as received" samples with equilibrated water content was used for the NH_3 -TPD measurements.)

- None of the cited references disclosed catalysts having the claimed combination of features. This was shown by the Appellant's counter-evidence concerning Example 7 of document (2).
- Likewise, there was no indication in the prior art that such a combination of features would positively influence the midbarrel selectivity.

VII. The Respondent, in essence, argued as follows:

- Contrary to the Appellant's previous line of argument that the NH_3 -TPD measurement was made on as-received samples with equilibrated water content, its new argument that pre-dried material was used amounted to a change of the legal framework of the case which was not allowable.
- Even if the Appellant's new argument should be accepted, vital information concerning the NH_3 -TPD measurement conditions was still missing since the patent in suit did not indicate any drying conditions; the alleged invention was therefore not sufficiently disclosed and the obtained values were not reliable.
- The claimed subject-matter was anticipated by the disclosure of document (2) as was shown by the Respondent's experimental results obtained by reworking Example 7 of document (2).

VIII. The Appellant requested that the decision under appeal be set aside and that the patent be maintained.

The Respondent requested that the appeal be dismissed.

Reasons for the Decision

1. *Sufficiency of disclosure*

The objection was raised with respect to the NH₃-TPD acidity strength value of less than 2.00 mentioned in Claim 1 as one of the essential features of the claimed subject-matter.

1.1 From the very beginning of the opposition proceedings, the Respondent challenged sufficiency of disclosure of this parameter on the basis of not being able to reproduce the value of 2.60 given for the non-modified starting zeolite LZ-Y82 (page 8, lines 8 to 11) and of insufficient description of the measuring method (page 4, line 41 to page 5, line 4). It pointed out in particular that the patent in suit did not say whether the 0.250 ± 0.001 gram starting sample was a dry sample or, if not, how much humidity it could contain.

1.2 The Respondent has never questioned whether the method of measurement disclosed in the patent in suit can be carried out on wet or dry samples. The gist of its arguments aims rather at a problem concerning the interpretation of a measured value.

The Board is satisfied that in the present case the lack of indication of certain measuring conditions is not detrimental to the sufficiency of the disclosure but could raise a clarity problem with the consequence that the particular value must be interpreted in a broad manner or, in other words, any value obtained by a NH₃-TPD method which falls within the claimed range of

values would, regardless of whether the sample was wet or dry, anticipate this range of values. Since the appeal fails for other reasons, it is not necessary to consider this further.

2. *Legal framework of the opposition*

The Respondent objected to the Appellant's change of argument concerning the dryness of the material to be tested by NH₃-TPD (see 1.1 above) as amounting to a change of legal framework of the opposition.

It has been the Respondent's own argument, in its response to the statement of grounds of appeal, that a skilled person would infer from the patent in suit that the zeolite samples used for NH₃-TPD measurement must be pre-dried. It seems to be clear that the Appellant was finally persuaded by this line of argument that - contrary to its initial conviction - the samples cannot be as-received samples containing water. Hence, the Appellant merely changed its opinion insofar as it now agrees with the Respondent as to the sample to be used for NH₃-TPD measurement. The Board does not see how the legal framework of an opposition which is defined by the extent to which and the grounds upon which a patent has been opposed (see Case Law of the Boards of Appeal, 3rd edition 1998, English version, page 434, last paragraph) can be changed by such an agreement.

3. *Novelty*

The patent in suit relates to a hydrocracking catalyst and to the production of middle distillates by hydrocracking a hydrocarbon feed in the presence of such a catalyst (page 2, lines 5 to 7). Hydrocracking

catalysts are known in the art, e.g. from document (2), and usually comprise a hydrogenation component, a refractory inorganic oxide and a zeolite (see patent in suit, page 2, lines 24 to 30 and document (2), column 2, lines 1 to 20).

3.1 The patent now aims at the provision of a mid-barrel hydrocracking catalyst with increased selectivity and suggests that this can be achieved if the catalyst contains a hydrogen form Y zeolite having a low concentration of bound hydroxyl groups on its surface as characterized by NH_3 -TPD as a means for determining the acidity strength of the zeolite (page 3, lines 52 to 56 and page 4, lines 34 to 35). According to Claims 1 and 8 of the patent in suit it is essential for such a catalyst that the zeolite contained therein has a NH_3 -TPD acidity strength value of less than 2,00 and, in addition, an ion exchange capacity greater than 0.07 as well as a unit cell size between 2,420 and 2,440 nm (see also page 5, lines 15 to 16 and 21 to 22).

3.2 Document (2) also relates to the increase of midbarrel selectivity. It is suggested in this citation to use a particular class of zeolites in the catalyst composition for this purpose (column 1, lines 51 to 57), preferably zeolites having a faujasite crystalline structure (column 3, lines 57 to 63) in hydrogen form (column 4, lines 1 to 3), which uncontestedly is a Y zeolite converted to its hydrogen form (see also Examples 1 and 7).

The only features of the catalyst of Claim 8 of the patent in suit which are not mentioned in document (2) are the NH_3 -TPD acidity strength value and the ion exchange capacity value for the selected zeolite.

Whilst document (2) is completely silent on these features, it is, however, stated there that the zeolite is obtained by a particular modification, including a steam treatment which will reduce the unit cell size and give a value of below 2,450 nm, preferably between 2,440 and 2,450 nm, for the preferred faujasites (column 4, lines 18 to 39). This product is thereafter re-exchanged to reduce further the sodium content and calcined (column 5, lines 43 to 51). One such modifying treatment is described in detail in Example 7. This example also describes the manufacture of a hydrocracking catalyst from the product zeolite obtained in combination with alumina as refractory inorganic oxide and nickel and tungsten as hydrogenation components. Example 8 then shows how middle distillates boiling below 363°C (685°F) are obtained by using this catalyst in a hydrocracking process.

- 3.3 With its notice of opposition, the Respondent filed an experimental report concerning its repetition of Example 7 of document (2) on the basis of a regular NaY zeolite which - after the steaming step - had a unit cell size of 2,445 nm. The final re-exchanged and calcined zeolite was determined to have a unit cell size of 2,434 nm.

Ion exchange capacity of the obtained product was determined according to the method described in document (10), since this was the only reference given in the patent in suit in this respect. It was found to be 0.073.

Further, the NH₃-TPD acidity strength of the product was determined by different measuring techniques to be

either 1.9 or 0.9 on a dry basis.

The Respondent concluded that the subject-matter of Claims 1 and 8 of the patent in suit was anticipated by that particular example.

3.4 The Appellant, with its statement of grounds of appeal filed experimental counter-evidence concerning Example 7 of document (2), allegedly carried out by different experienced workers. The Appellant argued that the description of this example was not detailed enough so that the gaps in the described method had to be filled from the skilled people's knowledge. Three rather different results were obtained, none of them comparable with the Respondent's results. Accordingly, none of them showed the features set out for the zeolite in Claims 1 and 8 of the patent in suit. From this, the Appellant concluded that Example 7 of document (2) did not automatically result in a catalyst having the claimed combination of features.

3.5 The Appellant argued that the unit cell size of 2,440 to 2,450 nm mentioned in column 4, lines 36 to 38 of document (2) was not related to the steam treated intermediate product of Example 7. Further, it was evident from Examples 15 and 16 that the products of document (2) may well be amorphous. Thus, a skilled person when trying to rework the teaching of document (2) had to select production conditions in relation to the product he wishes to obtain.

3.6 The Board finds neither the Appellant's arguments nor its counter-evidence to be convincing for the following reasons:

The Appellant mentioned only one particular parameter which had to be selected in document (2), namely the starting zeolite. The Board agrees with the Appellant that in document (2) this selection has to be made. However, document (2) already gives the necessary clear instructions to that end. It is stated that preferred zeolites are those which have a faujasite crystal structure (column 3, lines 61 to 62) and a unit cell size of below 2,440 nm after steaming (column 4, lines 35 to 39). Thus, the preferred starting zeolite to be used is defined by the unit cell size of the "intermediate zeolite product" after this treatment step. Since examples normally illustrate the preferred embodiments of an invention, these statements in the description of document (2) must have a significance for the material to be selected in the examples. This is not in contradiction to Examples 15 and 16 mentioned by the Appellant. Whilst the zeolite used in Example 15 is made by ion exchange and steam treatment according to the instructions of Example 1 and gives crystalline material even after rehydration and acid washing (see also Table 4 and 5), Example 16, which results in amorphous products after such further treatment, is a comparative example wherein the zeolite has obviously not been steamed.

The Board is therefore presented with conflicting experimental evidence and must decide - on the balance of probability - which party's evidence is more reliable.

In its experiments, the Respondent clearly indicated the composition of the starting zeolite and how this material was treated in accordance with the instructions given in Example 7 of document (2).

Accordingly, a regular NaY zeolite of given composition was ion exchanged and steam treated according to Example 1, to which Example 7 refers in this respect, to give a unit cell size of 2,445 nm and a sodium oxide content of at least 1% by weight as required for this stage (column 4, lines 28 to 39 and column 5, lines 11 to 20). After re-exchange and calcination of this product in accordance with Example 7 a final zeolite resulted having a unit cell size of 2,434 nm as well as a sodium oxide content and a pore size distribution similar to that given in Example 7. Since, document (2) itself does not leave any room for supposing that other results could be obtained by following these instructions, the Respondent provided convincing evidence of the fact it alleged, i.e. that a catalyst as claimed in Claim 8 of the patent in suit was already disclosed in Example 7 of document (2).

On the other hand, it is apparent from the three different results submitted by the Appellant, that its experiments must have been carried out under varying conditions. However, no details whatsoever have been given of how the different experienced workers conducted their experiments to rework Example 7 of document (2). It is, therefore, impossible to know how the different results were arrived at. In this respect, the Board considers it is not sufficient merely to indicate that gaps in the description of the method according to Example 7 of document (2) have been filled without specifying which gaps have been filled in what manner in which experiment.

The evidence the Appellant provided merely shows that - somehow or other - products may be obtained which fall outside the claimed combination of features. It did

not, however, present any evidence or arguments showing that another NaY zeolite and/or different experimental conditions than those used by the Respondent would not, without departing from the instructions cogently prescribed in the reworked Example 7, result in a product zeolite having the desired unit cell size, ion exchange capacity and NH₃-TPD acidity strength set out in Claims 1 and 8 of the patent in suit.

- 3.7 The Board, therefore, has no reason to doubt that the experiments of the Respondent are representative of the products which are necessarily obtained by a skilled person following the instructions of Example 7 of document (2). For these reasons, the Board concludes that this example and the use of the obtained catalyst according to Example 8 of document (2) anticipate the combination of features of the catalyst of Claim 8 and the process of Claim 1 of the patent in suit. Therefore, the subject-matter of these claims is not novel (Articles 52(1) and 54(1) (2) EPC).

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar:

The Chairman:

G. Rauh

P. Krasa