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Bezeichnung der Erfindung: Catalytic dewaxing of hydrocarbon oils with synthetic offretite
Title of invention:
Titre de l'invention :

Klassifikation / Classification / Classement : C10G

ENTSCHEIDUNG / DECISION
vom / of / du 21 February 1985

Anmelder / Applicant / Demandeur : Mobil Oil

~~Patentinhaber / Proprietor of the patent / Titulaire du brevet~~

~~Einsprechender / Opponent / Opposant~~

Stichwort / Headword / Référence :

EPÜ / EPC / CBE Art. 83, 123

"disclosure - by way of reference"
"amendment of claims"

Leitsatz / Headnote / Sommaire

Structural features of a means for performing a chemical process (here: the catalyst "offretite") which are not mentioned in the application documents themselves but in a document (here: a Canadian patent specification) to which they refer may be incorporated into a patent claim if they unequivocally form part of the invention for which protection is sought. However, all the essential structural features thus disclosed which belong together must be incorporated into the claim; it is not permissible to single out a particular one of their number (here: a silica/alumina ratio).

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Case Number: T 6 / 84

DECISION

of the Technical Board of Appeal 3.3.1

of 21 February 1985

Appellant: Mobil Oil Corporation
150 East 42nd Street
New York, New York 10017
U S A

Representative: Alan Harry West
Mobil Court
3 Clements Inn
London WC2A 2EB
England

Decision under appeal: Decision of Examining Division 029 of the European Patent Office dated 27 July 1983 refusing European patent application No 80 300 440.7 pursuant to Article 97(1) EPC

Composition of the Board:

Chairman: K. Jahn
Member: G. Szabo
Member: F. Benussi

Summary of Facts and Submissions

I. European patent application 80 300 440.7, filed on 15 February 1980, published on 1 October 1980 with publication number 16 530 and claiming the priority of an earlier application of 19 March 1979 (US-21 735), was refused by decision of the Examining Division 029 of the European Patent Office dated 27 July 1983. The decision was based on Claims 1 to 12 submitted on 28 August 1982. The main claim was worded as follows :

"1. A process for catalytic dewaxing a waxy hydrocarbon oil which comprises contacting said oil with a catalyst at dewaxing conditions characterized in that the catalyst is synthetic offretite having a silica/alumina mole ratio of 5 to 10."

II. The stated ground for refusal was that the subject-matter of claims 1 to 12 did not involve an inventive step. GB-A-2 001 668 (1) described a process for the catalytic dewaxing of waxy hydrocarbon oils by using a shape-selective zeolite catalyst and mentioned offretite in this respect. The process according to Claim 1 of the application under appeal differed from the cited known process by the use of an offretite having a silica/alumina mole ratio of 5 to 10, whilst the one used in the cited art had to have a ratio of "above 12". CA-A-934 130 (2), however, had already disclosed the preparation of a synthetic offretite type of material having a ratio of 5 to 10 for the purpose of non-selective cracking and hydrocracking large-size hydrocarbons. The same document also contemplated a similar treatment of "lower molecular weight hydrocarbons" or the mixture of both types. The utilisation of the offretite having a lower mole

ratio instead of material with a higher ratio, suggested in the state of the art, could not be regarded as an invention since the former was recommended for a very similar process, e.g. cracking or hydrocracking purposes irrespective of the size of the molecule.

III. On 28 September 1983 the applicants filed an appeal against the decision of the 27 July 1983, paying the fee at the same time. They submitted a Statement of Grounds on 25 November 1983.

IV. Invited by the Technical Board to provide further explanations, the appellants responded in due time, and submitted at the oral hearing held on 21 March 1985 a set of claims with the main claim limited as follows :

"1. A process for the catalytic dewaxing of a waxy hydrocarbon oil which comprises contacting the oil in the presence of hydrogen with a catalyst under dewaxing conditions, characterized in that the catalyst comprises

a) synthetic offretite having a compositional formula in terms of mole ratio of oxides of

$$(1.1\pm0.4)\ M_{2/n}O : Al_2O_3 : (5\text{-}10)SiO_2 : (0\text{-}8)\ H_2O$$

in which M is a mixture of hydrogen and a hydrogenating metal and n is the valence of M, and having an X-ray powder diffraction pattern substantially as follows :

.../...

2 Times Theta (Θ)	Relative Intensity I/I_o	Interplanar Spacing, d(A)
7.7	100	11.45
11.75	16.5	7.54
13.4	55.2	6.63
14.05	9.9	6.30
15.43	15.0	5.74
19.42	26.5	4.57
20.47	43.3	4.34
23.7	89.2	3.76
24.85	43.0	3.59
26.9	18.6	3.31
28.3	17.4	3.15
30.5	9.5	2.93
31.35	79.7	2.85
33.32	19.1	2.68
35.90	13.8	2.51

where theta (Θ) is the Bragg angle,

I is the observed peakheight,

I_o is the intensity of the strongest line or peak,

and d is the interplanar spacing in Angstrom units;

or (b) synthetic offretite as defined in (a) above but in which M is hydrogen, associated with a hydrogenation metal."

V. The appellants submitted substantially the following in support of the appeal :

(a) The skilled man would either dismiss from further consideration as irrelevant the idea of using offretite with a silica/alumina ratio above 12 or consider the same unsupported by the document as a whole. According to the available literature, offretites have silica/alumina ratios around 5 to 10, and none of the sources suggest a ratio higher than 12. This

.../...

is confirmed by Dr. Chen in his affidavit submitted on behalf of the appellant. The affiant also states that in order to provide such material, known methods would have to be applied which effectively dealuminise the lower-ratio material. Document (1) itself gives no guidance as to the origin of the offretite recommended for its purposes and gives no information about its ratio. In view of the requirement in the claim of the application under appeal that the offretite should be of the type having a ratio of 5 to 10, the process in question is novel and could not be derived from the cited documents.

(b) Although catalytic activity can generally be considered to be related to the silica/alumina ratio, many other factors influence such activity. In addition the zeolite is required to exhibit a very high order of selectivity for dewaxing which is more important than the degree of catalytic activity. Information now available suggests that the success of offretite in comparison with ZSM-5 is attributable to the puckered 12-membered oxygen rings which contain stacking faults.

(c) The specific selectivity of the relevant offretite was in no way suggested by document (2) (cf. also equivalent DE-A-1 806 154). In view of the express statement in the document that the cracking and hydrocracking is non-selective ("especially of larger-size hydrocarbons"), the actual selectivity and superiority over the more conventional and commercially accepted ZSM-5 dewaxing catalyst of (1) are surprising.

.../...

(d) The comparative tests in Example IV show that the claimed process results in a much better Viscosity Index (V.I.) whilst maintaining a satisfactory pour point when compared with the use of ZSM-5 with a metal. This, according to the appellants, proves that the invention solves the stated problem satisfactorily and unexpectedly.

VI. The appellants request that the decision under appeal be set aside and a European patent granted.

Reasons for the Decision

1. The appeal complies with Articles 106 to 108 and Rule 64 EPC and is, therefore, admissible.

2. There is no formal objection to the current version of the claims. The main claim relies for the definition of the synthetic offretite on p. 8, lines 25 to 27 of the specification. The relevant passage refers to CA-A-934 130 (2) which, in turn, specifies (cf. Claims 1, 2 and 4 on pages 19 and 20 of that document) the offretites used for the present invention as aluminosilicates having the given mole ratios for the oxides and a characteristic X-ray powder diffraction pattern. It is the view of the Technical Board that structural features of a means for performing a chemical process which are not mentioned in the application documents themselves but in a document to which they refer may be incorporated into a patent claim if they unequivocally form part of the invention for which protection is sought. However, all the essen-

tial structural features thus disclosed which belong together must be incorporated into the claim; it is not permissible to single out a particular one of their number.

3. As to the amendment of the main claim by incorporating certain characteristics from document (2) in order to define the offretites used for the purposes of the invention claimed in the present application, the Board is therefore of the opinion that the mere limitation to a silica/ alumina mole ratio of 5 to 10, as claimed before the Examining Division, is insufficient. In the absence of evidence that this feature alone, together with the general term "offretite", is a sufficient characterisation, it is necessary to recite fully the other components of the structure and the diffraction pattern figures, as they were originally disclosed and defined in document (2). Nevertheless, the further criterion also mentioned is the same document , i.e. the capability of absorbing cyclohexane at least to a specified degree, is considered unnecessary in the circumstances since this condition is assumed to be fulfilled in consequence of the essential features of the claim.

Some other features of the main claim are directly derived from former Claims 2 and 4 as originally filed, or are necessarily required when using the offretite in association with a hydrogenation metal in or on the offretite (cf. from p. 8 last paragraph to p. 9, line 9, and (2), pp. 19 and 20).

4. The closest state of the art in GB-A-2 001 668 (1) describes the preparation of high-quality oil from waxy crude oil by a process which includes the step of catalytic dewaxing by hydrogenation in the presence of an

aluminosilicate zeolite catalyst having a silica/alumina ratio above 12 and a constraint index between 1 and 12. The aim was to obtain a good yield and a very low pour point (p. 2, line 20). The only zeolite catalyst exemplified in the document was ZSM-5 with a hydrogenation metal such as nickel. Regarding criteria for selection (cf. page 5, lines 11 to 14), ZSM-5 tops the list with regard to constraint index and framework density. This particular zeolite was clearly preferred (cf. also page 1, line 63). Zeolites having mole ratios of at least 30 are apparently the best (p. 2, line 44).

5. The problem with which the invention was concerned was to improve the selectivity of performance and the yield with the catalyst, if not the activity, whilst reducing the pour point to -3.9°C or below. (cf. application page 4, lines 10 to 17; p. 5, line 25 to page 6, line 3). The problem was solved by the use of offretites known in the art as defined in Claim 1, which have - inter alia - a silica/alumina ratio of 5 to 10.

It is credible that this problem has been effectively solved, since the results are better than those achieved with ZSM-5 and a hydrogenation metal under comparable circumstances. Table IV of the present specification convincingly shows for instance (p. 15) that the yield and Viscosity Index (V.I.) which according to the appellants are characteristic of selectivity, are significantly improved with offretite (yield 101.3%, V.I. 98) as compared with ZSM-5 (yield 97.6%, V.I. 88) whilst the pour point remains at the same but acceptable level (-3.9°C). Table V in Example 4 nevertheless demonstrates that much lower pour points could be achieved with the claimed methods, if desired.

.../...

6. Document (1) contains no examples with offretite, although the general description mentions this type of zeolite as a possible means for dewaxing. In view of the fact that it is repeatedly emphasised that a suitable zeolite should have a silica/alumina mole ratio of at least 12 and preferably above 30 (p. 2, line 44), no use of offretites limited to a mole ratio of 5 to 10 is revealed or implied. The claimed solution, limited to this range, is therefore novel over the cited art (1).

Document (2), which on the other hand discloses the preparation and the general use of such offretites in the "non-specific hydrocracking" of large-size hydrocarbons, does not indicate the specific application of the technique to waxy oils containing, as is known, unbranched paraffins (cf. present specification p. 5, lines 15 to 19) . The use of the same zeolites with a selective effect for dewaxing purposes is therefore also novel in respect of document (2).

7. As already stated, the process disclosed in document (1) relates to the same problem of dewaxing oils and uses zeolites, including offretite, for the purpose. Nevertheless, there is no guidance at all as to how to obtain or select an offretite when it comes to the much emphasised silica/alumina ratio. It is disclosed that an offretite of otherwise unknown characteristics has a constraint index of 3.7 (p. 3, line 24) which falls within the range specified in the claims (1 to 12), but it must be assumed that the true ranges specified in the main claim of the cited patent, i.e. the mole ratios and the constraint indices, are independent criteria and the compliance with one does not necessarily imply compliance with the other. It adds to the uncertainty that

according to the appellants' submissions, and evidence from Dr. Chen, no direct reference to an offretite of a mole ratio above 12 could be located.

8. It appears that the mentioning of offretite in document (1) is, at worst, not an enabling disclosure or, at best, something which would hardly function advantageously in the light of the problem even if it had a ratio above 12. In neither case could the disclosure be considered as a proper basis for the skilled man to consider offretites with mole ratios from 5 to 10 to be a likely proposition which would offer an advantage or be worth trying. Doubts about suitability and reproducibility within the broadest claim in the cited patent cannot lead to the notion that all is well then in the twilight zone outside the limitations of the claim. Nor is there any reason to assume that the skilled man would have treated the repeated emphasis on the importance of high mole ratio as an obvious error to be replaced unequivocally by a much lower ratio. This would not have been been suitable for the other zeolites which clearly supports the validity of the high mole ratio as a condition.

9. It can therefore be concluded that the choice of offretites having a silica/alumina mole ratio within the range of 5 to 10 is in no way foreshadowed by or derivable from (1). Still less could it be predicted that the same catalyst would deliver the improved selectivity required by the problem to be solved by the invention. According to the appellants, the higher selectivity manifests itself in the better yield, and particularly in the significantly higher V.I. value when Pt/offretite is compared with Ni/ZSM-5 (cf. first and last column in

Table IV, p. 15). The latter has, admittedly, not only a higher catalytic activity than offretite but is also, because of this, a very much favoured and commercially used catalyst in the art.

The higher selectivity of the offretites recommended in the application under appeal is surprising since the comparison of the constraint indices for the two materials (1) (page 3, lines 18 and 24) would suggest the opposite. A recent publication by some of the inventors suggests that a low concentration of randomly distributed stacking faults in the offretite may be responsible for this surprising and useful anomaly (Chen, N.Y. et al, Journal of Catalysis, 1984, 86, 24).

10. Since document (2), disclosing offretites with the reqired 5 to 10 mole ratio, emphasises the non-selective character of its use in hydrocracking, the selectivity of its application in removing waxy substances from oils is unexpected. There was no good reason to replace the high activity zeolites of document (1) with a specific offretite of this kind, since this would have been quite contrary to the stated preferences for choice in the disclosure. The process using the catalyst now claimed therefore involves an inventive step. The same applies to the subject-matter of Claims 2 to 10, since these are fully dependent on the main claim.

11. The Board cannot therefore concur with the grounds and conclusions of the impugned decision of the first instance. However, the patent applied for cannot be granted at present as the description in the specification has not yet been brought into line with the claims as amended.

.../...

Order

It is decided that :

1. The decision of the Examining Division of 27 July 1983 is set aside.

2. The case is remitted to the Examining Division with the order to grant a European patent on the basis of Claims 1 to 10 submitted at the oral proceedings.

The Registrar

The Chairman