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D E C I S I O N
of 20 October 2000

Case Number: T 0609/97 - 3.3.6

Application Number: 88111807.9

Publication Number: 0305720

IPC: C10G 11/20

Language of the proceedings: EN

Title of invention:

Production of gaseous olefins by catalytic conversion of hydrocarbons

Patentee:

CHINA PETROCHEMICAL CORPORATION, et al

Opponent:

Akzo Nobel N.V.

Headword:

Olefin production/CHINA PETROCHEMICAL

Relevant legal provisions:

EPC Art. 56

Keyword:

"Inventive step - yes"

"Further improvement of a yield not foreshadowed in citations disclosing low yields"

Decisions cited:

G 0004/92, T 0133/92

Catchword:

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Case Number: T 0609/97 - 3.3.6

D E C I S I O N
of the Technical Board of Appeal 3.3.6
of 20 October 2000

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Decision under appeal: Interlocutory decision of the Opposition Division
of the European Patent Office posted 9 April 1997
concerning maintenance of European patent
No. 0 305 720 in amended form.

Composition of the Board:

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

Summary of Facts and Submissions

- I. This appeal is from an interlocutory decision of the Opposition Division to maintain European patent No. 0 305 720 relating to the production of gaseous olefins by catalytic conversion of hydrocarbons in amended form.
- II. In the notice of opposition, based on lack of inventive step, the following documents had, inter alia been submitted

(2) US-A-3 835 029 and

(3) US-A-3 849 291.

During the opposition proceedings, the Appellant (Opponent) further cited

(5) L.J. McPherson and M.F. Olive, Cracking and Reforming, in Modern Petroleum Technology, 5th Edition, Part I, Ed. G.D. Hobson et al., John Wiley & Sons 1984, chapter 14, pages 395 to 435;

(6) F.G. Dwyer et al., Octane enhancement in FCC via ZSM-5, NPRA Annual Meeting San Antonio 1987, pages 1 to 4; and

(7) G.T. Austin, Shreve's Chemical Process Industries, Mc Graw Hill Book Company, 1984, 5th Edition, pages 736 to 740.

- III. In its decision, the Opposition Division held that, starting from document (2) as the closest prior art, none of the other prior art documents hinted at the

claimed combination of features in order to attain the desired high yields in propylene and butylene from high boiling hydrocarbon feedstock by a catalytic cracking operation.

IV. In its statement of grounds of appeal, the Appellant objected to the amended Claim 1 under Article 123(3) EPC and maintained that the subject-matter of this claim lacked an inventive step in view of, inter alia, the following further documents

- (8) H.H. Voge, Chapter 5, Catalytic Cracking, Catalysis, Vol. VI (P.H. Emmet ed.), Reinhold, New York (1958), pages 407 to 411;
- (9) Advances in Catalysis, Volume VI, Academic press (1954), pages 360 to 365 and 402 to 417;
- (10) J.H. Gary and G.E. Handwerk, Petroleum Refining: Technology and Economics, Second Edition, Revised and Expanded (1984), pages 114 to 116;
- (11) E.V. Murphree, Advances in Chemistry No. 5, 30, (1951) pages 30 to 38;
- (12) US-A-3 758 403;
- (13) E.G. Wollaston et al., Hydrocarbon Processing, September 1975, pages 93 to 100; and
- (14) G.W.G. Mc Donald, Oil and Gas Journal, April 1, 1985, pages 111 to 115.

V. With its letter of reply dated 5 March 1998, the Respondents (Proprietors) filed an amended Claim 1 in

order to overcome any deficiencies under Article 123(3) EPC.

VI. In a communication dated 26 September 2000, the Board informed the parties that one issue to be decided was whether the amendments made to Claim 1 complied with the requirements of Article 123 EPC since it did not correspond to Claim 1 either as granted or as originally filed.

VII. Oral proceedings were held before the Appeal Board on 20 October 2000, in the absence of the Appellant as announced by a letter of 26 September 2000. During these proceedings, the Respondents withdrew their previous requests that late filed documents (8) to (14) be excluded and for apportionment of costs. They also filed a re-amended independent Claim 1 together with four dependent claims as their sole request, Claim 1 reading:

"1. A process for preparing gaseous olefins by catalytic conversion which comprises contacting petroleum hydrocarbon feedstock under cracking conditions with a solid acidic catalyst in the presence of steam,

characterized in that said feedstock comprises

- vacuum gas oil,
- residual oil and
- mixtures thereof, or
- crude oil; and

is contacted with a microspherical acidic zeolite catalyst comprising

pentasil shape selective molecular sieves and/or USY (ultra stable hydrogen Y) zeolites as active components and

matrix material selected from synthetic inorganic oxides, mineral clays and mixtures thereof in a fluidised or moving bed or transfer line reactor

at a temperature of from 500°C to 650°C and

at a pressure between 1.5×10^5 Pa and 3.0×10^5 Pa

with a weight space velocity of 0.2 to 20 hr⁻¹,

a catalyst-to-oil ratio of 2 to 12, and

a steam-to-feed ratio of 0.05 to 1:1 by weight,

to carry out the cracking reaction."

VIII. The Appellant in writing submitted four different approaches for the assessment of inventive step. In summary it argued as follows:

- Starting from document (2), a person skilled in the art wishing to increase the yield of gaseous olefins, would try to decrease space velocity, since it was known from documents (9) and (14) that high conversion gave high yields of olefins, and from document (10) that conversion was increased by a decreased space velocity. Moreover, a space velocity of below 20 h⁻¹ was within the scope of the teaching of document (2).
- The subject-matter of Claim 1 consisted in a cracking process which had been optimized to achieve a high yield in C₃/C₄ olefins. The way to optimise for this purpose the various parameters of the process was, however, part of the common general knowledge as represented in documents (5), (6) and (8) to (10).
- Document (3) was also concerned with the production of propylene and butylene, since these

were the olefins used for alkylation as taught in documents (6) and (14). However, no inventive difference existed between the claimed process and that disclosed in document (3).

- The process of document (12) differed from the claimed one only in a slightly lower temperature, and a particular pressure and steam to feed ratio. These features were, however, obvious from the teaching of documents (5), (7) and (9).

IX. The Respondents rejected the Appellant's arguments and submitted in essence:

- that document (2) disclosed a dilute phase cracking process which could not be operated at space velocities corresponding to the dense phase system defined in present Claim 1;
- that it was not permissible for the assessment of inventive step to select unrelated parameters from several references in order to piece together the claimed subject-matter;
- that document (3) primarily concerned the production of gasoline where high yield of aromatics was important and - like document (2) - also operated at dilute phase conditions;
- that document (12) was irrelevant since it suggested different operating parameters and mentioned a considerably lower maximum yield of C₃/C₄ olefins than document (2).

X. The Appellant requested in writing that the decision

under appeal be set aside and that the European patent No. 0 305 720 be revoked.

The Respondents requested that the appeal be dismissed and that the patent be maintained according to the request submitted during oral proceedings.

Reasons for the Decision

1. *Amendments*

- 1.1 With the exception of the term "microspherical" which is not explicitly mentioned in the application as originally filed, amended Claim 1 is based on Claims 1 to 3, 5 and 8 of the application as originally filed. The Appellant never objected to the introduction of the term "microspherical", and the Board is of the opinion that it is credible from document (5) that, at the priority date of the patent in suit, a person skilled in the art would have expected a zeolite catalyst to be in the form of microspheres, in particular when it is used in a fluid cracking process (page 416, third full paragraph). The introduction of this term does not, therefore, add subject-matter extending beyond the content of the application as filed. The conditions of Article 123(2) EPC are therefore met.

All amendments made to Claim 1 during the oral proceedings before the Appeal Board are based on Claim 1 as granted. They do not, therefore, extend the protection conferred by the claim (Article 123(3) EPC).

The other amendment which was introduced during the opposition proceedings consists in a restriction of the

process of Claim 1 with respect to the feedstock to be treated and is supported by granted Claim 4.

Dependent Claims 2 to 5 correspond to Claims 4, 7, 9 and 10 as originally filed and to Claims 2, 5, 6 and 7 as granted.

Further, the amendments do not give rise to objections under Article 84 EPC.

In summary, the Board concludes that the claims as amended fulfill the requirements of Articles 123(2)(3) and 84 EPC.

- 1.2 According to decision G 4/92 (OJ EPO 1994, 149) of the Enlarged Board of Appeal, a decision should not be made against a party failing to attend oral proceedings if such a decision would be based on new facts first submitted at those oral proceedings. Being based on Claim 1 as granted, the amendments to Claim 1 made in the present case by the Respondents during the oral proceedings in the elected absence of the Appellant are not new facts within the meaning of G 4/92. Moreover, the amendments are all intended to overcome deficiencies under Article 123(3) EPC, a ground of appeal which had been raised by the Appellant in its statement of grounds of appeal. Consequently, the Appellant had to expect that the Respondents would try to overcome this objection by suitable amendments (see also T 133/92, not published in the OJ EPO, reasons for the decision No. 7). Therefore, the Appellant's right to be heard was not violated by rendering this decision in the Appellant's absence.

2. *Inventive step*

Novelty was not contested either in the opposition or appeal proceedings. The only point at issue is, therefore, whether or not the claimed subject-matter is based on an inventive step.

2.1 Technical background

The patent in suit relates to the production of gaseous olefins, in particular to the production of propylene ($C_{3=}$) and butylene ($C_{4=}$) from heavy petroleum hydrocarbon feedstock such as vacuum gas oil, residual oil and crude oil, by a catalytic conversion using a solid acidic catalyst (page 2, lines 3 to 11, page 3, lines 6 to 8).

According to the patent in suit, several processes for the manufacture of C_2 -to- C_4 -olefins from gasoline or vacuum gas oil have been proposed in the state of the art, inter alia one using ZSM-5 as the catalyst. These processes are, however, all said to be disadvantageous in so far as they require high cracking temperatures of 600 to 800°C and are not sufficiently selective so that the yields of $C_{3=}$ and $C_{4=}$ are poor (page 2, lines 17 to 25).

Therefore, the technical problem to be solved by the claimed subject-matter could be seen in providing a process which gives a higher yield of $C_{3=}$ and $C_{4=}$ in comparison to conventional catalytic cracking processes, even at a lower reaction temperature which ensures that expensive steel material for the apparatus is not necessary (page 3, lines 33 to 40).

2.2 Closest prior art

As already indicated, a number of documents relate to the production of C₃₌ and C₄₌ from petroleum hydrocarbons utilizing solid acidic conversion catalysts. According to document (2), amounts of 10 to 40% by volume of propylene and butylene can be obtained. These values unarguably correspond to about 6 to 24% by weight due to the specific gravity of propylene and butylene of about 0.6 g/cm³. These yields are mentioned for a process carried out at a reaction temperature in the range of 538 to 613°C (1000 to 1135°F) under dilute phase conditions, with contact times of about 0.2 to 5 seconds (column 2, lines 37 to 55), at a differential pressure of about 1.3 to 2.1 x 10⁵ Pa (5 to 15 p.s.i.g.) (column 2, lines 20 to 26), at a catalyst to oil ratio (C/O) of 5 to 25 by weight (column 2, lines 12 to 14) and in the presence of steam (column 2, lines 4 to 12). The catalyst is a commercially available zeolite or molecular sieve (column 1, lines 63 to 64). The applied feedstock and apparatus used were said not to be critical (column 1, lines 67 to 72), but virgin gas oil having a boiling range of between 316 to 593°C (600 to 1100°F) and a specific gravity of 28 API is mentioned as an example (column 2, lines 63 to 66), indicating a rather heavy hydrocarbon feed. The apparatus is a downflow reactor (Figure). The weight hourly space velocity WHSV can be obtained by the well-known correlation (see document (10), middle of page 115)

$$\text{WHSV (h}^{-1}\text{)} = 3600 / \text{contact time (sec)} \times \text{C/O.}$$

As agreed by the parties, the above values given in document (2) for contact times between 0.2 and 5 seconds and C/O ranging from 5 to 25, result in a WHSV ranging from 28.8 to 3600 h⁻¹ (see also decision of the Opposition Division, page 6, last paragraph). These

values as well as the downstream flow conditions in the reactor vessel (column 1, lines 54 to 57 and column 2, line 63 to column 3, line 5) indicate that the disclosed process is carried out under the dilute phase conditions mentioned above.

Therefore the Board considers document (2) as the most promising starting point for someone seeking a catalytic cracking process suitable for producing a gaseous product stream rich in propylene and butylene from a heavy petroleum hydrocarbon feedstock, in particular since - of all the documents on file - it mentions the highest yield of C₃₌ and C₄₌.

2.3 Technical problem and its solution

Considering that the range of the process temperature according to Claim 1 is not lower than but encompassed within that of the process of document (2), the technical problem to be solved as against this document (2) amounts simply to a further improvement of the C₃₌+C₄₌-yields.

The solution to this problem suggested by Claim 1 of the patent in suit is the combination of the following features within a catalytic cracking process:

1. a particular feedstock (comprising vacuum gas oil, residual oil or crude oil);
2. a particular catalyst (comprising pentasil shape selective molecular sieves or ultra stable hydrogen Y (USY) and inorganic oxides or clay as a matrix material);

3. a particular reactor (fluidised, moving bed or transfer line);
4. a particular process temperature (500 to 650°C);
5. a particular process pressure (1.5 x 10⁵ Pa and 3.0 x 10⁵ Pa);
6. a particular WHSV (0.2 to 20 hr⁻¹); and
7. a particular steam-to-feed ratio (0.05 to 1:1 by weight).

As is shown in the examples, under these conditions the process of the patent in suit gives C₃₌+C₄₌-yields ranging from about 30 to about 43% by weight. The Board concludes, therefore, that the above mentioned technical problem has plausibly been solved by the claimed process.

2.4 It remains to be decided whether, in view of the available prior art documents, it was obvious for someone skilled in the art to solve this technical problem by the means claimed.

2.5 The Appellant submitted that the values for the contact time and the C/O disclosed in document (2) were not strictly defined values but merely vague values of e.g. about 5 seconds contact time and about 25 C/O. Therefore, the resulting lower WHSV value was also not limited to 28.8 h⁻¹. A person skilled in the art would, therefore, understand document (2) as also teaching a WHSV of below 20 h⁻¹.

Moreover, if a person skilled in the art was to change

anything in the process of document (2) in order to obtain a product rich in gaseous olefins, he or she would try to increase the conversion as suggested in document (9) (see page 362, paragraph surrounding Table VIII), in document (14) (see page 112, right-hand column, lines 1 to 11) and in document (10) (see middle of page 115) by applying a low space velocity (document (10), loc. cit.). All other features of Claim 1 were usual in catalytic cracking processes.

2.6 The Board is not convinced by this line of argument for the following reasons:

2.6.1 Concerning the argument that document (2) covered WHSV values of below 20 h^{-1} , the Board agrees with the Respondents' submission that the term "about" cannot be given the meaning of a deviation of " $\pm 40\%$ ". In the Board's opinion, "about" merely means that deviations within the normal margin of error of measurement are covered. The Appellant did not provide evidence that a margin of error of $\pm 40\%$ would apply to the measuring of contact time, C/O or WHSV, nor does the Board see any reasons why that should be. The upper limit for the WHSV of the claimed process is, in the Board's judgment, therefore, clearly lower than the lowest value proposed in document (2).

2.6.2 Concerning the Respondent's second argument, the Board considers that it may be accepted that the feedstock mentioned in document (2) is comparable with that claimed, and that the temperature and pressure conditions as well as the C/O are within or overlapping those defined in present Claim 1. Also it may be accepted that the steam to feed ratio (3 to 45 pounds of steam per barrel of feed) mentioned in document (2)

(see column 2, lines 10 to 12) overlaps with the claimed range. Document (2) does not, however, contain any suggestion how modifications of the process conditions could yield a product still more rich in C_3+C_4 .

In particular, document (2), by merely mentioning zeolites in general as the catalyst and using a WHSV considerably higher than that of the process of Claim 1, does not suggest that a specific zeolite catalyst in combination with a substantially reduced WHSV could be suitable for this purpose.

2.6.3 The Board accepts the Appellant's submission that the state of the art, in particular documents (9), (10) and (14), contained several suggestions that a decrease in space velocity could enhance the low olefin yield of the respective processes disclosed in these citations. However, in the Board's opinion, none of these documents offers appropriate guidance how further to improve the C_3+C_4 -yield of document (2), if only for the reason that the latter is already considerably higher than the amounts of propylene and butylene mentioned (if at all) in citations (9), (10) and (14).

Thus document (9), a general review article from 1954, mentions on page 362 that "olefin contents of the C_3 and C_4 fractions vary over a wide range depending upon the catalyst, feed stock, and operating conditions". It goes on to say that normally a "high degree of unsaturation is favoured by the use of high-boiling feed, high temperature, low pressure and **low conversion**". However, high conversion - **up to a certain point** - gives increased yields of olefins, because "gas yield is enough higher to outweigh the decreased degree

of unsaturation" (page 362, first complete sentence after the table). Being silent about any definitions of the terms "low" and "high", this statement is, in the Board's opinion, rather vague and imprecise since it leaves a skilled reader in doubt as to the exact point to which conversion might be increased and whether such increase would actually increase the C₃₌ and C₄₌ fractions or merely produce more ethylene. Further, he is left in doubt about the other parameters influencing high olefin yield, in particular the feed stock and the catalyst.

Document (14) more clearly indicates that the yield of propylene and butylene increases with increasing reactor temperature and conversion (page 112, right-hand column, lines 1 to 11). However, Figure 4 on page 113, to which this statement refers, shows that a maximum amount of C₃₌ and C₄₌ can be obtained at 566°C (1050°F) and 90% conversion, the total yield being, however, about only 14% wt. Document (14) does not suggest that this yield could be considerably further increased, but indicates that at severe cracking conditions the ultimate result depends on two competing reactions, the one producing and the other consuming olefins (page 113, left-hand column, first full paragraph). Moreover, document (14) also realises that the results are further influenced by other parameters, such as reactor design, feedstock quality and catalyst type, without however giving any details of these (page 113, left-hand column, second full paragraph).

Document (10), a handbook on the subject of petroleum refining, also gives on page 115 a general overview of how conversion is interrelated with other operating variables. It is stated that high conversion results

from high reaction temperature, high C/O, high catalyst activity, high contact time, and low space velocity. Typical operating conditions for a fluidized bed reactor are given on page 116 in Table 7.2 with temperatures, pressures, WHSV and C/O overlapping the respective claimed ranges. It is however not indicated what kind of feedstock and catalyst have been used therein. Moreover, the Board accepts the Respondents' argument that footnote e below the table clearly shows that the total amount of the C₃ and C₄ fractions, including both saturated and unsaturated components, amounts to 30% by volume, or only 18% by weight.

Moreover, document (2) relates to a process carried out under dilute phase conditions in a downstream type reactor. As submitted by the Respondents, any substantial reduction of the space velocity in such a system would additionally require at least either procedural or constructional amendments which would influence the overall product quality and distribution of fractions in an unforeseen manner. This was not contested by the Appellant.

The Board, therefore, concludes that a skilled person would not find any hint in documents (9), (10) and (14) that decreasing the WHSV in the process of document (2) by a factor of at least 40% could considerably increase the propylene and butylene yield.

- 2.6.4 The Board also agrees with the Appellant that there exists prior art, i.e. documents (3), (6) and (12), which uses in a catalytic cracking process a catalyst of the claimed type. However, the total amount of the C₃ and C₄ fractions, where mentioned at all, is again much lower than that of the process of document (2), namely

about 9% by weight according to Tables 5 and 6 of document (6) and up to about 15% by weight according to the examples given in document (12) (see Example 3).

Document (3) is silent on such yields. It is in the first place concerned with gasoline production at low coke formation (column 2, lines 49 to 59) and teaches short contact times of up to 15 seconds, e.g. 2 to 15 seconds, and C/O of 6 to 25 for recycle and coker feed material for this purpose. It is said that operating space velocities are relatively high. A range for the WHSV of 10 to 300 h⁻¹ can be calculated from these figures (column 2, line 60 to column 3, line 2 and column 7, lines 48 to 60). Still lower WHSV are indicated in Figures I and II, which illustrate coke formation in relation to different catalysts, however at temperatures below 500°C and atmospheric pressure. It is also mentioned that operating conditions are employed which generally maximize the gasoline yield. Under some conditions production of high amounts of olefins for use in alkylation may, however, be desirable. The conditions to be selected for olefin production are not indicated. It is only mentioned that in this case no ZSM-5 type catalyst should be used (column 20, lines 10 to 18). Since ZSM-5 falls within the group of pentasil shape selective molecular sieves used in the claimed process, document (3) rather suggests using other types of catalyst for the production of olefins, thereby teaching away from the claimed subject-matter.

Document (6) is silent on any conditions of temperature, pressure, WHSV, C/O and steam and, therefore, would not have been considered by the skilled person looking for a solution to the technical

problem in question.

Document (12) in all its examples discloses process temperatures below 500°C without indication of a particular pressure. According to the general teaching of document (12), cracking temperatures of between 200 and 700°C (400 to 1300°F) may be employed under reduced atmospheric or superatmospheric pressure (column 10, lines 16 to 19). It is, however, stated that vacuum must be employed above 400°C (750°F) in order to avoid thermal cracking (column 10, lines 38 to 41) which is contrary to the corresponding requirement in Claim 1 of the patent in suit.

Therefore, the Board concludes that none of documents (3), (6) and (12) indicates how to improve the C₃₌+C₄₌₋ yield over that of document (2).

- 2.6.5 As citations (5), (7), (8), (11) and (13) do not mention the present technical problem, these citations would not have been considered by a skilled person seeking a solution to this problem.
- 2.7 The Board concludes therefore that, while the various parameters of the claimed process were known from different catalytic cracking processes, their particular combination as in the process of Claim 1 to obtain a product with enhanced C₃₌+C₄₌₋ yield was not hinted at in the prior art documents, either individually or in combination.
- 2.8 No other result is obtained if one starts either from document (3) or from document (12) as the closest prior art as suggested by the Appellant.

The problem to be solved as against these documents also consists in providing products with increased yields of $C_{3=}$ and $C_{4=}$. As indicated above, documents (3) and (12) do not give any hint as to the solution of this problem. The only document on file which proposes a solution is document (2). This document, however, by working at dilute phase conditions, including high space velocities WHSV ranging from 28.8 to 3600 hr^{-1} , leads one away from the claimed process.

2.9 All the other documents on file are less relevant to the issue of inventive step than those discussed above and do not provide any incentive for the claimed solution either.

3. The Board holds, therefore, that the cited prior art documents either alone or in combination do not render obvious the claimed solution of the present technical problem, and concludes that the process of Claim 1 is based on an inventive step within the meaning of Article 56 EPC.

Dependent Claims 2 to 5, which refer to preferred embodiments of Claim 1, are based on the same inventive concept and derive their patentability from that of Claim 1.

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 to maintain the patent with Claims 1 to 5 of the request as submitted during the oral proceedings and a description to be adapted thereto.

The Registrar:

The Chairman:

G. Rauh

P. Krasa