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**D E C I S I O N**  
**of 22 September 1999**

**Case Number:** T 0933/94 - 3.3.1

**Application Number:** 88200403.9

**Publication Number:** 0281208

**IPC:** C07C 2/24

**Language of the proceedings:** EN

**Title of invention:**

Process for two-stage catalytic conversion of an olefins-  
containing feed

**Patentee:**

Shell Internationale Research Maatschappij B.V.

**Opponent:**

Degussa-Hüls Aktiengesellschaft

**Headword:**

Two-stage catalytic process

**Relevant legal provisions:**

EPC Art. 123(2), (3), 56

**Keyword:**

"Transfer of an opposition (yes)"  
"Inventive step - main request (no)"  
"Auxiliary request (yes)"

**Decisions cited:**

G 0004/88, T 0686/91, T 0298/93, T 0219/83, T 0270/90,  
T 0181/82

**Catchword:**

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Boards of Appeal

Chambres de recours

**Case Number:** T 0933/94 - 3.3.1

**D E C I S I O N**  
**of the Technical Board of Appeal 3.3.1**  
**of 22 September 1999**

**Appellant:** Shell Internationale Research  
(Proprietor of the patent) Maatschappij B.V.  
Carel van Bylandtlaan 30  
2596 HR Den Haag (NL)

**Representative:** -

**Respondent:** Degussa-Hüls Aktiengesellschaft  
(Opponent) Patente und Marken  
Standort Marl  
Bau 1042/PB 15  
45764 Marl (DE)

**Representative:** -

**Decision under appeal:** Decision of the Opposition Division of the  
European Patent Office posted 17 October 1994  
revoking European patent No. 0 281 208 pursuant  
to Article 102(1) EPC.

**Composition of the Board:**

**Chairman:** A. J. Nuss  
**Members:** P. F. Ranguis  
J. P. B. Seitz

## Summary of Facts and Submissions

I. The Appellant (proprietor of the patent) lodged an appeal against the decision of the Opposition Division by which the European patent No. 281 208 was revoked in response to an opposition which had been filed against the patent as a whole on the grounds that the claimed subject-matter was not patentable under Article 54 or Article 56 EPC.

II. The decision was based on five requests filed at the oral proceedings before the Opposition Division, i.e. a main request and four auxiliary requests, said requests therefore superseding the claims as granted.

III. The Opposition Division held in particular that the feature "that at least part of the amount of metal(s) (X) have been incorporated into the (first stage) catalyst by means of ion exchange" contained in each of the claims 1 of the five requests, was not supported by the application as filed (Article 123(2) EPC).

In addition, the Opposition Division considered that this feature extended the protection conferred by the patent as granted (Article 123(3) EPC).

IV. At the oral proceedings held on 22 September 1999 before the Board, the Appellant abandoned the five requests on which the contested decision was based; he requested that the decision under appeal be set aside and,

- as a main request, that the patent be maintained in the form as granted and,

- as an auxiliary request, that the patent be maintained in an amended form, on the basis of the claims 1 to 9 submitted at the oral proceedings.

The Respondent (Opponent) requested that the Appeal be dismissed.

V. Claim 1 of the main and auxiliary request read as follows:

(i) Claim 1 of the main request:

"Process for two-stage catalytic conversion of an olefins-containing feed wherein the feed is contacted in a first stage under substantially non-oligomerizing conditions comprising a temperature from 20 to 150°C, with a catalyst comprising at least one metal (X) selected from the group consisting of metals from Groups 1a, 1b, 2a, 2b, 4b, 5b, 6b and 8 of the Periodic Table of the Elements and effluent from the first stage is contacted in the second stage under olefin oligomerization conditions at a temperature which is at least 50°C above the operating temperature of the first stage with a catalyst comprising at least one metal (Z) selected from the group consisting of metals from Groups 1b, 2a, 2b, 4b, 5b, 6b and 8 on a mordenite-type of crystalline trivalent metal (Q) silicate".

(ii) Claim 1 of the auxiliary request:

"Process for two-stage catalytic conversion of an olefins-containing feed wherein the feed is contacted in a first stage under substantially non-oligomerizing conditions comprising a temperature from 20 to 150°C

with a catalyst comprising at least one metal (X) selected from the group consisting of metals from Groups 1a, 1b, 2a, 2b, 4b, 5b, 6b and 8 of the Periodic Table of the Elements on a modernite-type carrier and effluent from the first stage is contacted in the second stage under olefin oligomerization conditions at a temperature which is at least 50°C above the operating temperature of the first stage with a catalyst comprising at least one metal (Z) selected from the group consisting of metals from Groups 1b, 2a, 2b, 4b, 5b, 6b and 8 on a mordenite-type of crystalline trivalent metal (Q) silicate".

VI. In the appeal proceedings, the Respondent only maintained his opposition on the ground of lack of inventive step and supported his submissions on the basis of the documents:

D1: US-A-2 828 347

D3: G. Wendt et al.: "Dimerization of n-butenes on nickel mordenite catalysts", React. kinet. Catal. Lett., vol. 31, No. 2 (1986) pages 383-388

In addition to those documents, three other documents which had been cited by the Respondent in the course of the opposition proceedings were also considered in the appeal proceedings:

D2: GB-A-1 135 938

D4: EP-B-90 569

D5: Ashim K. Ghosh et al.: "A Fourier-Transform

Infrared Spectral Study of Propene Reactions on Acidic zeolites", Journal. of Catalysis **100**, (1986) pages 185-195

Furthermore, in support of the inventive step of the patent in suit, the Appellant cited in the appeal proceedings six other documents:

D6: US-A-2 381 198

D7: US 2 581 228

D8: Kirk and Othmer, vol. 4, Pages 358-360, 3rd edition (1978)

D9: Kirk and Othmer, vol. 4, Pages 716-719, 4rd edition (1992)

D10: Kirk and Othmer, vol. 10 Pages 196-197 (1953)

D11: Catalysis by Metals, G. C. Bond, page 285 (1962)

VII. In the course of the written proceedings and during the oral proceedings, the Appellant argued essentially that for both the main and auxiliary request the technical problem to be solved is the provision of a process for the catalytic conversion of olefin to dimers and oligomers in which olefinic impure feeds such as those available from refineries can be used without adversely affecting catalyst stability.

The Appellant submitted that document D1 disclosed a process for polymerizing olefin which comprises contacting a polymerisable olefin feed under

polymerizing conditions, at a temperature preferably in the range from 25°C to 150°C, with a catalyst comprising nickel chloride or nickel bromide supported on an amorphous silica-alumina carrier. He further argued that the first step set out in the example, consisting of passing the olefin-containing feed stream over a copper-on-pumice catalyst in order to remove acetylene, butadiene and sulphur compounds, before it was contacted at a temperature of 71.1°C (160°F) with a nickel chloride catalyst, is an hydrogenation step which takes place at 150-200°C.

In support of this assertion, the Appellant cited the documents D6, D7, D8, D9, D10 and D11. He argued essentially that documents D6 and D7, cited as references in D1, suggested passing the feed stream through a bed of hydrogenation catalyst, e.g. nickel on kieselguhr or silica gel at elevated temperature (200°C) (see page 2, right hand column, lines 29 to 30 of D6 and column 9, lines 6 to 7 of D7). Documents D8 and D9 disclosed the removal of dienes impurities from olefin-containing feed by selective catalytic hydrogenation. Document D10 showed that the yields of light gases from various cracking processes contain always a few amounts of hydrogen and document D11 showed that copper-on-pumice is one of the catalysts used in the hydrogenation of acetylene, indicating a temperature range of 150°C to 195°C.

He further submitted that the submission of the Respondent according to which the first step of D1 would be carried out at room temperature was pure speculation and that the Respondent had adduced no evidence for his allegation.



According to the Appellant, an alternative way for interpreting the example in document D1 would be that the temperature of the first stage is the same as the second stage i.e. 71.1°C.

Regarding document D3 which relates to the dimerization of n-butenes on nickel mordenite catalysts, the Appellant admitted that this document disclosed the second stage of claim 1 of the patent in suit, with the reservation that the olefinic feedstock of D3 did not contain any dienes and therefore could not suggest to the person skilled in the art as to how the technical problem addressed in the patent in suit might be solved.

Regarding the documents D2, D4 and D5, The Appellant considered that they did not relate to the same kind of catalysts and did not address the technical problem mentioned above.

The Appellant further pointed out that an important feature of the present process is the temperature difference between the first and second stages - the first stage being carried out at a temperature from 20 to 150°C and the second stage being carried out at a temperature higher (at least 50°C higher) than the first stage.

In conclusion, the Appellant submitted that D1 and D3 would not have led the person skilled in the art to develop a process as claimed in the patent in suit, nor was there any reason to believe that such a process would be advantageous, resulting in good rates of olefin conversion over extended periods of time. In

support of that, he relied upon experiments 1 to 5 (2 and 4 being comparative examples) set out in the description.

In support of the inventive step of the auxiliary request, the Appellant submitted essentially the same arguments.

VIII. Regarding the main request, the Respondent maintained that the subject-matter of the present claims did not involve an inventive step in view of documents D1 and D3 taken in combination.

In particular, he disputed that the first step of the Example 1 of D1 (passing the feed gas over copper-on-pumice catalyst) was carried out at high temperature. In his view, the said step was carried out at room temperature for the following reasons:

- (a) the fact that no temperature is mentioned for this step means simply that the reaction is carried out at room temperature,
- (b) The said reaction cannot be made under hydrogenation conditions because no hydrogen is necessarily present in the gas composition (D1 mentions carbon monoxide but not hydrogen). In that context, documents D6 and D7 are not relevant as they relate to the removal of sulphur compounds and not olefinic compounds, by hydrogenation catalysts.
- (c) It is not credible that the removal of acetylene, dienes and sulphur compounds on copper-on-pumice

be made at 150°C-200°C as this step does not lead to the oligomerization of ethylene or propylene. In this connection, the Respondent pointed out that copper was mentioned in the two stages of the claimed subject matter. In the second stage of the claimed subject matter, at the temperature (150°C to 330°C) recommended by the patent in suit (page 3, line 11), the oligomerization occurs. Therefore, the removal of dienes can only be made at a lower temperature. The same conclusion applies to the first step of Example 1 of D1.

Regarding inventive step, The Respondent considered that Example 1 of D1 addressed the same technical problem as that of the patent in suit i.e. oligomerizing ethylene and propylene while removing, in a first step, the catalyst poisons such as dienes. Furthermore, it would have been obvious for one skilled in the art to use the mordenite catalyst according to D3 in the process of Example 1 of D1 and as a result develop a process as now claimed.

The Respondent no longer contested the inventive step of the patent in suit on the basis of documents D2, D4 and D5.

In support of the lack of inventive step of the auxiliary request, the Appellant submitted essentially the same arguments.

IX. At the end of the oral proceedings the decision of the Board was given orally.

## Reasons for the Decision

1. The appeal is admissible.

### *Identity of the Respondent/Opponent*

2. The opposition was filed by Hüls AG, Marl, Germany. The Respondent informed the Board on 10 August 1999 that Hüls AG had merged with Degussa AG forming the new firm Degussa-Hüls AG. A notarized certificate was provided. The Board concludes, according to the decision G 4/88 (OJ EPO 1989, 480), that the opposition should be treated as transferred to Degussa-Hüls AG. Therefore, the Board considers that this firm is entitled to take over the Hüls AG's rights in the present proceedings.

### *Main request*

3. After examination of the cited prior art documents, the Board has reached the conclusion that the subject-matter as defined in the claims as granted is novel. Since novelty was no longer disputed, it is not necessary to give reasons for this finding.
4. The remaining issue to be dealt with is whether the subject-matter of the claims as granted involves an inventive step.
  - 4.1 The Board considers, in agreement with the parties, that the closest state of the art with respect to the two-stage catalytic conversion of an olefin-containing feed according to present claim 1 is the disclosure of document D1. Indeed, according to numerous decisions

(see in particular T 686/91, point 4 of the reasons; T 298/93, point 2.2.2 of the reasons; both not published in OJ EPO), the "closest prior art" is a prior art document disclosing subject-matter aiming at the same objective as the claimed invention and having the most relevant technical features in common.

Document D1, in particular its example, is the sole disclosure related to a two-stage catalytic process for polymerization of olefins stemming from a cracked gas, wherein the first stage consists in removing the poisons such as butadiene, acetylene or sulphur compounds on copper-on-pumice and the second stage in polymerizing the olefins on a nickel chloride or oxide catalyst supported on a silica-alumina carrier at 160°F (71.1°C) as set out column 3, lines 25 to 54.

- 4.2 An important question to be answered with regard to the disclosure of document D1 is, in the Board's view, that of the temperature at which the removal of dienes, acetylene and sulphur compounds is carried out.

The parties have made contradictory assertions regarding the temperature at which the feed gas is passed over copper-on-pumice catalyst in the example of D1. None of them have submitted clear evidence which would have enabled the Board to base its decision on absolute conviction.

On the one hand, it is a general principle that the patent proprietor (Appellant) is given the benefit of the doubt in case of contrary assertions regarding facts. The burden of proof that the feed gas is passed over the copper-on-pumice at room temperature lay with

the Respondent. However, the Respondent has submitted no relevant information to support his assertion. Therefore, the objection is dismissed (T 219/83, OJ EPO 1986, 211, in particular point 12 of the reasons).

On the other hand, the Appellant has submitted two alternative ways regarding the temperature at which the feed gas is passed over copper-on-pumice: either at hydrogenation temperature (150 to 200°C) or at 71.1°C (160°F), which is the temperature at which the polymerization occurs.

As a general principle, the Boards decide the issues before them on the basis adduced by the parties in applying a balance of probabilities (T 270/90, OJ EPO 1993, in particular point 2.1 of the reasons). In the present case, it is to be decided whether the temperature of 150 to 200°C is more likely to be true than the 71.1°C temperature, or the opposite.

In support of the temperature of 150 to 200°C, the Appellant cites the documents D6 to D11. However none of them mentions the hydrogenation of dienes by copper (only D11 relates to hydrogenation of acetylene). The Board notes furthermore that copper is one of the metals (supported on crystalline modernite) used for the second step of the patent in suit. The temperature of dimerization is between 150°C and 330°C (see page 3, line 11 of the patent in suit). By contrast, the temperature of the first step in D1 is such that no dimerization occurs. Although it is likely that the crystalline modernite support is involved to some degree in the catalytic reaction and that it cannot be directly compared to an amorphous support such as

pumice, it still remains that the Appellant has submitted nothing relevant showing that, although polymerisation using copper on crystalline modernite occurs between 150°C and 300°C, this does not happen when copper is used alone or on another support, nor has the Appellant shown that in the presence of copper-on-pumice at 150-200°C no dimerization occurs. Moreover, on page 2, lines 51 to 56, the patent in suit mentions that the first stage (comprising the use of copper, be it alone or supported), must preferably be carried out between 30°C and 100°C to limit the oligomerization. It does not seem credible that for the same reaction the temperature, in the patent in suit, must be limited to 30-100°C to avoid any oligomerization, while it may be of 150-200°C in D1.

On the contrary, the temperature of 71.1°C is the more likely one to have been used in both steps of the example of D1.

Therefore, in accordance with one of the interpretations offered by the Appellant, the Board is satisfied that the temperature at which in D1 the feed gas is passed over the copper-on-pumice catalyst is 71.1°C.

- 4.3 In the next step, the technical problem which the invention addresses in the light of the closest prior art is to be determined.

In view of the closest prior art, i.e. document D1, the technical problem underlying the patent in suit consists in providing a **further** process for two-stage catalytic conversion of an olefins-containing feed

without affecting catalytic stability. In that context, the comparative tests 2 and 4 mentioned in the Table of the patent in suit can be of no use as they do not relate to a process such as disclosed in D1 (T 181/82, OJ EPO 1984, 401, in particular point 5 of the reasons).

This problem is solved by the process according to claim 1 (see point V(i) above) of the patent as granted.

4.4 The further step which consists in determining whether the problem is solved for all the solutions falling within the scope of the claimed subject-matter need not be examined in view of the considerations made in point 4.5 below.

4.5 It remains to be decided whether or not the proposed solution to the problem underlying the patent in suit is obvious in view of the cited prior art.

Document D1, i.e. the closest prior art (see point 4.1 above), teaches as a general description a process for polymerizing olefins which comprises contacting a polymerisable olefin feed under polymerising conditions with a nickel chloride-silica-alumina "at temperatures which vary within a rather wide range but generally are not much lower than 0°C nor appreciably above about 225°C. It is preferred to operate within the range of about 25°C to 150°C. Temperatures in the neighbourhood of 100°C seem to be superior since at such level polymerisation is accelerated without undue side reactions." (see column 2, line 70 to column 3, line 5). The specific example describes a two-stages



catalytic process for conversion of olefins in which it has been established that both steps are carried out at 71.1°C (see point 4.2 above). The first stage is made on a copper-on-pumice catalyst to remove acetylene, butadiene and sulphur compounds and the second (polymerisation) stage is made on nickel chloride-silica-alumina catalyst or nickel oxide silica-alumina catalyst as a comparative experiment. However, it is within the ambit of the person skilled in the art, in view of the general description, to vary the temperature of polymerisation between 0°C and 225°C or at least between 25°C and 150°C.

Nevertheless, in document D1, the nickel halide-silica-alumina catalyst (or the nickel oxide-silica-alumina catalyst, see comparative test) used in the polymerisation stage is structurally amorphous because D1 (see column 1, lines 25 to 29) refers directly to D6 (US 2 381 198) and D7 (US 2 581 228) which disclose silica gel containing minor amounts of alumina (see, in particular, column 2, lines 34 to 41 of D7, column 2, lines 18 to 22 of D6 and column 2, lines 37 to 44 of D1), while, in the patent in suit, the catalyst used in the polymerisation stage is, amongst others, a nickel modernite-type of crystalline trivalent metal (Q) silicate.

However, when trying to solve the above stated technical problem, it would have been obvious for the man skilled in the art to replace, in the example of D1, the nickel oxide or nickel chloride supported on silica-alumina catalyst by a nickel modernite catalyst such as disclosed in D3 and carry out the polymerization of the purified olefinic feed at 100°C

(373°K) because the latter teaches those specific (and sole) conditions for the dimerisation of pure butene (without dienes or acetylene). Although the Board admits that this does not lead completely to the claimed subject matter, because the difference of temperature between both stages would be only about 30°C or so, instead of the claimed difference of at least 50°C between the two stages, there is no reason to believe that the selection of the temperature range was a purposive selection and not an arbitrary selection. Prima facie, in the Board's view, it would have been within the ambit of the person skilled in the art to optimize the temperature of the second stage as a matter of routine. Additionally, the Board notes that document D4 teaches that mordenite-type catalysts can be used between 20°C and 180°C (see column 5, lines 48 to 60), confirming that, as for the polymerisation catalysts of D1, the temperature of polymerisation with mordenite-type catalysts may also vary in a wide range.

The Appellant has submitted in the course of the proceedings that "it was an important feature of the process of the present invention that the **first stage** (in which diene removal is effected) is carried out at a temperature in the range from **20°C to 150°C** and **the second stage** (in which olefin oligomerization occurs) is carried out at a temperature which is **at least 50°C** above the operating temperature of the first stage" (emphasis added by the Appellant), (see page 5, paragraph (ii) of the statement of grounds of appeal).

However, nothing relevant was submitted to substantiate that the stated difference in temperature between the two stages is an essential feature. Although it is

plausible that this is the case when both catalysts comprise a modernite-type carrier (see auxiliary request), the Board is not convinced that this feature is critical for any carrier which might be used in the first stage. In other terms, in the present situation, the conditions of temperature are arbitrary and therefore irrelevant for the assessment of inventive step.

The Board concludes that the man skilled in the art would have been led by the teaching of D1 taken in combination with that of D3 to achieve a two-stage catalytic process involving, in a first stage, the removal of undesirable compounds such as dienes by a suitable catalyst and, in a second stage, involving the polymerisation of the olefin-containing feed by a nickel on modernite-type carrier, the choice of the temperature being either meaningless or at best a question of simple optimization that the skilled person would have easily determined.

For this reason, claim 1 of the main request does not involve an inventive step in the sense of Article 56 EPC.

*Auxiliary request*

5. The present auxiliary request differs from the main one in that claim 1 was amended to specify that the metal (X) is supported "on a modernite-type carrier" (see point V(ii) above).
6. In the Board's judgment, this amendment neither contravenes Article 123(2) EPC (see page 3, lines 22 to

31; page 6, lines 18 to 24 of the application as filed), nor Article 123(3) EPC as the protection conferred is not extended thereby. This amendment was not objected to by the Respondent either.

7. As for the main request, the Board considers that D1 is the closest prior art and that the technical problem underlying the patent in suit consists in providing a **further** process for two-stage catalytic conversion of an olefins-containing feed without affecting catalytic stability.

The Board is satisfied, in view of tests 1, 3 and 5 contained in the patent in suit, that the difference of temperature of 50°C between the first and the second stage is an essential one for all the solutions encompassed by claim 1. This finding has not been challenged by the Appellant. Therefore, the Board accepts that the process as defined in claim 1 solves the above stated technical problem.

It remains to be decided whether or not the proposed solution to the problem underlying the patent in suit in the form of the auxiliary request is obvious in view of the cited state of the art.

Document D1, i.e. the closest prior art (see point 4.1 above), teaches a two-stage catalytic process for conversion of olefins. In Example 1, for which it has been established that both steps are carried out at 71.1°C (see point 4.2 above), the first stage is made on a copper-on-pumice catalyst to remove acetylene, butadiene and sulphur compounds and the second (polymerisation) stage is made on nickel chloride-

silica-alumina catalyst whereby for this step a wide temperature range between 0°C and 225°C (column 2, lines 70 to column 3, line 1) may be envisaged.

When questioned by the Board at the oral proceedings, the parties did not contest that pumice is a naturally occurring foam glass and that glass is an inorganic product of fusion that has cooled to a rigid condition without crystallizing. Therefore pumice is structurally amorphous, while the modernite-type carrier is crystalline.

Furthermore, in document D1, the nickel chloride-silica-alumina catalyst (or the nickel oxide-silica-alumina catalyst, see comparative test) used in the polymerisation stage is structurally amorphous because D1 (see column 1, lines 25 to 29) refers directly to D6 (US 2 381 198) and D7 (US 2 581 228) which disclose silica gel containing minor amounts of alumina (see, in particular, column 2, lines 34 to 41 of D7, col 2, lines 18 to 22 of D6 and column 2, lines 37 to 44 of D1).

When dealing with the main request, where the catalyst in the first step is not required to have a modernite-type carrier, the Board has already indicated that it would have been obvious to the man skilled in the art to use in the process known from D1 the nickel modernite catalyst of D3 for carrying out the second stage. However, in the process now claimed the catalyst used in the first stage cannot be one such as described in D1 and document D3 teaches nothing about the removal of dienes, nor do documents D2, D4 or D5 contain any relevant information related to this removal. D2

discloses a process involving a pure isobutylene, D4 mentions simply the removal of butadiene before the polymerisation reaction and D5 discloses a process involving substantially pure propene.

The prior art provides, therefore, no incentive for the person skilled in the art, first, to replace the copper-on-pumice carrier by a modernite-type carrier in the first stage and, furthermore, to achieve the two stages under specific temperature conditions (first stage from 20 to 150°C, and second stage, at least 50°C above the operating temperature of the first stage), both features in combination leading to a non obvious further process.

The Board concludes that it was not obvious to propose a process for two-stage catalytic conversion of olefins-containing feed such as defined in claim 1 of the auxiliary request and, therefore, its subject-matter involves an inventive step within the meaning of Articles 52(1) and 56 EPC.

For the same reasons, the Board concludes that the subject-matter of dependent claims 2 to 9 involves an inventive step.

**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 in amended form, namely:

**Claims:** 1 to 9

**Description:** pages 2 to 5

submitted as the auxiliary request at the oral proceedings on 22 September 1999.

The Registrar

The Chairman

E. Görgmaier

A. Nuss