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**Datasheet for the decision
of 15 November 2011**

Case Number: T 2013/07 - 3.3.07

Application Number: 99303973.4

Publication Number: 1053789

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B01J 23/78, C07C 17/152,
C07C 17/156, C07C 19/045

Language of the proceedings: EN

Title of invention:
Catalyst and oxychlorination process using it

Patent Proprietors:
Ineos Technologies (Vinyls) Limited

Opponents:
ALSBMARLE Netherlands B.V.
BASF SE

Headword:
-

Relevant legal provisions:
EPC Art. 56

Keyword:
"Inventive step - main and first auxiliary request (no)"
"Admissibility - second auxiliary request (no)"

Decisions cited:
-

Catchword:
-



Case Number: T 2013/07 - 3.3.07

DECISION
of the Technical Board of Appeal 3.3.07
of 15 November 2011

Appellants: BASF SE
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Decision under appeal: **Decision of the Opposition Division of the European Patent Office posted 16 November 2007 rejecting the opposition filed against European patent No. 1053789 pursuant to Article 102(2) EPC 1973.**

Composition of the Board:

Chairman: J. Riolo
Members: D. Semino
D. T. Keeling

Summary of Facts and Submissions

I. The appeal of the opponents lies against the decision of the Opposition Division announced at the oral proceedings on 8 November 2007 to reject the oppositions against European Patent 1 053 789. The granted patent comprised 10 claims, independent claims 1 and 9 reading as follows:

"1. A catalyst comprising a carrier and catalytically active material comprising copper supported thereon, the copper being present in an amount of 1-12 wt % on the dry catalyst, wherein the catalyst is in the form of a hollow cylinder having the following dimensions:-

$$4.0 \leq D_e \leq 7.0$$

$$2.0 \leq D_i \leq 2.8$$

$$6.1 \leq L \leq 6.9$$

$$2.0 \leq D_e/D_i \leq 2.5$$

wherein D_e is the external diameter (mm), D_i is the internal diameter (mm) and L is the length (mm), respectively of the hollow cylinder."

"9. Use of the catalyst of any of claims 1 to 8 in the oxychlorination of hydrocarbons."

Dependent product claim 2 included a limitation in all four ranges of dimensions and dependent use claim 10 related to the specific oxychlorination of ethylene to 1,2-dichloroethane.

II. Two notices of opposition had been filed against the granted patent requesting revocation of the patent on the grounds of lack of novelty and lack of inventive step in accordance with Article 100(a) EPC. The

oppositions were *inter alia* supported by the following documents:

D1: US-A-4 366 093

D2: US-A-5 166 120

III. The decision under appeal can be summarised as follows:

- (a) The catalyst in the form of a hollow cylinder of granted claim 1 was novel with respect to the disclosure of D1, because D1 did not disclose a length of the hollow cylinder in the range 6.1 to 6.9 mm.

- (b) D1, which was the closest prior art, dissuaded the skilled person from increasing the length to more than 6 mm, as supported by the comparative examples which showed that the hot spot temperature became unfavourably high if a height greater than 6 mm was selected. Therefore, the skilled person would not expect that both the pressure drop and the hot spot temperature might be improved by increasing the length beyond the range disclosed in D1. Moreover, even if the skilled person, starting from D1, ought to decrease the pressure drop at any cost, he would not be in a one-way street situation, since there were at least three possibilities, namely changing the length, the external diameter or the internal diameter/wall thickness. In addition, even if the skilled person chose to increase the length beyond the range of D1 in view of D2, this would not necessarily lead to a value within the range 6.1 to 6.9 mm. For those reasons, the skilled person

did not find any motivation in the prior art to increase the length to a value of 6.1 to 6.9 mm in order to provide a catalyst with lower pressure drop and hot spot temperature. Therefore, the catalyst of granted claim 1 was inventive.

- IV. The opponents 02 (appellants) appealed that decision.
- V. In a communication sent in preparation of the oral proceedings the Board summarised the main issues raised by the parties.
- VI. With letter of 11 October 2011 the patent proprietors (respondents) filed a set of 9 claims together with an amended description as first auxiliary request. In claim 1 according to that request the ranges of D_e , D_i , L and D_e/D_i had been limited to 4.5-5.5, 2.0-2.6, 6.2-6.6 and 2.1-2.3 respectively, according to claim 2 as granted.
- VII. With letter of 13 October 2011 the appellants maintained their previous objections of lack of novelty and lack of inventive step and further elaborated on them.
- VIII. Oral proceedings were held on 15 November 2011 in the announced absence of the opponents 01 (party as of right to the proceedings). During the oral proceedings the respondents submitted a second auxiliary request, which included a single claim, reading as follows:

"The use of a catalyst comprising a carrier and catalytically active material comprising copper supported thereon, the copper being present in an

amount of 1-12 wt % on the dry catalyst, wherein the catalyst is in the form of a hollow cylinder having the following dimensions

$$4.5 \leq D_e \leq 5.5$$

$$2.0 \leq D_i \leq 2.6$$

$$6.2 \leq L \leq 6.6$$

$$2.1 \leq D_e/D_i \leq 2.3$$

in increasing the selectivity of the catalytic oxychlorination of ethylene to 1,2-dichloroethane which comprises reacting ethylene, oxygen and hydrogen chloride in a fixed bed reactor in the presence of the catalyst."

IX. The arguments of the appellants (opponents 02), as far as relevant to the present decision, can be summarised as follows:

Inventiveness of claim 1 according to the main request and to the first auxiliary request

- (a) D1 was correctly considered as the closest prior art by the Opposition Division and disclosed a catalyst which differed from the claimed one only marginally in its length.
- (b) No other effect could be acknowledged to that difference other than a reduction in pressure drop when the catalyst was used in tubular reactors. The tests in the patent were not able to convincingly show any other improvement in performance, since they did not reproduce the examples of D1 and they were not realistic. In particular, the incredibly large excess in ethylene in those tests explained the high

conversion in hydrogen chloride and rendered the results irrelevant. Moreover, it was unclear how the flowrates of the reactants were changed in order to give the desired oxygen excess and no information was given on the measurement errors and on the relevance of variations in selectivity in the fourth significant digit. In any case, it was not credible that a catalyst with lower surface area and higher void fraction could have a better activity and the results showed that the results strongly depended on the operating conditions. The problem solved was therefore the provision of a catalyst which allowed a reduction in pressure drop.

- (c) According to document D2 that problem could be solved by increasing the length of the hollow cylinder, its external diameter or both. Since D1 taught the disadvantages of increasing the external diameter and suggested to keep the ratio between the length and the external diameter as close as possible to one and not exceeding the value of 2, the increase in length remained the only possible solution to the posed problem. The comparative examples in D1 confirmed the disadvantages of values of external diameter larger than 6 mm and of a ratio between the length and the external diameter greater than 2. The skilled person, while increasing the length and keeping the ratio between the length and the external diameter as close as possible to one, would inevitable fall in the range of lengths of granted claim 1 without exercising any inventive activity.

- (d) The same arguments applied to claim 1 of the first auxiliary request, where the ranges of dimensions had only be slightly limited.

Admissibility of the second auxiliary request

- (e) The single claim of the second auxiliary request filed during the oral proceedings was significantly different from granted use claim 9 since it specified an increase in selectivity which did not appear in the granted claims. That feature was not clear, had no basis in the application as filed and was not supported by the examples. Since the request was undoubtedly late filed and opened up a large number of new issues, it should not be admitted into the proceedings.

- X. The arguments of the respondents (patent proprietors), as far as relevant to the present decision, can be summarised as follows:

Inventiveness of claim 1 according to the main request and to the first auxiliary request

- (a) The claimed catalyst differed from the one disclosed in the closest prior art document D1 in the length of the hollow cylinder.
- (b) The problem solved by means of that feature was the provision of a catalyst which satisfied in use in oxychlorination reactors the requirements of lower pressure drop, better heat exchange together with better activity and selectivity. The examples

in the patent showed that the problem was solved by the claimed catalyst. They were realistic, since it was normal practice to use a large ethylene excess in industrial reactors using oxygen instead of air, as confirmed in D2, and they were clear, since the skilled person could understand without difficulties how to modify the oxygen flowrate to obtain the desired small variations in oxygen excess. Moreover, example B in the patent was a reasonable reproduction of example 1 of D1, since no significant variations in the results could be expected in view of the minimal variations in the dimensions and there was no proof that the measurements were not reliable. The results showed not only a clear decrease in pressure drop and hot spot temperature, but a consistent improvement in conversion and selectivity, so that in view of them and on the balance of probabilities the problem posed in the patent in suit had to be considered as solved over the whole breadth of the claim.

- (c) There was no hint in the available prior art that the length of the catalyst of D1 should be increased in order to solve the posed problem. The claimed catalyst involved therefore the required inventive step.

- (d) The same arguments applied a *fortiori* for the catalyst of the first auxiliary request, which was even more remote from the one of D1 and restricted the invention to the most preferred ranges of catalyst dimensions, for which the claimed effects were indisputably proven.

Admissibility of the second auxiliary request

(e) The second auxiliary request was late filed, but was to be seen as a reaction to the latest submissions of the appellants, which were filed a month before the oral proceedings. Its single claim was clear, had a basis in paragraphs [0010], [0029] and [0034] of the original application, was supported by the examples in the patent and did not introduce any new issue in the proceedings. Therefore, the request had to be admitted.

XI. The appellants (opponents 02) requested that the decision under appeal be set aside and the European patent be revoked.

XII. The respondents (patent proprietors) requested that the appeal be dismissed or in the alternative that the patent be maintained according to the claims of the first auxiliary request filed by letter of 11 October 2011 or of the second auxiliary request filed during the oral proceedings.

Reasons for the Decision

1. The appeal is admissible.

Main request

2. *Inventiveness of claim 1*

- 2.1 Document D1, which like the patent in suit concerns a catalyst in the form of a hollow cylinder for the oxychlorination of hydrocarbons (see claim 1 and paragraph [0001] of the patent in suit together with D1 claim 1, column 2, lines 64-67 and column 4, lines 2-6), has been considered both in the decision under appeal and in the main submissions of the parties as the closest prior art. The Board has no reason to deviate from this choice.

- 2.1.1 D1 discloses a catalyst having a cylindrical shape, the external diameter being from 3 to 6 mm, the internal diameter being at least 1.0 mm, the thickness of the wall being at most 1.5 mm and the length (referred to as height in D1) being from 3 to 6 mm (claim 1). Preferred dimensions are 4 to 5 mm for the external diameter, 1.5 to 2.5 mm for the internal diameter, 1.0 to 1.5 mm for the wall thickness and 3 to 6 mm for the length (column 1, line 67 to column 2, line 2). The catalyst has a carrier and a catalytically active material (column 2, lines 58-64), which is typically a copper halide when the catalyst is used for oxychlorination (column 2, lines 64-67).

- 2.1.2 The catalysts of the examples of D1 (examples 1 to 4) contain 18 % by weight of cupric chloride (column 4,

lines 20-24), which corresponds to 8.17 % by weight copper. The catalyst of example 1 has a cylindrical shape with an external diameter of 5 mm, an internal diameter of 2.2 mm (therefore a ratio external/internal diameter of 2.27) and a length of 5 mm (table bridging pages 3 and 4). The catalysts of the examples of D1 were tested in an externally cooled nickel tube 1200 mm long with an internal diameter of 26.3 mm to which 21.6 Nl/h of ethylene, 40 Nl/h of hydrogen chloride and 57 Nl/h of air were fed (column 4, lines 25-33 and 45-49).

- 2.2 The catalyst of granted claim 1 of the patent in suit differs therefore from the catalyst of D1 (in particular of example 1 of D1) only in the length of the hollow cylinder, which is in the range 6.1 to 6.9 mm according to granted claim 1.
- 2.3 The problem to be solved according to the patent in suit is to provide a catalyst for effective use in oxychlorination reactions which satisfies the requirements of lower pressure drop of the catalytic bed, better heat exchange and good effectiveness (paragraph [0015]). The same problem is, however, addressed by D1 (see column 1, lines 8-14 and 42-44; column 2, lines 64-67), so that the question arises whether a further improvement in pressure drop, heat exchange and effectiveness (intended as related to the activity and selectivity of the catalyst) can be acknowledged for the claimed catalyst with respect to the catalyst of D1.
- 2.4 In order to establish which improvements have been supported by evidence it is crucial to take into

account the tests available on file, in particular the examples in the patent.

- 2.4.1 Catalyst A and catalyst B, for which tests are reported in the patent, are meant to be representative of the claimed catalyst and of a catalyst according to D1 (paragraph [0031] in the patent). The values of the external diameter, internal diameter and length are 4.90 mm, 2.25 mm and 6.35 mm for catalyst A and 4.90 mm, 2.25 mm and 5.00 mm for catalyst B respectively.
- 2.4.2 The reactor used in the tests is an externally cooled nickel tube 8 mm long with an internal diameter of 27.75 mm to which 5200 Nl/h of ethylene, 600 Nl/h of oxygen, 2300 Nl/h of hydrogen chloride and 1000 Nl/h of nitrogen were fed at a pressure of 6 barg and with a coolant temperature of 220°C (paragraph [0030]). Those conditions were intended to be truly representative of an industrial reactor (paragraph [0029]).
- 2.4.3 Three trials were accomplished for each catalyst (table 3 in the patent) with an excess of oxygen versus hydrogen chloride of 0.8%, 3.0% and 6.8% for catalyst A (trials A1, A2 and A3) and of 2.0%, 4.3% and 6.6% for catalyst B (trials B1, B2 and B3). The pressure drop was 1.6 bar in the tests on catalyst A and 1.9 bar in the tests on catalyst B. The hot spot temperature was 253°C in the tests on catalyst A and 256°C in the tests on catalyst B. The hydrogen chloride conversion was 98.4, 99.9 and 100 in tests A1, A2 and A3 and 98.5, 99.8 and 100 in tests B1, B2 and B3 respectively (all values are in % mol). The selectivity to 1,2-dichloroethane was 98.28, 98.63 and 98.68 in tests A1,

A2 and A3 and 98.19, 98.40 and 98.55 in tests B1, B2 and B3 respectively (all values are in % mol).

2.5 The Board cannot agree with the appellants that those tests are unrealistic and unclear and that the values of selectivity are not reliable. Large excess in one of the reactants (in the present case ethylene) is a common possibility in reactive processes, as is confirmed specifically for the oxychlorination of ethylene with pure oxygen in the analysis of the prior art in D2 (column 1, lines 51-56). Oxygen excess of 0.8 to 6.8 mol% versus hydrogen chloride (minimum and maximum values in the tests on catalysts A and B) with a flowrate of 2300 Nl/h hydrogen chloride corresponds to an oxygen flowrate of 580 to 614 Nl/h, so that the specification of an oxygen flowrate of 600 Nl/h (paragraph [0030]), albeit imprecise, can be taken as an indication of an average value. As to the values of selectivity to 1,2-dichloroethane, no evidence has been provided by the appellants that it is not possible to obtain the level of precision given in the results in the patent, so that those values have to be appreciated as they are.

2.6 However, the examples in the patent do not provide a reproduction of the examples of D1. The dimensions of the hollow cylinder of catalyst B in the patent are not the same as the ones of the closest example of D1 (example 1 with an external diameter of 5.0 instead of 4.9 mm in catalyst B and an internal diameter of 2.2 mm instead of 2.25 mm in catalyst B) and the conditions under which the catalysts are tested are completely different (see points 2.1.2 and 2.4.2 above) in terms of reactor size and operating conditions (small

flowrates, use of air and almost stoichiometric quantities in D1, larger flowrates, use of oxygen and large excess of ethylene in the patent).

2.7 Moreover, the results in the patent show a strong sensitivity of the results to the operating conditions, since small changes in the excess of oxygen lead to relevant changes of the hydrogen chloride conversion and in the selectivity to 1,2-dichloroethane (the values for trial B3 with catalyst B according to D1 are better than those of trial A1 with catalyst A according to the patent and the operating conditions differ only in a small variation in the oxygen flowrate).

2.8 Under such circumstances, the presence of a single example (with slightly different trials) with a single combination of dimensions and a single set of operating conditions, which are not a reproduction of the examples of the prior art, which example is compared with a catalyst, which does not correspond to the catalyst of the closest example of the prior art, cannot be sufficient to render credible that a simultaneous improvement in pressure loss, hot spot temperature, activity and selectivity can be achieved over the whole breadth of the claim when the claimed catalyst is used. The Board considers that over the balance of probabilities it cannot be concluded that such a combined improvement has been proven.

2.9 As to the size of the compared catalysts, while it is true that the difference in external diameter between catalyst B in the patent and example 1 of D1 is only 0.1 mm (in addition to a difference of 0.05 mm in internal diameter), a similar difference is all that

distinguishes the claimed catalyst from the catalyst of D1 (the length in D1 goes up to 6 mm and the minimal length in the patent is 6.1 mm), so that if it is accepted that no significant difference in the results exists between catalyst B in the patent and example 1 of D1 (as the respondents alleged), then the same holds true for the claimed catalyst with respect to the catalyst of D1.

2.10 The only effect which can be acknowledged with respect to D1 is the decrease in pressure loss, which takes place if the length of the hollow cylinder is increased, all other dimensions remaining the same. This is supported not only by the examples in the patent, but also by those of D1, where all the pairs of examples and comparative examples which differ only in the length (example 1 and comparative example 4, examples 2 and 3, example 2 and comparative example 3 in the table overlapping pages 3 and 4) consistently show that an increase in length corresponds to a reduction in pressure drop. Moreover, it is in agreement with physical considerations, since when only the length is increased, the loading of the catalytic bed decreases, with higher void fraction and lower surface per unit volume, which clearly results in a reduction of pressure drop.

2.11 The problem effectively solved with respect to D1 is therefore the provision of a catalyst for effective use in oxychlorination reactions with lower pressure drop within the catalytic bed.

2.12 D2 in the context of a catalyst for the same reaction (selective oxychlorination of ethylene, column 1, lines

6-10) and with the same hollow cylindrical shape and similar dimensions (column 2, lines 19-25) discloses that it is generally known that the pressure drop can be reduced across a bed of annular catalytic bodies by using larger external diameters and/or lengths of the catalytic bodies.

2.13 The Board considers that this passage of D2 suggests three solutions to the problem of decreasing the pressure drop (increase the external diameter, increase the length or increase both) and that none of these solutions is inventive in view of the disclosure in D2. Moreover, D1 indicates clearly that the external diameter should not exceed 6 mm in order to avoid disadvantages in catalytic activity, selectivity and heat conductivity (column 3, lines 42-44) and that the ratio of the length to the external diameter of the hollow cylinder should be close to one and not exceed 2 (column 3, lines 36-42). Therefore, the skilled person, starting from the catalyst of D1 and addressing the problem of decreasing the pressure drop, would in view of the disclosure of D2 and the limitations given in D1 increase the length of hollow cylinder, thereby obtaining a catalyst according to granted claim 1 without exercising any inventive activity.

2.14 For these reasons, the catalyst of granted claim 1 does not involve an inventive step.

First auxiliary request

3. *Inventiveness of claim 1*

3.1 Claim 1 according to the first auxiliary request differs from granted claim 1 only in that preferred ranges are specified for the external diameter, internal diameter, length and ratio of external to internal diameter.

3.2 However, these limitations do not affect the comparison with D1 as the closest prior art, since the difference with respect to it (in particular to its example 1) remains still only the length of the hollow cylinder. The amendments, therefore, do not change the analysis of inventive step with respect to D1, which leads to the conclusion that the catalyst of claim 1 of the first auxiliary request is not inventive for the same reasons as detailed for the main request (see paragraphs 2.1 to 2.14 above).

Second auxiliary request

4. *Admissibility*

4.1 The second auxiliary request filed during the oral proceedings before the Board included a single claim in the form of a use claim which, in spite of partially using the wording of original dependent claims 10 (as far as the use of the catalyst in the specific oxychlorination of ethylene to 1,2-dichloroethane) and 2 (as far as the definition of the catalyst is concerned), introduces a use of the catalyst "in increasing the selectivity" of the specific

oxychlorination reaction of granted claim 10, which never appeared in the claims during prosecution of the case.

4.2 The filing of the second auxiliary request cannot be seen as a legitimate reaction to the communication of the Board, in which only the points raised by the parties were summarised, nor to the letter dated 13 October 2011 of the appellants, which did not introduce any new grounds or evidence, but only further elaborated the arguments related to lack of inventive step, nor to the discussion during the oral proceedings, which addressed the critical point of inventive step by taking into account the facts, evidence and arguments which had been in the proceedings since the filing of the oppositions.

4.3 Moreover, the single claim of the second auxiliary request opens up a number of new issues, which have never been addressed by the parties and the Board during the proceedings and would require therefore a new analysis of the case, including lack of clarity (it is not defined with respect to what an increase in selectivity takes place), lack of basis in the original application (paragraphs [0010], [0029] and [0034] which have been indicated as a basis by the respondents do not contain the wording of the new use and refer either to the prior art or to specific tests) and sufficiency of disclosure (how could the skilled person know which catalysts provide an increase in selectivity and under what process conditions), without apparently solving the critical issue of inventive step with respect to D1 (which discloses the same reaction and aims at a good selectivity).

4.4 Under these circumstances and by exercising its discretion under Article 13 of the Rules of Procedure of the Boards of Appeal, the Board finds it appropriate not to admit the second auxiliary request into the proceedings.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.
2. The patent is revoked.

The Registrar

The Chairman

S. Fabiani

J. Riolo