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File Number: T 23/90 - 3.3.1

Application No.: 83 104 238.7

Publication No.: 0 093 434

Title of invention: Preparation of polyalkylene polyamines

Classification: C07C 85/06

D E C I S I O N  
of 18 September 1991

Proprietor of the patent: UNION CARBIDE CORPORATION

Opponent: Hüls Aktiengesellschaft

Headword: Polyamines/Union Carbide

EPC Articles 54 and 56

Keyword: "Novelty (yes)" - "Inventive step (yes)"

Headnote



Case Number : T 23/90 - 3.3.1

**D E C I S I O N**  
of the Technical Board of Appeal 3.3.1  
of 18 September 1991

**Appellant :**  
(Proprietor of the patent)

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**Respondent(s) :**  
(Opponent)

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**Decision under appeal :**

Decision of Opposition Division of the European  
Patent Office posted on 2 November 1989 revoking  
European patent No. 0 093 434 pursuant to  
Article 102(1) EPC.

**Composition of the Board :**

**Chairman :** R. Andrews  
**Members :** J.M. Jonk  
J.A. Stephens-Ofner

Summary of Facts and Submissions

I. The grant of the European patent No. 0 093 434 in respect of European patent application No. 83 104 238.7 was announced on 13 August 1986 (cf. Bulletin 86/33).

II. A Notice of Opposition was filed on 28 April 1987 requesting the revocation of the patent on the grounds of lack of novelty and inventive step, and insufficient disclosure. The opposition was supported by several documents including:

- (5) US-A-3 121 115,
- (6) US-A-4 036 881 (= (4) DE-A-2 624 109),
- (8) US-A-4 103 087, and
- (11) US-A-4 044 053 (= (3) DE-A-2 624 135)

which are relevant to the present decision.

III. By a decision dated 2 November 1989 the Opposition Division revoked the present European patent.

The Opposition Division held that the subject-matter of Claim 1 submitted to the Opposition Division on 5 January 1989, reading:

"A process for producing predominantly linearly extended polyalkylene polyamines comprising:

- (a) contacting (i) an alkylenediamine with; (ii) a difunctional hydroxy alkylene compound selected from the group consisting of alkylene glycols and alkanolamines; (iii) in the presence of a catalytically effective amount of a phosphorus acid or acid derivative compound;

- (b) removing water as it is formed during the reaction;  
and
- (c) recovering the polyalkylene polyamines from the relatively anhydrous reaction mixture."

did not involve an inventive step. The reasons for this finding were that the process according to this Claim 1 differed from the prior art, as represented by document (6) or (11), in that the water of reaction was removed. However, this feature would have been obvious to the skilled person because it was recommended in documents (5) and (8). Moreover, the improvement of the conversion rate was the logical consequence of the shift of equilibrium of a reversible reaction in accordance with the law of mass action.

- IV. A Notice of Appeal was submitted against this decision on 2 January 1990 and the appeal fee was paid on the same date.

A Statement of Grounds of Appeal was submitted on 13 February 1990.

- V. The Appellant contended in his written submissions and during oral proceedings held on 18 September 1991 that the removal of the water as it was formed during the production of the predominantly linearly extended polyalkylene polyamines was not obvious to the skilled person, because both documents (6) and (11), representing the closest state of the art, disclosed that it was not critical to control the amount of water of reaction and that it was desirable to retain the water in the reaction zone. According to Example IV of document (6) water was even added.

Documents (5) and (8), recommending the removal of water of reaction, were both related to a different process, namely the preparation of tertiary substituted nitrogen compounds. Moreover, this prior art was discussed in the documents (6) and (11), showing that the proprietors of these patents had specific knowledge thereof and, nevertheless, had a different view about the control of the water of reaction.

The Appellant also argued that the present reaction was irreversible, so that it could not be expected that the absence of water would affect the reaction efficiency.

During the oral proceedings the Appellant submitted a new set of 12 claims (auxiliary request), Claim 1 being identical with Claim 1 of the main request with the exception of the following additional feature under (a):

"(iv) at temperatures from 250°C to 350°C and at a pressure sufficient to provide a reaction mixture in a liquid state";

VI. The Respondent argued that the subject-matter of the opposed patent lacked novelty in the light of the disclosure in document (6) that it was not critical to control the amount of water of reaction present during the heating of reactants and catalyst, such as by removal thereof as it was formed, because this statement implied the performance of experiments wherein the water of reaction was in fact removed. In this connection he referred to a passage in GRUR Int. 1991, Volume 6, page 449, left column, last whole paragraph, which is related to the non-published decision T 28/89 of 18 September 1989.

Regarding inventive step the Respondent disputed Appellant's allegation that the present reaction would be irreversible by referring to

(13) Sammlung Göschen Bd. 698/698a - Allgemeine und Physikalische Chemie, 2. Teil - de Gruyter (Berlin 1956), Seite 105, Abschnitt 72.

From the disclosure of this document the Respondent concluded that the present catalytic reaction was an equilibrium reaction so that the law of mass action would be valid.

VII. The Appellant requested that the decision under appeal be set aside and the patent be maintained on the basis of claims submitted to the Opposition Division on 5 January 1989 (main request) or on the basis of claims submitted in the course of oral proceedings (auxiliary request).

The Respondent requested that the appeal be dismissed.

VIII. At the conclusion of the oral proceedings, the Board's decision to maintain the patent on the basis of the main request of the Appellant was announced.

#### Reasons for the Decision

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

2.1 There are no objections to the present claims under Article 123 EPC. The amendment under (b) of Claim 1 is based on page 7, line 54 and the amendment under (c) on page 7, line 61 of the printed patent specification (cf. also page 17, lines 11 and 25 of the originally filed patent application).

2.2 The first issue to be dealt with is, whether the subject-matter of Claim 1 is novel.

2.2.1 In accordance with the established jurisprudence of the Boards of Appeal, this issue has to be decided on the basis of whether document (6) as a whole makes available to the skilled person as a technical teaching the subject-matter for which protection is sought, i.e. a process for producing predominantly linearly extended polyalkylene polyamines in which the water of reaction is removed as it is formed.

2.2.2 Document (6) discloses the preparation of predominantly non-cyclic polyalkylene polyamines by reacting an alkyleneamine with an alkanolamine in the presence of a catalytically effective amount of a phosphorus-containing substance (cf. column 2, lines 41 to 63). Furthermore, it contains the following statement at column 5, line 65 to column 6, line 3:

"It is not critical to control the amount of water of reaction present during the heating of reactants and catalyst, such as by removal thereof as it is formed. Usually, we prefer to retain the water in the reaction mass during recovery of the predominantly non-cyclic polyalkylene polyamines."

2.2.3 The Respondent contended that the claimed process lacked novelty because this statement, teaching that the removal

of water of reaction was not critical, implied that the removal of the water has been tried.

However, in the Board's view, this argument fails because the opinion indicated in this statement is not necessarily based on actual comparative experiments involving the same process parameters save for the removal of water, but may be based on other grounds such as purely theoretical considerations or particular knowledge derived from reactions of closely related compounds.

Therefore, in the Board's judgement, the claimed process has not been made available to the skilled person as a technical teaching (cf. also T 310/88 of 23 July 1990, Supplement to OJ 6/1991, pages 21 and 22). The subject-matter of Claim 1 is, consequently, novel.

In the unpublished decision T 28/89 of 18 September 1989, it was held that a certain passage in a prior art document implied that the two possible nozzle orientations in a pulsed liquid-liquid extraction column had, at least, already been tried. However, the facts of the present case are different insofar as the clear intention of the above-quoted passage from document (6) was to emphasise the contrast between the process of this document and that of document (5) (referred to in column 2, lines 23 to 33) which required the continuous removal of water.

2.3 The next issue to be dealt with is whether the subject-matter of Claim 1 involves an inventive step.

2.3.1 After consideration of the prior art documents cited during the proceedings, the Board finds that documents (6) and (11) represent the closest state of the art.



Documents (6) and (11) are both related to a process for preparing predominantly, non-cyclic polyalkylene polyamines. According to document (6) an alkyeneamine is reacted with an alkanolamine whereas according to document (11) the conversion of the alkyleneamine is carried out with a diol compound (cf. (6), column 2, lines 41 to 52 and (11), column 2, lines 33 to 43). The reactions are carried out in the presence of a catalytically effective amount of a phosphorus-containing substance, such as boron phosphate (cf. (6), the paragraph bridging columns 4 and 5 and (11), column 5, lines 17 to 27).

However, the conversion rates of the reactants to the predominantly linearly polyalkylene polyamines were unsatisfactory.

2.3.2 Therefore, in the light of this closest prior art, the technical problem underlying the subject patent can be seen in providing a process whereby the conversion rate to the desired linearly extended polyalkylene polyamines is increased (cf. also page 1, lines 22 to 24 and lines 62 to 64 of the printed patent specification).

2.3.3 According to Claim 1, this technical problem is solved by removing water as it is formed during the reaction.

In view of the undisputed test results indicated in the examples and the comparative examples (cf. particularly Tables 2 and 4) the Board is satisfied that the above technical problem is credibly solved.

2.3.4 It remains to be decided whether, in view of the technical problem to be solved, the requirement of inventive step is met by the claimed process.

2.3.5 Referring to document (13) the Respondent contended that the present reaction was reversible, so that the removal of water of reaction would inevitably lead to higher yields of the desired products.

However, document (13) only concerns the common general knowledge that catalysts already in small amounts affect the speed of both the forward and backward reaction in the same way and do not influence the equilibrium state. Therefore, this document does not provide the skilled person with any information as to whether the present reaction would be reversible in the sense that the desired reaction product readily reacts with water to generate the starting compounds.

Contrary to Respondents allegation it is indicated in the published patent specification that the present reaction is irreversible and that the effect of the removal of reaction water probably results in the reversible formation of a catalytically active phosphorus ester not appreciated by the prior art (cf. page 2, lines 35 to 64).

In the light of the conflict of credible evidence, which the Board is in no position to resolve of its own motion (Article 114(1) EPC), the Board finds that the Respondent has failed to discharge the onus of proving the facts he alleges, so that the argument he based on those facts cannot be accepted.

2.3.6 As previously mentioned, document (6) discloses the preparation of predominantly non-cyclic polyalkyleneamines by reacting alkyleneamines with alkanolamines in the presence of catalytic amounts of phosphorus-containing substances. In view of the statement referred to in paragraph 2.2.2 above and the fact that in all the

examples the water of reaction is retained in the reaction mixture and in Example IV water is added to the reaction mixture, the Board considers that the disclosure of this document leads away from the present process.

The same conclusion must be drawn from the disclosure of document (11), which only differs from that of document (6) by the use of a diol instead of an aminoalkanol (cf. section 2.3.1 above, second paragraph), because it contains exactly the same statement about the desirability of retaining the water of reaction in the reaction mixture (cf. column 6, lines 18 to 24) and because it discloses only examples whereby the reaction is carried out in an autoclave.

Therefore, these two documents do not give any incentive to the skilled person to remove the reaction water as it is formed.

- 2.3.7 Document (5) relates to a process for aminoalkylating an amine or phenol comprising heating of the alkylatable compound with an aminoalkanol in the presence of an acid condensing agent such as a phosphoric acid compound (cf. column 1, lines 62 to 67 and column 2, lines 6 to 21). The water of reaction is preferably removed as it is formed (cf. column 2, lines 64 to 70). The essential teaching of this document is concerned with the discovery that, since only a catalytic amount of the acid condensing agent is required to effect the reaction, the aminoalkylated product is obtained as a free base. Thus a neutralising step is no longer required (cf. column 1, line 70 to column 2, line 5 and column 5, lines 3 to 13). However, document (5) does not contain any indication that the alkylatable amine could be an alkylene-diamine, let alone that the disclosed process could be used for the preparation of linearly extended polyalkylene polyamines.

Therefore, in the Board's judgement, a skilled person faced with the problem of improving the process for the preparation of linearly extended polyalkylene polyamines according to document (6) or (11) would have disregarded the teaching of this document.

Moreover, this finding of the Board is confirmed by the fact that the closest prior art, namely documents (6) and (11), contain a discussion of document (5) including the indication that the process is carried out with continuous water removal (cf. (6), column 2, lines 23 to 38 and (11), column 1. line 56 to column 2, line 22). This means, that although the skilled persons involved in the development of the processes according to documents (6) and (11) had specific knowledge of document (5), they still expressed a preference for retaining water in the reaction mixture, i.e. the complete opposite of the present process (cf. paragraph 2.3.6 above).

2.3.8 Document (8) is related to a further development of the prior art process of document (5) and discloses the reaction of an amine having one labile hydrogen with a tertiary aminoalkanol in the presence of aluminium phosphate and, particularly, the production of di-(N,N-disubstituted amino) alkanes (cf. column 2, lines 6 to 37, and Claim 1). This prior art process differs essentially from the known process of document (5) in that the aminoalkanol starting compound is restricted to a monovalent tertiary aminoalkanol falling outside the scope of present Claim 1 and to the use of a particular catalyst. This means that the prior art of document (8) is still less relevant than that of document (5) discussed above and would, therefore, be of no assistance to the skilled person seeking a solution to the technical problem underlying the disputed patent.

2.3.9 Consequently, in the Board's judgement, the proposed solution to the technical problem underlying the patent in suit is inventive. Thus, Claim 1 of the main request is allowable.

Dependent Claims 2 to 12, which relate to preferred embodiments of the process according to Claim 1, are likewise allowable.

3. Since the claims according to the main request are allowable, there is no need to consider the auxiliary request.

Order


For these reasons, it is decided that:

1. The decision under appeal is set aside.
2. The case is remitted to the Opposition Division with the order to maintain the patent on the basis of the main request of the Appellant.

The Registrar:

  
E. Gorgmaier

The Chairman

  
R. Andrews