DECISION of 16 March 2000

Case Number: T 0907/97 - 3.3.3
Application Number: 91906281.0
Publication Number: 0477376
IPC: C08G 18/16

Language of the proceedings: EN

Title of invention: Process for producing polyurethane

Applicant: DAICEL CHEMICAL INDUSTRIES, LTD.

Opponent: -

Headword: -

Relevant legal provisions: EPC Art. 56

Keyword: "Inventive step (yes) - unobvious solution"

Decisions cited: -

Catchword: -
Case Number: T 0907/97 - 3.3.3

DECISION
of the Technical Board of Appeal 3.3.3
of 16 March 2000

Appellant: DAICEL CHEMICAL INDUSTRIES, LTD.
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Decision under appeal: Decision of the Examining Division of the European Patent Office posted 4 April 1997 refusing European patent application No. 91 906 281.0 pursuant to Article 97(1) EPC.

Composition of the Board:
Chairman: C. Gérardin
Members: P. Kitzmantel
V. Di Cerbo
Summary of Facts and Submissions

I. This appeal, which was filed on 16 June 1997, lies against the decision of the Examining Division dated 4 April 1997, refusing European patent application No. 91 906 281.0 filed on 19 March 1991 in the name of DAICEL CHEMICAL INDUSTRIES, LTD., and published under No. 0 477 376. The appeal fee was paid together with the Notice of Appeal and the Statement of Grounds of Appeal was filed on 13 August 1997.

II. The decision under appeal was based on two sets of each six claims of a main request and an auxiliary request, both filed with a submission dated 13 February 1997. Independent Claims 1, 5 and 6 of the main request read as follows:

"1. A process for producing a polyurethane which comprises

- preparing a dialkyl carbonate without using phosgene,

- reacting the carbonate with a diamine to give a urethane compound,

- thermally decomposing the urethane compound to give a diisocyanate, said diisocyanate containing chlorine in an amount of 10 ppm or below, and

- reacting the diisocyanate with a polyol to produce the polyurethane, said reaction being activated by the addition of a protonic acid and/or a Lewis
acid."

"5. A polyurethane obtainable according to the process of any one of claims 1 to 4."

"6. A coating composition comprising the polyurethane according to claim 5 and a pigment."

The further claims 2 to 4 were dependent on Claim 1.

The claims of the auxiliary request differed from those according to the main request by the insertion into the introductory portion of Claim 1 after "A process for producing a polyurethane" of the statement "which does not contain phosgene, hydrogen chloride or a compound having a carbamoyl or carboxyl group, ...".

III. The appealed decision held that the subject-matter of the main and of the auxiliary requests was obvious over document

D1: EP-A-0 323 514 (the decision refers to the wrong number EP-A-0 323 524),

because the use, in the preparation of polyurethanes, of acid catalysts was known from documents

D7: Saunders/Frisch, "Polyurethanes, Chemistry and Technology", pages 160 to 173; R.E.Kieger, Florida 1987, and

IV. In the course of the written phase of the appeal proceedings the Appellant filed a number of requests all of which were abandoned at the oral proceedings on 16 March 2000. During these oral proceedings the following set of four claims was submitted as the Appellant's sole request:

"1. A process for producing a polyurethane comprising the steps of

- preparing a dialkyl carbonate without using phosgene,

- reacting the carbonate with a diamine to give a urethane compound,

- thermally decomposing the urethane compound to give a diisocyanate, said diisocyanate containing chlorine in an amount of 1 ppm or below, and

- reacting the diisocyanate with a polyol to produce the polyurethane, said reaction being activated by the addition of a protonic acid selected from nitric acid, sulfuric acid, phosphoric acid, phosphorous acid, saturated organic acids having 1 to 18 carbon atoms, unsaturated acids having 3 to 18 carbon atoms, alkyl- and alkenyl-substituted derivatives of aromatic organic acids with the alkyl or alkenyl substituent having 1 to 18 carbon atoms, polybasic acids having 2 to 18 carbon atoms and partial esters thereof, alkylsulfuric acids having 1 to 18 carbon atoms, alkenyl sulfuric acids having 2 to 18 carbon atoms, alkylphenylsulfuric acids having 6 to 24 carbon
atoms, phosphinic and phosphonic acids each having an alkyl group having 1 to 18 carbon atoms, an alkenyl group having 2 to 18 carbon atoms or an alkylphenyl group having 6 to 24 carbon atoms, and phosphites and phosphates each having an alkyl group having 1 to 18 carbon atoms, an alkenyl group having 2 to 18 carbon atoms or an alkylphenyl group having 6 to 24 carbon atoms."

"2. The process of claim 1, wherein said dialkyl carbonate is prepared from carbon monoxide, oxygen and an alkanol."

"3. The process of claim 1, wherein said dialkyl carbonate is prepared by preparing propylene [spelling error "proplyene" corrected] carbonate from propylene oxide and carbon dioxide and reacting the formed carbonate with an alkanol."

"4. The process of claim 1, wherein said dialkyl carbonate is prepared from an alkyl nitrite and carbon monoxide."

In the Appellant's view the subject-matter of Claim 1 was not obvious over D1, (i) because this document contained no information regarding the conditions of polyurethane formation from the low reactive, low chlorine diisocyanates, with whose preparation it was concerned, and (ii) because documents D7 and D8 were totally silent on the protonic acids to be used as catalysts according to present Claim 1.

V. The Appellant requested that the decision under appeal be set aside and a patent be granted on the basis of
Claims 1 to 4 submitted during the oral proceedings.

**Reasons for the Decision**

1. The appeal is admissible.

2. *Amendments (Article 123(2) EPC)*

Claim 1 is based on original Claim 1 and on the statements on page 33, line 24 to page 34, line 6 as well as on page 35, line 22 to page 37, line 7 of the original application.

The requirement of Article 123(2) EPC is therefore complied with.

3. *Novelty*

Claim 1 of document D1 relates to a process for the preparation of isocyanate compounds, which comprises (1) the first stage reaction of reacting a diamine with dimethyl carbonate to synthesize a corresponding urethane compound, and (2) the second stage reaction of thermally decomposing this urethane compound in a high boiling solvent under reduced pressure and in the presence of a suitable catalyst. According to page 1, lines 12 to 19 this method for the preparation of isocyanate compounds was developed in order to avoid the use of phosgene, a highly toxic compound used for the preparation of isocyanate compounds according to the most conventional preparation technique.
While D1, thus, discloses the preparation of diisocyanates by the route specified in present Claim 1, it is silent on the conditions to be employed when these diisocyanates are reacted with polyols to polyurethanes. On page 27, lines 10 to 12 D1 only mentions that "(t)his diisocyanate is valuable as the starting material for the production of polyurethanes, medicines, agricultural chemicals and the like."

The subject-matter of Claim 1 and of Claims 2 to 4, which are appended thereto, is thus novel over D1.

4. Invention step

4.1 Problem and solution

The problem underlying the subject-matter of Claim 1 resides in the concretisation of the reaction conditions, including the choice of appropriate catalysts, to be applied in the manufacture of polyurethanes from the diisocyanates prepared according to D1.

According to the application in suit this problem is solved by the use of specific protonic acid catalysts as recited in Claim 1.

The data in Table 2 (page 78 of the original application) show that this problem is effectively solved by the use of several such protonic acids (cf. Examples 6 to 13, pages 68, line 20 to page 72, line 5 of the original application).

4.2 Obviousness
4.2.1 The Section of D7 relied upon by the Examining Division relates to the catalysis of the isocyanate-hydroxyl reaction.

The only protonic acid disclosed therein is hydrogen chloride (cf. page 161, lines 6 to 7 from bottom; page 162, Tables XXIII and XXIV). Since none of the many other compounds envisaged in this document as catalysts for the isocyanate-hydroxyl reaction is a protonic acid (cf. Tables XXV, XXVIII to XXX and XXXII on pages 163 to 169 and 171), no conclusion as to the effectivity of protonic acids in general as catalysts for the isocyanate-hydroxyl reaction can be drawn from the disclosure of this document.

4.2.2 Similarly D8, the third document considered in the decision under appeal, identifies a large number of compounds which may be used as catalysts for the isocyanate-hydroxyl reaction, but does not disclose a single protonic acid (cf. Tables II to IX and XI on pages 207 to 209).

4.2.3 The cited prior art cannot, therefore, suggest the use of protonic acids, other than hydrochloric acid, as catalysts for the isocyanate-hydroxyl reaction.

4.2.4 Moreover, the statement in point 4.6 of the decision under appeal, namely that "(t)he catalytic action of mineral- or organic carboxylic acid-catalysts in the polyurethane forming reaction is to be regarded as being derivable from the prior art ..." - in this generality - is open to doubt. Contrastingly, it is set out in Section 3.4.2 of the Polyurethane Handbook (G. Oertel, Hansa Verlag (1985), pages 96 to 97) that some
Brönstedt- and also Lewis-acids may even retard the proton transfer to the isocyanate groups and, thus, the formation of the urethane units. Such statement in a textbook, reflecting thus common general knowledge, would be a disincentive to consider a solution to the above defined problem along these lines.

4.2.5 The subject-matter of present Claim 1, thus, complies with the requirement of Article 56 EPC.

4.2.6 The same conclusion applies a fortiori to the subject-matter of the dependent Claims 2 to 4.

5. Although the present set of claims overcomes the objection which led to the refusal of the application, a patent cannot be granted at this stage in view of the extensive adaptation of the description called for. For that purpose only the Board avails itself of the discretion stipulated in Article 111(2) EPC and remits the case to the Examining Division.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the Examining Division with the order to grant a patent on the basis of the set of claims submitted during the oral proceedings and after any necessary adaptation of the description.
The Registrar: 

E. Görgmaier

The Chairman:

C. Gérardin