DECISION of 28 September 2000

Case Number: T 1215/97 - 3.3.3
Application Number: 93119982.2
Publication Number: 0601502
IPC: C08G 64/30

Language of the proceedings: EN

Title of invention: Process for producing (co)polycarbonate

Applicant: DAICEL CHEMICAL INDUSTRIES, LTD.

Opponent: -

Headword: -

Relevant legal provisions: EPC Art. 54(1), (2), 111(1)

Keyword: "Novelty - prior disclosure - implicit features (no)"

Decisions cited: -

Catchword: -
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DECISION
of the Technical Board of Appeal 3.3.3
of 28 September 2000

Appellant:
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Decision under appeal:
Decision of the Examining Division of the
European Patent Office dated and issued in
writing on 15 July 1997 refusing European patent
application No. 93 119 982.2 pursuant to
Article 97(1) EPC.

Composition of the Board:
Chairman: C. Gérardin
Members: R. Young
A. Lindqvist
Summary of Facts and Submissions

I. European patent application No. 93 119 982.2, filed on 10 December 1993 claiming JP priorities of 11 December 1992 (JP 331421/92) and 21 October 1993 (JP 263420/93), respectively, and published under No. 0 601 602, was refused by a decision of the Examining Division dated and issued in writing on 15 July 1997. The decision was based on a set of Claims 1 to 14 filed on 19 November 1996. Claim 1 read as follows:

"1. A process for producing a (co)polycarbonate which comprises:

conducting a melt-polycondensation of a dihydroxy compound and a carbonic diester as monomers in the presence of a transesterification catalyst in a tank reactor, while removing a monohydroxy compound formed by the melt-polycondensation, thereby obtaining a reaction mixture comprising a prepolymer in a molten state (the first step),

cooling the molten reaction mixture obtained in the first step to solidify it (the second step), and

conducting a melt-polycondensation of the reaction mixture comprising the prepolymer in a horizontal polycondensation reactor, thereby obtaining a reaction mixture comprising a high-molecular-weight (co)polycarbonate (the third step),

wherein the prepolymer obtained in the first step has a terminal hydroxyl group content of 50 molar % or below based on all the terminal groups of the prepolymer."
Claims 2 to 14 were dependent claims directed to elaborations of the process according to Claim 1.

II. According to the decision, the application did not meet the requirements of Article 52(1) EPC, because the subject-matter of Claim 1 was not novel in the sense of Article 54(1) and (2) EPC, in the light of the disclosure of:


In particular, D1 disclosed, in Preparative Example 1 (see pages 11 and 12), a process for producing a polycarbonate comprising the following steps:

- conducting a melt-polycondensation of bisphenol A and diphenyl carbonate (mole ratio 1 : 1.023) in the presence of boric acid, tetramethylammonium hydroxide and sodium hydroxide in a tank reactor, while removing phenol formed by the melt-polycondensation, thereby forming a prepolymer in a molten state (first step);

- passing the formed prepolymer through a die in a nitrogen atmosphere into a strand, followed by cutting into pellets using a cutter. The preformed prepolymer has an intrinsic viscosity of 0.32 dl/g as measured at 20°C in methylene chloride (second step);

- conducting a melt-polycondensation of the formed prepolymer into a double-screw stirring polymerizer, thereby obtaining a polymer having an intrinsic viscosity of 0.45 dl/g (third step).
All the features of present Claim 1, except the solidification of the prepolymer obtained in the first step and a content of terminal hydroxyl groups of 50 molar % or below based on all the terminal groups of the prepolymer were therefore explicitly disclosed in D1. However these above two other features were implicitly disclosed to the skilled reader of D1:

- The solidification of the prepolymer was self-evident since the cutting into pellets meant that the molten reaction mixture obtained in the first step had been solidified.

- The terminal groups of the polycarbonate prepolymer were either hydroxyl or ester groups, which corresponded to the unreacted part of a bisphenol A or of a carbonic diester. Since in Preparative Example 1 of D1 a mole ratio bisphenol A to diphenyl carbonate of 1 : 1.023 was used, it was obvious according to a basic chemical knowledge that statistically the prepolymer obtained in the X first step had to have a terminal hydroxyl group content below 50 molar % based on the terminal groups of the prepolymer.

Since the disclosure of D1 was not only limited to what was explicitly described in that document, but also included any features implicit to a person skilled in the art, the subject-matter of present claim 1 lacked novelty in view of D1.

III. On 23 September 1997, a Notice of Appeal against the above decision was filed, the prescribed fee being paid on the same day.
With the Statement of Grounds of Appeal, filed on 3 November 1997, the Appellant (Applicant) submitted a new set of Claims 1 to 8 in replacement of the previous claims.

According to the submission of the Appellant, further features had been introduced into the Claim 1, in particular the requirement, in the second step in the process, that the molten reaction mixture was cooled down to the glass transition temperature of the prepolymer or below and, after cooling the molten reaction mixture in the second step, the mixture was pelletized.

In relation to the above claims, the Appellant argued substantially as follows:

(a) Whilst the relevant Example 1 of Document 1 disclosed a process for preparing an aromatic polycarbonate, comprising the steps of conducting a melt-polycondensation of Bisphenol A and diphenyl carbonate in the presence of a catalyst in order to obtain a prepolymer in the molten state, pelletizing the prepolymer, and conducting a melt-polycondensation of the formed prepolymer, D1 did not disclose that the molten reaction mixture obtained in the first step was solidified by cooling the reaction mixture to the glass transition temperature of the prepolymer of below. The reference in D1 to the reaction mixture from the first step being cut into pellets neither explicitly nor inherently disclosed that the molten reaction mixture was solidified by cooling the mixture down to the glass transition temperature of the prepolymer or below. Consequently the subject-matter of claim 1 was novel over the disclosure of D1.
(b) The object of providing a process for producing a colourless (co)polycarbonate having a high molecular weight as well as excellent thermal resistance and impact resistance, had been solved by the process according to the new Claim 1. By cooling the prepolymer down to its glass transition temperature or below between the two polymerization steps exposure of the prepolymer to high temperature (of about 200°C) was avoided. This cooling step resulted in a final polycarbonate which was colourless and had a hue value of 0.06 to 0.09, as evidenced by Examples 1 to 11 of the application in suit.

If, contrary to the above, the prepolymer were not cooled down to the glass transition temperature or below between the two polymerization steps, but were exposed to high temperature, the prepolymer would be decomposed by heat and the final polycarbonate would have a hue value of 0.18 to 0.19, as evidenced by Comparative Examples 1 and 2.

These effects were neither disclosed nor rendered obvious by the cited prior art. Consequently the subject-matter was not only novel but inventive.

IV. Following a communication of the Board, issued on 26 September 2000 by fax, notifying the intention of the Board to summon the Appellant to attend oral proceedings on the 15 December 2000, and raising objection, under Article 123(2) EPC, in relation to the absence of an apparently essential limitation in Claim 1 filed together with the Statement of Grounds of Appeal, the Appellant filed, together with a further submission dated 26 September 2000, and received on 27 September 2000, a further set of claims 1 to 8. Claim 1 of the latter set reads as follows:
"1. A process for producing a (co)polycarbonate comprising the steps of:

(1) conducting a melt-polycondensation of a dihydroxy compound and a carboxylic diester as monomers in the presence of a transesterification catalyst in a tank reactor, while removing a monohydroxy compound formed by the melt-polycondensation, thereby obtaining a reaction mixture comprising a prepolymer in a molten state, said prepolymer having a terminal hydroxyl group content of 50 mol% or below based on all the terminal groups of the prepolymer, and said reaction mixture having a limiting viscosity number $[\eta]$ of 0.1 to 0.4 dL/g and a hydroxyl group concentration of $1 \times 10^{-3}$ mol/g or below,

(2) cooling the molten reaction mixture obtained in the first step to the glass transition temperature of the prepolymer or below by using an inert substance being at a temperature of 50°C or below in order to solidify it, and pelletizing the cooled mixture, and

(3) conducting a melt-polycondensation of the reaction mixture comprising the prepolymer in a horizontal polycondensation reactor, thereby obtaining a reaction mixture comprising a high-molecular-weight (co)polycarbonate, said reaction mixture having a limiting viscosity number $[\eta]$ of 0.3 to 1.0 dL/g, a hydroxyl group concentration of $5 \times 10^{-4}$ mol/g or below and a terminal hydroxyl group content of 20 mol% or below based on all the terminal groups of the high-molecular-weight (co)polycarbonate."

Claims 2 to 8 are dependent claims directed to elaborations of the process according to Claim 1.
The Appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of the set of Claims 1 to 8, filed on 27 September 2000. An auxiliary request to fix oral proceedings (Notice of Appeal, paragraph b) was withdrawn if the Board decided to remit the application to the Examining Division for further examination (submission dated 26 September 2000, final paragraph).

Reasons for the Decision

1. The appeal is admissible.

2. Admissibility of amendments

Claim 1 represents a combination of the features of Claims 1 and 9 to 15 of the application as originally filed, together with the further limitation on step (2) of the process, that the cooling of the molten reaction mixture obtained in the first step is to the glass transition temperature of the prepolymer or below.

The latter feature is disclosed explicitly in the general description of the application as originally filed, on page 6, first paragraph, according to which, it had been found that, "when the formed prepolymer is solidified by cooling it to its glass transition temperature or below in order to avoid the holding of the prepolymer in a molten state under heating... so that the time taken for exposing the prepolymer to a high temperature is reduced as a whole, the progress of the thermal decomposition reaction can be inhibited and
no undesirable influence is exerted on the hue, thermal resistance, residence stability in the moulding machine, water resistance, weather resistance, etc., of the intended polymer* (printed application, page 3, lines 15 to 20).

In view of the above, it is evident that cooling, in the relevant step (2), of the prepolymer to its glass transition temperature or below, is a feature which is regarded as crucial to obtaining the desired effects. It is thus a quite general requirement of the process, which should be incorporated in Claim 1 for a definition of the process corresponding to the description (Article 84 EPC).

Dependent Claims 2 to 7 are based on Claims 2 to 7, respectively, of the application as originally filed, and Claim 8 on Claim 16 of the application as originally filed.

In summary, no objection arises under Articles 123(2) and 84 EPC to the present amended claims of the application in suit.

3. Novelty

According to the relevant passage of Example 1 of D1, relied upon in the decision under appeal, "Thus formed prepolymer withdrawn by means of a gear pump from the bottom of the evaporator was passed through a die in a nitrogen atmosphere into a strand, followed by cutting into pellets using a cutter."

Whilst it is evident that the nitrogen atmosphere is capable of cooling the prepolymer to the extent that it is sufficiently solid to form a strand which can be cut
with a cutter, neither the temperature of the strand nor of the nitrogen atmosphere is disclosed in D1. Nor is there any indication of the glass transition temperature of the prepolymer in the strand. Even if the latter could be determined, for instance by repeating the Example, however, the absence of any information as to the temperature to which the strand is cooled means that it is not derivable, even in principle, from this disclosure, whether the latter temperature lies above or below the relevant glass transition temperature.

For this reason, the disclosure of D1 is not novelty-destroying for the subject-matter of Claim 1 of the application in suit.

It follows from the above, that the subject-matter of Claims 1 to 8 is novel. Hence, the decision under appeal must be set aside.

4. Inventive step; remittal

Whilst the Statement of Grounds of Appeal contains a number of arguments in support of the recognition of an inventive step in the subject-matter claimed in the application in suit, in particular that Examples 1 to 11 of the application in suit provided evidence that the distinguishing feature of cooling to the glass transition temperature or below was responsible for the desirable effects obtained (section III., above), it is noted that, whilst Examples 1 to 9 include a cooling step which may be read in the light of the general reference to this feature at the top of page 6, forming the basis of the corresponding limitation in Claim 1, Examples 10 and 11 contain no such cooling step, and
yet apparently lead to a product of satisfactory hue of 0.09 (Examples 10 and 11). Thus there is some doubt as to whether the evidence of the examples entirely supports the position of the Appellant in this respect.

It is, however, evident from the examination file that the Examining Division had not reached a final determination on the issue of inventive step, and the Board would not wish to prejudge the issue.

Consequently, it is the intention of the Board to exercise its powers under Article 111(1) EPC to remit the case to the Examining Division for further prosecution, there being no need under these circumstances to appoint oral proceedings before the Board.

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 for further prosecution.

The Registrar:  

E. Gorgmeier

The Chairman:

C. Gérardin

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