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D E C I S I O N
of 10 August 2000

Case Number: T 1187/97 - 3.3.3

Application Number: 91111989.9

Publication Number: 0468360

IPC: C08G 64/26

Language of the proceedings: EN

Title of invention:

Single-step method for preparing polycarbonates from
bischloroformates

Applicant:

GENERAL ELECTRIC COMPANY

Opponent:

-

Headword:

-

Relevant legal provisions:

EPC Art. 123(2), 84, 54

Keyword:

"Amendments - added subject-matter (no)"
"Claims - clarity (yes)"
"Novelty - prior disclosure of controlled final step"

Decisions cited:

T 0006/84, T 0689/90, T 0590/94

Catchword:

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Boards of Appeal

Chambres de recours

Case Number: T 1187/97 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 10 August 2000

Appellant: GENERAL ELECTRIC COMPANY
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Decision under appeal: Decision of the Examining Division of the
European Patent Office dated 17 March 1997 and
issued in writing on 14 July 1997, refusing
European patent application No. 91 111 989.9
pursuant to Article 97(1) EPC.

Composition of the Board:

Chairman: C. Gérardin
Members: B. ter Laan
C. Rennie-Smith

Summary of Facts and Submissions

- I. European patent application No. 91 111 989.9, filed on 18 July 1991, claiming priority of 27 July 1990 from an earlier application in the USA (US 559130) and published on 29 January 1992 under No. 0 468 360 (Bulletin 92/05) was refused by a decision of the Examining Division of the European Patent Office dated 14 July 1997. That decision was based on a set of twenty claims, of which Claim 1, filed on 12 January 1996, read:

"A method for preparing an aromatic polycarbonate which comprises:

effecting contact between an aromatic bischloroformate composition and a chain-terminating amount of a monohydroxyaromatic compound or salt thereof, in a heterogeneous reaction medium comprising water and an inert, substantially water-insoluble organic liquid, by introducing into the mixture of said bischloroformate composition and monohydroxyaromatic compound an interfacial polycarbonate formation catalyst and an aqueous alkali metal base, said base being introduced without pH control and in an amount which is at least stoichiometrically sufficient to convert all of the bischloroformate to polycarbonate of the desired molecular weight and to finally form a non-emulsified heterogeneous system; and continuing base addition incrementally as necessary to bring the pH of the aqueous phase of the reaction mixture to a value within the range of about 11-14."

Dependent Claims 2 to 20, which had not been amended, referred to preferred embodiments of the process according to Claim 1.

The Examining Division held that the claimed subject-matter did not satisfy the requirements of Articles 84 and 54 EPC.

- (i) As regards Article 84 EPC, it was found that the expressions "without pH control" and "in an amount which is at least stoichiometrically sufficient" were contradictory as the latter term did imply pH control. Also, the Applicant's interpretation of the former expression was not in conformity with the process described in the description and examples.
- (ii) Regarding novelty, the process defined by the present wording of the claims was disclosed by all of the cited documents:

- D1: US-A-4 743 676,
- D2: EP-A-0 293 891,
- D3: US-A-3 646 102,
- D4: US-A-4 737 573 and
- D5: EP-A-0 456 052.

In particular, in the control example of D1 a process comprising capping and polymerization in one step was described and it was shown that the resulting polymer had a higher molecular weight than the product prepared by separate capping and polymerization.

D2 disclosed a process according to which the components were added in the same order as now claimed without any measurement of the pH. The components used were not excluded by the present definition in Claim 1 of the "bischloroformate composition".

In D3 a bischloroformate composition containing bisphenol A was contacted with an endcapping agent and a catalyst, followed by addition of a base. The present application did not exclude the possible presence of bisphenol A in the bischloroformate composition and the reaction conditions of D3 were such that the pH requirements and those for phase separation were met. The mere measurement of a parameter did not establish novelty over a document that did not explicitly mention that parameter.

The latter argument was also valid regarding D4.

D5, a prior art document pursuant to Article 54(3) EPC, disclosed a method for preparing polycarbonates by contacting of a bischloroformate composition with a catalyst and a base. Determining the base requirements of the reaction system and then adding the precalculated amount of base in order to avoid the inaccurate pH reading as a result of emulsification, specifically mentioned in D5, corresponded exactly to the procedure applied in the present application.

Therefore, the claimed subject-matter was not novel.

III. On 8 September 1997 a Notice of Appeal was lodged against that decision, together with payment of the prescribed fee. Although in the Notice of Appeal reference was made to a new set of claims to be filed with the Statement of Grounds of Appeal, the latter, filed on 24 November 1997, did not contain any such claims. However, by a fax received on 7 August 2000, an alternative set of 20 claims was filed.

At the oral proceedings before the Board, held on 10 August 2000, after several objections by the Board concerning the wording of the claims according to the

main and the auxiliary requests, both requests were abandoned and replaced by a new set of nineteen claims as the sole request. Claim 1 of that request reads a follows:

"A method for preparing an aromatic polycarbonate which comprises:

- (a) effecting contact between an **essentially phosgene free aromatic bischloroformate composition obtainable by passing phosgene into a heterogeneous mixture consisting essentially of water, a substantially inert, substantially water-insoluble organic liquid and at least one dihydroxyaromatic compound in a tank reactor, and simultaneously introducing an aqueous alkali or alkaline earth metal base solution at a rate to maintain the aqueous phase of said mixture at a pH in the range of 8-11; the temperature of said mixture being maintained in the range of about 15°-50°C., the volume ratio of aqueous to organic phase at the conclusion of phosgenation being in the range of about 0.4-1.0:1 and the total amount of phosgene employed being at least about 1.1 mole per mole of dihydroxyaromatic compound; said mixture being agitated under conditions at least sufficient to prevent segregation of the aqueous and organic liquid phases, and a chain-terminating amount of a monohydroxyaromatic compound or salt thereof, in a heterogeneous reaction medium comprising water and an inert, substantially water-insoluble organic liquid, by introducing into the mixture of said bischloroformate composition and monohydroxyaromatic compound an interfacial polycarbonate formation catalyst and an aqueous alkali metal base, said base being introduced in an amount of from 0.25-0.75 equivalent per mole**

of structural units in the bischloroformate composition which is at least stoichiometrically sufficient to convert all of the bischloroformate to polycarbonate of the desired molecular weight and to finally form a non-emulsified heterogeneous system; and

- (b) continuing base addition incrementally to bring the pH of the aqueous phase of the reaction mixture to a value within the range of **10-13.**" (Amendments to the claim as originally filed are indicated in bold by the Board)."

Dependent Claims 2 to 19 refer to preferred embodiments of the process according to Claim 1.

V. The Appellant's arguments submitted in writing and during oral proceedings can be summarised as follows:

- (i) Regarding Article 123(2) EPC, the application as originally filed provided an adequate basis for the amendments.
- (ii) Regarding Article 84 EPC, the present wording of the claims provided a clear definition of the claimed subject-matter, particularly regarding a) the implicit absence of bisphenol A in the polymerization mixture, b) the amount of alkali metal base added in the first step, which was carried out without direct pH control and c) the second polymerization step which was carried out under controlled pH conditions.
- (iii) Regarding novelty, D1 differed from the claimed subject-matter in that the process involved only one step which was carried out while maintaining

the pH at a certain value. The breaking of the emulsion was not mentioned, nor the continuation of base addition under controlled pH conditions.

Both D2 and D3 had bisphenol A present in the polymerization mixture, which in the present application, in view of the process features now claimed, was excluded. Also, no chain terminating agent was added, nor was there any indication of continued base addition and pH control after breaking of the emulsion.

In D4 the reaction was controlled by progressive addition of the catalyst, whereas the pH was to be maintained within a certain range by means of base addition. No pH control after emulsion breakage was mentioned.

D5 described a method for making end-capped polycarbonates from phenol monochloroformate polycarbonate oligomers. Bischloroformates were only described in comparative examples. Unlike the present application, that method involved pH control of the reaction mixture. Also, no incremental continuation of base addition after emulsion breakage was indicated.

- VI. The Appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of Claims 1 to 19 as filed during the oral proceedings.

Reasons for the Decision

1. The appeal is admissible.

Article 123(2) EPC

2. With respect to the wording of Claim 1 as originally filed, the present version differs by (i) the requirement that the bischloroformate composition should be essentially phosgene-free, (ii) the definition of the bischloroformate compositions by means of process features for preparing it, (iii) the deletion of the feature "without pH control", (iv) the amount of alkali base, (v) the deletion of the term "as necessary" before the final pH adjustment and (vi) the values of the final pH range.
 - 2.1 According to original page 4, lines 11 to 13 of the application, feature (i) is an essential characteristic of the bischloroformate compositions used in the invention, which should therefore be present in Claim 1 (see also Article 84 EPC).
 - 2.2 Feature (ii) is supported by the explicit reference to US-A-4 737 573 (document D4 in the present proceedings) in the application in suit (page 4, lines 16 to 20 as originally filed), which discloses an appropriate method for the preparation of bischloroformate compositions from phosgene and a bisphenol. The Board considers the insertion of a passage from a prior art document in this case permissible as the document was specifically "incorporated by reference herein" (original application, page 4, lines 18 to 19), thus making its contents an integral part of the application in suit. By incorporating in Claim 1 the complete passage corresponding in extenso to step (A) of the process described in D4, all the essential process

features described there as essential for preparing a bischloroformate oligomer composition have been considered in combination and, thereby, also become essential for a complete definition of the present process (see also decisions T 6/84 (OJ EPO 1985, 238), T 590/94 of 3 May 1996, not published in OJ EPO and T 689/90(OJ EPO 1993, 616)).

- 2.3 The feature "without pH control" had not been defined in the original application. From the Appellant's explanations, in line with the description, it appeared that the pH was not monitored rather than not controlled. According to the original description, page 7, line 16 to page 9, line 2, in particular page 7, lines 16 to 24, the control of the pH is carried out by adding a certain precalculated amount of base to the reaction mixture, which amount is now inserted in Claim 1 (feature (iv)). Therefore, the deletion of feature (iii) and the insertion of feature (iv) serve to clarify that step and as such are allowable.
- 2.4 The addition of further base in reaction step (b) "as necessary" indicated that, depending on the circumstances, additional base was added or not in order to bring the pH to the desired value of 10 to 13. However, in all examples further base was added after the mixture had become non-emulsified. This, together with the original wording of the claim, provides sufficient support for the deletion of "as necessary" (feature (v)).
- 2.5 Feature (vi) serves to bring the required pH range in the second process step in conformity with the values disclosed on page 9, line 17 of the original application.

- 2.6 In view of the above, the Board is satisfied that the requirements of Article 123(2) are met.

Article 84 EPC

3. The present wording of the claims also satisfies the requirements of Article 84 EPC in terms of clarity and conformity with the description.
- 3.1 In particular, the process features for the preparation of the bischloroformate composition overcome the discrepancy between the description, which gives a long list of suitable substantially inert, substantially water-insoluble organic liquids to carry out the polycondensation reaction (cf. page 5, line 31 to page 6, line 9), and the specific examples, in which the solvent is comprised in the bisphenol A bischloroformate oligomer composition used as the starting compound for that polycondensation reaction.
- 3.2 The unclear expression "without pH control" has now been replaced by the addition of a specified amount of base, in conformity with the information contained in the original description.
- 3.3 By the deletion of the term "as necessary" in process step (B) the addition of further base has become an obligatory feature, in conformity with the original description, page 9, lines 11 to 15 and the examples.

Novelty

4. The novelty objection of the Examining Division was based on five documents.

4.1 D1 describes a method for preparing a linear aromatic polycarbonate of controlled molecular weight which comprises the steps of:

- (A) reacting an aromatic bischloroformate composition with a monohydroxyaromatic compound or salt thereof in a reaction system also comprising water, a substantially inert, substantially water-insoluble organic liquid and an alkali or alkaline earth metal base, to produce a partially capped bischloroformate composition; and
- (B) contacting said partially capped bischloroformate composition with an interfacial polycarbonate formation catalyst and an aqueous alkali metal or alkaline earth metal base to form said linear aromatic polycarbonate (Claim 1).

According to the description (column 4, lines 65 to 66; column 5, lines 1 to 6), the pH in step (A) is stabilized at about 11.8 to 12.3 while maintaining a relatively high flow rate in the vicinity of the pH monitoring means in order to prevent fouling. In step (B) the pH is in excess of about 10, preferably in the range of about 11 to 14 (column 6, lines 20 to 26). In the examples the pH is maintained at a fixed value of 8.5 during step (A), after which additional base and catalyst are introduced.

D1 hence discloses a two step process in which the partial endcapping of bischloroformate is followed by polymerization, which is initiated by the addition of an appropriate catalyst. Although the present wording of Claim 1 does not exclude the possibility of carrying out the first step of the process in the same way as

defined by D1, that document does not mention a further step in which the pH is raised to a value of 10 to 13 after the emulsion formed during polymerization was broken. In fact, the formation of an emulsion is not mentioned at all and does not play a role in the process according to D1.

4.2 D2 describes a process for producing high impact resistant polycarbonate consisting of a main chain and terminal groups bonded thereto, said main chain comprising a repeating unit represented by a certain specific formula, and at least one of the terminal groups being a cumylphenoxy group represented by another certain specific formula and having a viscosity average molecular weight of at least 10,000 and an acetone soluble content of not more than 3.5% by weight, which process comprises reacting a polycarbonate oligomer derived from bisphenol A, bisphenol A and cumylphenol while stirring so that the reaction mixture is in a turbulent flow condition; adding an aqueous solution of alkali hydroxide at the point of time that the viscosity of the reaction mixture increases; and subsequently continuing the reaction in a laminar flow condition (Claim 5). In Example 1, to a polycarbonate oligomer composition a solution containing a base and bisphenol A is added. A catalyst and a chain terminator are then introduced. After further addition of base the reaction is completed. In Example 5, a polymerization modifier is added to a polycarbonate oligomer composition. A solution containing a base and a catalyst is added to the former mixture, after which a third composition containing bisphenol A and base is introduced.

In the polymerization mixture of D2 bisphenol A is present, which, according to the Appellant, is not the case with the composition obtainable by the method now defined in Claim 1. However, although the Appellant's explanation in itself can be accepted, the wording of present Claim 1 does not exclude the presence of bisphenol A in the heterogenous reaction medium ("... comprising water and an inert, substantially water-insoluble organic ..."). Furthermore, present Claim 1, step (A), merely indicates that a catalyst and a base are introduced into the mixture of chain-terminator and bischloroformate, so that the addition of base after increase of the viscosity is not excluded either. However, the combination of introducing a catalyst and a base to a composition containing bischloroformate and a chain terminator, polymerizing until a non-emulsified heterogeneous system is formed and then additionally increasing the pH is not mentioned in D2. Therefore, D2 does not destroy the novelty of the subject-matter of Claim 1 as it is now worded.

4.3 D3 describes a method for continuously preparing polycarbonate oligomer which comprises (Claim 1):

- (a) introducing an aqueous caustic alkali solution of dihydroxy compound and an organic solvent into a tubular reactor;
- (b) said organic solvent being capable of dissolving said polycarbonate oligomer;
- (c) forming a mixed phase stream of the organic solvent and aqueous solution in said tubular reactor;

- (d) feeding phosgene thereto and causing said phosgene to react with said mixed phase stream in a parallel-current flow while allowing the heat of reaction which evolves at the phosgenation reaction to vaporize organic solvent;
- (e) and then condensing the vaporized organic solvent near the reactor outlet by cooling the outside of the reactor.

In the examples the polycarbonate oligomer thus produced is polymerized in the presence of a base, bisphenol A, a catalyst and a chain terminator. No mention is made of emulsion formation or of continuing the polymerization at a higher pH after the reaction mixture returned to a heterogeneous state.

4.4 D4 describes a method for preparing a linear polycarbonate which comprises the steps of:

- (A) preparing a bischloroformate oligomer composition by passing phosgene into a heterogeneous mixture consisting essentially of water, a substantially inert, substantially water-insoluble organic liquid and at least one dihydroxyaromatic compound in a tank reactor, and simultaneously introducing an aqueous alkali or alkaline earth metal base solution at a rate to maintain the aqueous phase of said mixture at a pH in the range of 8-11; the temperature of said mixture being maintained in the range of about 15-50°C, the volume ratio of aqueous to organic phase at the conclusion of phosgenation being in the range of about 0.4-1.0:1 and the total amount of phosgene employed being at least about 1.1 mole per mole of dihydroxyaromatic compound; said

mixture being agitated under conditions at least sufficient to prevent segregation of the aqueous and organic liquid phases;

(B) passing an interfacial polycarbonate formation catalyst into a mixture of said bischloroformate composition, water and a substantially inert, substantially water-insoluble organic liquid in a tank reactor, with effective agitation, and simultaneously introducing an aqueous alkali or alkaline earth metal base solution at a rate to maintain the aqueous phase of said mixture at a pH in the range of about 10-14, the rate of catalyst addition being adjusted to effect polycarbonate formation under controlled conditions; and

(C) recovering said linear polycarbonate.

In the sole example in which a bischloroformate oligomer composition is polymerized to a polycarbonate (Example 7), the pH of the reaction mixture containing the bischloroformate is adjusted to 12 and the catalyst solution which also contains phenol, is metered in over 5 minutes. The pH is maintained during the reaction. No mention is made of emulsion formation, nor of continuing base addition after its breakage in order to bring the pH value within the range of 10 to 13.

Step (A) of D4 is now incorporated in Claim 1 as the process by which the bischloroformate composition is obtainable, so that this process step cannot render the subject-matter of present Claim 1 novel. Also, the wording of present Claim 1 does not exclude the possibility of carrying out the first step of the

present process in the same way as defined by D4. Although the addition of base is related to the pH value of 12, whereas in present claim the amount is correlated to moles of structural units in the bischloroformate composition, this may still imply the same amount of base as required in present Claim 1, which interpretation was not contradicted by the Appellant. However, D4 does not mention a further step in which the pH is raised to a value of 10 to 13 after the emulsion formed during polymerization was broken. As in D1 and D3, the formation of an emulsion is not mentioned at all and does not play a role in the process according to D4.

4.5 D5 (Claim 1) describes a method for making polycarbonate having a ratio of condensed phenolic endcapping units to condensed bisphenol carbonate units having a value from about 0.005 to 0.08, which comprises

- (1) introducing phosgene into an agitated mixture comprising bisphenol at a temperature of 15 to 50°C under interfacial reaction conditions and a pH between about 3.0 to about 11.2,
- (2) continuing the phosgenation of the mixture of (1) and allowing the further introduction of aqueous alkali metal hydroxide to proceed in accordance with a previously established set point between pH 3-10.5 using either a pH Control System A which allows the introduction of aqueous alkali metal hydroxide at a rate sufficient to substantially maintain the mixture at the previously established pH set point, or a pH Control System B, which allows the introduction

of aqueous alkali metal hydroxide at a rate sufficient to provide an introduction rate ratio of moles of aqueous alkali metal hydroxide, to moles of phosgene having a value of up to about 2.5,

- (3) allowing the introduction of aqueous alkali metal hydroxide to proceed in accordance with (2) until a sudden increase in base demand is noted with Control System A or a sudden decrease in pH of at least one unit is noted with respect to Control System B,
- (4) continuing the introduction of phosgene and alkali metal hydroxide in accordance with (3), until an additional 0.0025 to 0.09 mol of phosgene, per mol of bisphenol is introduced,
- (5) discontinuing the introduction of phosgene and alkali metal hydroxide, while continuing to agitate the resulting mixture until the mixture is substantially free of phosgene, while the pH is maintained between about 4 to 8,
- (6) introducing sufficient endcapping phenol into the mixture of (5) to provide the aforesaid mole ratio of condensed phenolic end capping units to condensed bisphenol carbonate units followed by or along with tertiary organic amine to provide a proportion of from about 0.05 to 2 mole of tertiary organic amine, per 100 mol of bisphenol, and sufficient alkali metal hydroxide to maintain the pH of the resulting mixture of between about 9 to 12,

- (7) allowing the polymerization of the bisphenol monochloroformate oligomer to proceed, and
- (8) recovering phenolic end-capped polycarbonate from the mixture of (7).

Although the disclosure of D5 concerns the polymerization of monochloroformate oligomers to polycarbonate, comparative Runs 4 to 6 refer to the polymerization of bischloroformate which, instead of monochloroformate, is produced by using higher levels of phosgene (page 6, lines 33 to 36). Polymerization is carried out under pH control by adding base, with a set point of 9. Exceeding the set point caused the base addition to be shut off (page 6, lines 43 to 45). On page 6, lines 45 to 50, the fouling problems when using bischloroformate oligomers are described, as well as the solution to that problem: by characterization of the bischloroformate prior to base addition, the base requirement for polymerization is calculated; the calculated amount of base is then added as a constant flow rate over a period of 5 minutes. Although no exact amounts are given, the requirement in present Claim 1 that the amount of base should be at least stoichiometrically sufficient to convert all of the bischloroformate to polycarbonate cannot be regarded as a distinguishing feature; similarly, the recommendation to calculate the amount of base by way of titration (original application, page 7, line 25 to page 8, line 29) is in effect not different from the necessity to calculate the amount of base, although formulated in a more precise way. Therefore, that quantitative

feature cannot serve to establish novelty. However, D5 does not disclose continued base addition after breaking of the emulsion to raise the pH to a value of 10 to 13.

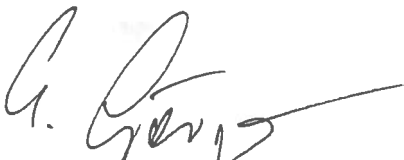
- 4.6 From the above analyses of D1 to D5 it is clear that none of those documents discloses the second process step as now required in Claim 1 of continuing the addition of base after the emulsion has ceased to exist, to bring the pH to a value of 10 to 13, in combination with the details of process step (A) as now required in Claim 1.
- 4.7 For the above reasons, the Board comes to the conclusion that the subject-matter of Claim 1 is novel.
5. As Claim 1 of the main request is novel, the same goes for dependent Claims 2 to 19, the novelty of which is supported by that of Claim 1.
6. The decision under appeal referred only to novelty and did not address the issue of inventive step. Therefore, the Board, in order not to deprive the Appellant of the possibility of being heard by two instances, does not consider it appropriate to deal with the issue of inventive step. Accordingly, the Board remits the case to the first instance for further prosecution pursuant to Article 111(1) EPC.

Order

For these reasons it is decided that:

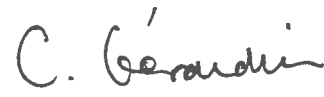
1. The decision under appeal is set aside.
2. The case is remitted to the Examining Division for further prosecution on the basis of Claims 1 to 19 of the main request as submitted during the oral proceedings.

The Registrar:



E. Görgmayer

The Chairman:



C. Gérardin

