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**D E C I S I O N**  
**of 29 October 2002**

**Case Number:** T 0410/00 - 3.3.3

**Application Number:** 94303447.0

**Publication Number:** 0626409

**IPC:** C08G 64/24

**Language of the proceedings:** EN

**Title of invention:**

Method for making aromatic polycarbonates

**Applicant:**

GENERAL ELECTRIC COMPANY

**Opponent:**

-

**Headword:**

-

**Relevant legal provisions:**

EPC Art. 56, 84

**Keyword:**

"Inventive step (yes) - problem and solution"  
"Claims - clarity (after amendment)"

**Decisions cited:**

-

**Catchword:**

-



Case Number: T 0410/00 - 3.3.3

**D E C I S I O N**  
of the Technical Board of Appeal 3.3.3  
of 29 October 2002

**Appellant:** GENERAL ELECTRIC COMPANY  
1 River Road  
Schenectady, NY 12345 (US)

**Representative:** Szary, Anne Catherine, Dr.  
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**Decision under appeal:** Decision of the Examining Division of the  
European Patent Office issued in writing on  
22 October 1999 refusing European patent  
application No. 94 303 447 pursuant to  
Article 97(1) EPC.

**Composition of the Board:**

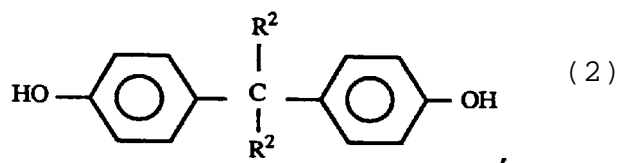
**Chairman:** R. Young  
**Members:** A. Däweritz  
U. Tronser

### Summary of Facts and Submissions

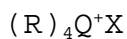
- I. European patent application No. 94 303 447.0, filed on 13 May 1994, claiming the priority of 24 May 1993 of an earlier application in the United States of America (066 382) and published under No. 0 626 409 on 30 November 1994, was refused by a decision of the Examining Division, issued in writing on 22 October 1999.
- II. The decision was based on a set of nine claims, Claims 1 and 2 having been submitted with a letter dated 25 January 1999, and Claims 3 to 9 in their initial version.

The claims read as follows:

- "1. A method for making polycarbonate, comprising,  
(A) effecting reaction under interfacial reaction conditions at a pH in the range of 9 to 12.5 between a phenolic chain-stopper and a bisphenol of the formula,



where R<sup>2</sup> is selected from the same or different C<sub>(1-4)</sub> alkyl group, and a substantially stoichiometric amount of phosgene in the presence of an amount of phase transfer catalyst of the formula,



which is effective for providing polycarbonate having a molecular weight in the range of 25K to 180K compared to a polystyrene standard, and (B) substantially eliminating chloroformate end groups in the resulting mixture of (A) prior to polycarbonate recovery,

where R can be the same or different C<sub>(4-10)</sub> alkyl group, Q is a nitrogen or a phosphorus atom, X can be a halogen atom, or an -OR<sup>1</sup> group, R<sup>1</sup> is a member selected from the group consisting of H, C<sub>(1-18)</sub> alkyl and C<sub>(6-18)</sub> aryl, and R<sup>2</sup> is selected from the same or different C<sub>(1-4)</sub> alkyl group.

2. A method in accordance with claim 1, where the bisphenol is bisphenol A.
3. A method in accordance with claim 1, where the phase transfer catalyst is tetrabutylammonium hydroxide.
4. A method in accordance with claim 1, where the phase transfer catalyst is tetrabutylphosphonium bromide.
5. A method in accordance with claim 1, where the phase transfer catalyst is tetrabutylammonium bromide or tetrabutylammonium chloride.
6. A method in accordance with claim 1, where the phenolic chain-stopper is phenol.
7. A method in accordance with claim 1, where the phenolic chain-stopper is cumyl phenol.
8. A method in accordance with claim 1, where the

chloroformate end groups are eliminated by addition 100 ppm to 1000 ppm of triethylamine followed by the addition of sufficient phosgene to react with any residual phenolic hydroxy.

9. A method in accordance with claim 1, where the chloroformate end groups are eliminated by addition of an aromatic phenol compound."

Whilst novelty was acknowledged in the decision, the request for grant of a patent was refused for lack of inventive step in the light of the disclosures of

D2: EP-A-0 013 904 and

D3: DE-A-2 503 150.

The Examining Division held that the claimed method differed from that in D3 only by the use of a different quaternary ammonium salt as the catalyst and that there was no evidence on file that the use of the different catalyst was associated with a technical effect.

Therefore, the technical problem to be overcome was seen only in the provision of a further process for making polycarbonate, which was solved in an obvious way by selecting further suitable quaternary ammonium salt phase transfer catalysts, such as those described in D2.

- III. On 13 December 1999, a Notice of Appeal against the above decision was lodged by the Appellant (Applicant). The prescribed fee was paid on the same date.

In the Statement of Grounds of Appeal, received on 1 March 2000, the Appellant requested that the decision be set aside and a patent be granted on the basis of

the claims on file.

The arguments submitted by the Appellant can be summarised as follows:

The application related to a method for making polycarbonate by (A) phosgenating a bisphenol in the presence of a specific phase transfer catalyst (**PTC**) according to the definition in Claim 1, whereby the bisphenol and phosgene were used in substantially stoichiometric amounts, and (B) eliminating chloroformate end groups in the resulting mixture of (A) prior to polycarbonate recovery.

The problem to be overcome by the application was seen in the provision of high molecular weight polycarbonates free of residual chloroformate end groups. Such end groups would cause the resins to discolour to a high degree upon extrusion.

Furthermore, in the prior art, the use of tertiary organic amines as effective condensation catalysts in this reaction had been found to lead to excessive phosgene usage as a result of phosgene hydrolysis and/or chloroformate hydrolysis prior to the termination of capping. The rate of these side reactions was now found to be considerably reduced when the specific PTC according to Claim 1 were used.

The process of Document D3, which was considered in the contested decision to represent the closest state of the art, differed from the claimed subject-matter both in the choice of the PTC catalyst and in the absence of step (B), ie the removal of chloroformate groups.

Document D2 concerned the problem of forming polycarbonates from sterically hindered bisphenols which was overcome by the choice of quaternary ammonium salts as the catalyst. In that process, a substantial excess of phosgene was used, and the above step (B) was missing. It would not be derivable from D2 that it was possible to prepare the desired polycarbonate when using substantially stoichiometric amounts of phosgene and bisphenol.

Neither D3 nor D2 would have taught to use the specific PTC as defined in the claims nor to remove the chloroformate end groups from the polymer before its recovery.

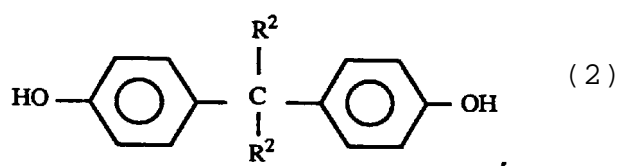
- IV. In a conversation by telephone on 17 September 2002 between the Representative and the Rapporteur, the meaning of the definitions of R and X in the phase transfer catalyst was discussed. Another point discussed was that whilst, according to the application text, the reaction in step (A) is carried out in the presence of an aqueous phase, the wording of Claim 1 is silent in this respect.

In reply to this conversation, it was argued by the Appellant that effecting the reaction in step (A) "under interfacial reaction conditions" implicitly disclosed "the presence of an organic phase and an aqueous phase", and two amended versions of Claim 1, followed in each case by an unchanged Claim 2, were submitted ("Main request" and "Auxiliary request"; letter of 24 September 2002). In both versions of Claim 1, the expressions "R can be" and "X can be" were replaced by "R is selected from" and "X is selected from", respectively.

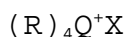
Claim 1 of the main request, as amended, reads:

"1. A method for making polycarbonate, comprising,

(A) effecting reaction under interfacial reaction conditions at a pH in the range of 9 to 12.5 between a phenolic chain-stopper and a bisphenol of the formula,



where R<sup>2</sup> is selected from the same or different C<sub>(1-4)</sub> alkyl group, and a substantially stoichiometric amount of phosgene in the presence of an amount of phase transfer catalyst of the formula,



which is effective for providing polycarbonate having a molecular weight in the range of 25K to 180K compared to a polystyrene standard, and

(B) substantial[ly] eliminating chloroformate end groups in the resulting mixture of (A) prior to polycarbonate recovery,

where R is selected from the same or different C<sub>(4-10)</sub> alkyl group, Q is a nitrogen or a phosphorus atom, X is selected from a halogen atom, or an -OR<sup>1</sup> group, R<sup>1</sup> is a member selected from the group consisting of H, C<sub>(1-18)</sub> alkyl and C<sub>(6-18)</sub> aryl, and R<sup>2</sup> is selected from the same or different C<sub>(1-4)</sub> alkyl



group."

In Claim 1 according to the auxiliary request, the passage ", in the presence of an organic solvent and an aqueous phase," was inserted after "interfacial reaction conditions" (in step (A)).

- V. The Appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of Claims 1 and 2, as submitted with the letter dated 24 September 2002 and headed "Main request", and Claims 3 to 9 as originally filed or, alternatively, Claims 1 and 2, as submitted with the same letter and headed "Auxiliary request", and the above Claims 3 to 9.

### **Reasons for the Decision**

1. The appeal is admissible.
2. *Article 123(2) EPC*

Claim 1, as amended according to the main request, is based on the following passages of the application as originally filed: Claim 1, page 3, lines 13 and 15.

The additional feature inserted in Claim 1 according to the auxiliary request is supported by page 5, lines 7/8 and 24 to 29; page 6, lines 19 to 22; page 8, lines 7 and 10; and page 9, line 1.

The remaining claims are identical to those as originally filed.

Consequently, the claims of both requests comply with

Article 123(2) EPC.

3. *Article 84 EPC (main request)*

3.1 Due to the amendment of the definitions of groups R and X, the scope of the formula defining the phase transfer catalyst has become clear and unambiguous.

3.2 According to Ullmann's Encyclopaedia of Industrial Chemistry, 5<sup>th</sup> edition 1992, volume A21, pages 210 to 212, chapter 3.1 "Interfacial Polycondensation", "Interfacial polycondensation is currently used for the industrial production of polycarbonates [2],[5],[6]. Bisphenol A is reacted with phosgene at 20 to 40°C in a two-phase mixture consisting of an aqueous, alkaline phase and an immiscible organic phase." In Table 3 in "Encyclopedia of Polymer Science and Engineering", John Wiley & Sons, New York ... 1987, volume 8, page 226, reference is made to "Typical Interfacial Polycondensations with Stirring" and, *inter alia*, to "carbonate" polymer. In all these reactions, one reactant (such as phosgene) is present in an organic phase and the others (eg Bisphenol A ["**BPA**"] and tertiary amine) in an aqueous phase. According to Kirk-Othmer, Encyclopedia of Chemical Technology, 4<sup>th</sup> edition, volume 19, John Wiley & Sons, New York 1996, page 595, this reaction is carried out in the same way even after the filing date of the application in suit: "Interfacial Polymerization. Most BPA polycarbonate is produced by an interfacial polymerization process utilizing phosgene [75-44-5]. The interfacial process for polycarbonate preparation involves stirring a slurry or solution of BPA and ... chain stopper ... in a mixture of methylene chloride and water, while adding phosgene (qv) in the presence of a tertiary amine

catalyst ...". A similar statement can also be found in "Kunststoff-Handbuch" volume 3/1, Carl Hanser Verlag München 1992, page 142, chapter 3.3.2.3. It can be concluded from these encyclopaedias that the reference to an interfacial reaction for a person skilled in this art implicitly includes the presence of both an aqueous and an organic solvent phase.

Therefore, the Board is satisfied that the requirements of Article 84 EPC are met by the main request.

4. *Novelty*

Novelty of the claimed subject-matter was acknowledged in the decision under appeal. The Board has no reason to take a different view.

It follows that the requirements of Article 54 EPC are met.

5. *Problem and solution*

5.1 The patent application in suit concerns a method for making aromatic polycarbonate.

5.2 Such a method is known from D3 which the Board, like the Examining Division, regards as representing the closest state of the art.

5.2.1 In this citation, the problems are described which occurred in previously known methods of preparing polycarbonate by phosgenating aqueous solutions of alkali salts of aromatic dioxy compounds ("**bisphenol salts**"), such as BPA, in the presence of an organic solvent, capable of dissolving phosgene and

polycarbonate (paragraph bridging pages 1/2).

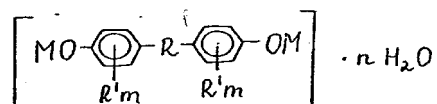
In one such previous method, carried out in a two-phase system of an aqueous alkali solution of BPA and an organic solvent as mentioned before, the bisphenol was polycondensed with phosgene in the presence of catalysts and monophenols as chain terminators. Intermediate products obtained in the first step of this method were terminated by chloroformate groups which could then be polycondensed further to polycarbonate having the desired molecular weight (paragraph bridging pages 2/3).

However, this previously known method had a number of disadvantages. Thus, significant amounts of aqueous alkali hydroxide and phosgene were wasted due to their consumption in side-reactions, ie by the hydrolysis of phosgene and/or the said chloroformate groups. Both side-reactions occurred when the reaction mixture contained an excess of alkali (pages 3 and 4).

- 5.2.2 According to Document D3, these disadvantages are avoided by means of a three-stage process for the preparation of polycarbonates wherein, in the first stage, the said bisphenol salts are prepared by reacting bisphenols suspended in the organic solvent, referred to above, a stoichiometric amount of the alkali hydroxide and an amount of water sufficient to provide, together with the water released by this reaction, the number of water molecules in the crystal hydrate formed in this stage (page 9, last paragraph to the bottom of page 10). In the second stage, the said bisphenol salts, in the form of small solid particles evenly suspended in an organic solvent as mentioned before, are subjected to phosgenation in the presence

of a catalyst and monophenols as chain terminators in an interfacial polycondensation to oligocarbonates, which are then, in the third stage, polycondensed further to polycarbonates in the presence of an amount of 0.3 to 0.7 mols, per mol of the bisphenol salt, of an aqueous solution of alkali hydroxides (of a 5 to 20% strength). Thereafter, the end product is recovered (pages 11 to 13).

The said crystal hydrates have the general formula



wherein R is >C(CH<sub>3</sub>)<sub>2</sub> or cyclohexyl; R' is Cl or Br; M is K or Na; n is an integer corresponding to the number of water molecules in the crystal hydrate; and m is 2 (Claim 1, page 5).

Whilst in the definition of the symbols in the above formula m is limited to the value of 2, the crystal hydrate of the disodium salt of 2,2-bis-(4-hydroxyphenyl) propane (BPA), ie a compound wherein m is zero, is referred to as the preferred starting compound at the top of page 6. This is confirmed by a number of physical properties of polycarbonate derived from BPA, most of the examples and particulars of the conversion of BPA to polycarbonate given in the description (page 8, last paragraph and subsequent pages; Examples 1, 2, 4 to 9, 11 to 14).

Whilst in the previously known method, tertiary amines or quaternary ammonium salts, in general, were used as catalysts, D3 refers only to tertiary amines or benzyltriethylammonium chloride and, in particular, the

latter compound (D3: page 2, last paragraph and page 12, lines 4 to 6; the examples).

Since the salts are reacted with phosgene in the absence of free water and free alkali, the consumption of water, phosgene and alkali can be reduced significantly (water to a tenth, phosgene and alkali by 20% compared to the previously known processes). Thus, the salt and phosgene are suitably kept during the phosgenation at a molar ratio of 1:1. Since the phosgenation can be carried out at the constant boiling point of the organic solvent, the molecular weight distribution of the polycarbonate can also be improved (page 6, paragraph 3 to page 8, paragraph 1; page 11, last paragraph).

5.3 According to introductory statements in D3, the presence of water and alkali in the phosgenation of bisphenols catalysed by tertiary amines or quaternary ammonium compounds results in a significant loss in phosgene due to side-reactions. In the light of these shortcomings and in line with the introductory statements in the patent application (page 2, line 23 to page 3, line 2), the technical problem underlying the patent application in suit may thus be seen in the definition of a simpler and improved process for the preparation of aromatic polycarbonates of high molecular weights from specific bis(4-hydroxyphenyl) alkanes whilst avoiding significant losses of phosgene due to side reactions and without the need to prepare, prior to the phosgenation, a specific intermediate form of a starting material in an additional reaction requiring sensitive control of the reaction conditions.

5.4 According to the patent application in suit, this

problem is solved by reacting, in a step (A), bis(4-hydroxyphenyl)alkanes according to the above formula (2) with a substantially stoichiometric amount of phosgene in the presence of a phenolic chain-stopper and a phase transfer catalyst of the formula  $(R)_4Q^+X$  (see Claim 1, section IV, above) under interfacial reaction conditions, at a pH value in the range of 9 to 12.5 to provide polycarbonate having a molecular weight in the range of 25K to 180K, compared to polystyrene standard, and then, before recovering the polycarbonate, substantially eliminating chloroformate end groups from the polymer (step (B)).

In both examples of the application in suit, BPA was phosgenated in a mixture of an aqueous phase and a methylene chloride phase in the presence of tetrabutyl ammonium hydroxide. Phosgene was used in excesses of only 2 and 1 mol%, respectively, based on the total amount of bisphenol and phenol. During this reaction, the pH value was maintained at 10.5 to 11 and 10.8, respectively, by means of an aqueous sodium hydroxide solution. In a second step, chloroformate terminal groups were eliminated from the polymer obtained in the first step, resulting in polycarbonates having molecular weights of 41.5K, 39.5K and 4.14K, respectively.

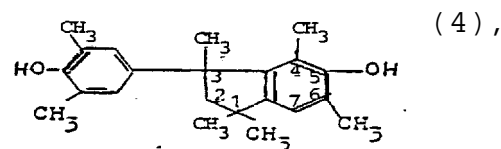
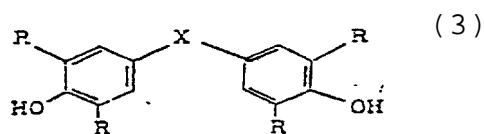
5.5 Consequently, the aspects of the above defined technical problem are evidently effectively solved by the method as defined in Claim 1.

6. *Inventive step*

It remains to be decided whether this solution was obvious to a person skilled in the art having regard to the state of the art relied upon by the Appellant.

6.1 It is evident from the above considerations that in D3 the disadvantages of the previous processes due to hydrolysis side-reactions were avoided in a completely different way by preparing a solid crystal hydrate of the bisphenol salt in a preliminary step, and explicitly working in the absence of any free water and alkali during the phosgenation. Indeed, it is doubtful, in view of the apparent substantially complete absence of an aqueous phase, whether this prior disclosure can properly be regarded as an interfacial process at all, as normally understood by a person skilled in this art (see section 3.2, above). Hence, there was no hint to the claimed solution of the technical problem, but on the contrary, this document teaches clearly away from the solution found according to the application in suit.

6.2 Document D2 relates to a process for preparing thermoplastic high molecular weight aromatic polycarbonates exclusively or predominantly from bis(tetra-ortho-C<sub>1</sub>- to -C<sub>3</sub>-alkylphenols), ie compounds according to the two general formulae





wherein R is C<sub>1</sub> to C<sub>3</sub> alkyl, and X denotes a direct single bond, C<sub>1</sub> to C<sub>5</sub> alkylene, C<sub>2</sub> to C<sub>5</sub> alkylidene, cyclohexylene, cyclohexylidene, -O- or -S-, in a catalysed phosgenation reaction at a pH value within a range of 11 to 14 (Claim 1; page 12, lines 2 to 4).

Previously, such polymers could be prepared only by using specific methods which disadvantageously, and in contrast to the process of D2, required the use of relatively high quantities of amines. Instead, quaternary ammonium salts are used in the process of D2 in amounts of 0.01 to 5 mol% as catalysts, optionally together with tertiary amines in amounts not exceeding 8 mol% (both relative to the total amount of the bisphenols; Claim 1; page 1 to page 4, line 11; page 5, last paragraph).

Further remarkable features of the process of D2 are seen in the facts that phosgene needs to be used in amounts of less than 170 mol%, in general less than 150 mol%, relative to the total amount of bisphenols charged, and that the products have low contents of terminal phenolic groups (page 7, lines 7 to 17). Thus, in Example 1, phosgene is used in an amount of approximately 150 mol% (cf. letter dated 13 July 1999, second paragraph). It is evident from these figures that, in this document, the consumption of phosgene in side-reactions is taken for granted.

In addition, the document refers to physical and chemical properties of the resulting polymers, ie an excellent resistance against solvolysis, hydrolysis or aminolysis and a high thermal stability under load. Other properties mentioned high second order transition temperatures and high temperature resistance (page 15,

line 13 *et seq.*).

However, the document does not refer to the above relevant technical problem, ie to avoid phosgene-consuming side-reactions and to obtain high molecular weight polycarbonates derived only from unsubstituted bis(4-hydroxyphenyl)alkanes (see the above formula (2)), and, therefore, cannot contribute to its solution.

- 6.3 It follows that the method of Claim 1 according to the main request would not be obvious to a person skilled in the art in view of D3, by itself, or D3 in combination with D2 and, therefore, involves an inventive step.
  
7. Claims 2 to 9 of the main request, which relate to specific embodiments of the method of Claim 1, are supported by the patentability of the main claim and thus also allowable.
  
8. Since the main request is successful, the auxiliary request need not be dealt with. Nor is it necessary to hold oral proceedings.

## Order

### For these reasons it is decided:

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 Claims 1 and 2, as submitted by letter dated 24 September 2002 and headed "Main request", and Claims 3 to 9, as originally filed, after any necessary consequential amendment of the description.

The Registrar:

The Chairman:

E. Görgmaier

R. Young