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### DECISION of 5 October 2000

**Case Number:** T 0747/99 - 3.3.3

Application Number: 95303430.3

Publication Number: 0687710

**IPC:** C08L 69/00

Language of the proceedings: EN

Title of invention: Copolyester-carbonate resin/PCT blends

#### Applicant:

GENERAL ELECTRIC COMPANY

# Opponent:

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### Headword:

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Relevant legal provisions: EPC Art. 56

Keyword: "Inventive step - obvious combination of known features"

Decisions cited:

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Catchword:



Europäisches Patentamt European Patent Office Office européen des brevets

Beschwerdekammern

Boards of Appeal

Chambres de recours

**Case Number:** T 0747/99 - 3.3.3

#### D E C I S I O N of the Technical Board of Appeal 3.3.3 of 5 October 2000

Appellant:	GENERAL ELECTRIC COMPANY		
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Decision under appeal:	Decision of the Examining Division of the	
	European Patent Office posted 2 March 1999	
	refusing European patent application	
	No. 95 303 430.3 pursuant to Article 97(1) EPC.	

Composition of the Board:

Chairman:	C.	Gérardin
Members:	в.	ter Laan
	Α.	Lindqvist

### Summary of Facts and Submissions

I. The European patent application No. 95 303 430.3, filed on 23 May 1995, claiming the priority of 14 June 1994 of an earlier application in the United States of America (US 259589) and published under No. 0 687 710 on 20 December 1995 (Bulletin 95/51), was refused by a decision of the Examining Division issued in writing on 2 March 1999.

> That decision was based, as main request, on a set of 10 claims filed on 28 April 1998 and, as auxiliary request, on an amended version of these claims filed on 19 October 1998.

Claim 1 of the main request reads as follows:

"1. A thermoplastic molding composition comprising:

from 50 to 99% by weight of at least one copolyester-carbonate resin which is a reaction product of at least one dihydric phenol, at least one carbonate precursor, and at least one aliphatic alpha omega dicarboxylic acid or ester precursor thereof, wherein the at least one aliphatic alpha omega dicarboxylic acid or ester precursor has from 9 to 40 carbon atoms and is present in the copolyester-carbonate in quantities ranging from 2 to 30 mole % based on the dihydric phenol; and

from 1 to 50% by weight of at least one polyester resin which is a condensation product of at least one diacid and at least one polyol in which the at least one diacid is comprised of greater than about 50% by weight of terephthalic acid and the at least one polyol is comprised of greater than about 50% by weight of

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1,4-cyclohexanedimethanol, which resin has a minimum melting point peak of about 291.0°C and which is one of a homopolymer, a copolymer, and mixtures thereof which polymers are melt blended at a temperature above about 300°C whereby the polymers are miscible in a single phase melt blend and the blend remains transparent when pelletized and molded into an article."

Claim 6 of the main request reads as follows:

"6. An article of improved color, comprising: an article which has been subjected to sterilization by ionizing radiation and which has been molded from a thermoplastic molding composition comprised of:

from 50 to 99% by weight of at least one copolyester-carbonate resin which is a reaction product of at least one dihydric phenol, at least one carbonate precursor, and at least one aliphatic alpha omega dicarboxylic acid or ester precursor thereof, wherein the at least one aliphatic alpha omega dicarboxylic acid or ester precursor has from 9 to 40 carbon atoms and is present in the copolyester-carbonate in quantities ranging from 2 to 30 mole % based on the dihydric phenol; and

from 1 to 50% by weight of at least one polyester resin which is a condensation product of at least one diacid and at least one polyol in which the at least one diacid is comprised of greater than about 50% by weight of terephthalic acid and the at least one polyol is comprised of greater than about 50% by weight of 1,4-cyclohexanedimethanol, which resin has a minimum melting point peak of about 291°C and a maximum melting point peak of about 299°C and which is one of a

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homopolymer, a copolymer, and mixtures thereof which polymers are melt blended at a temperature above about 300°C whereby the polymers are miscible in a single phase melt blend and the article is transparent."

The amendment in Claim 1 of the auxiliary request consists in the deletion of the feature "which resin has a minimum melting point peak of about 291.0°C and" in the definition of the polyester resin.

The amendment in Claim 6 of the auxiliary request consists in the deletion of the features "which resin has a minimum melting point peak of about 291°C and a maximum melting point peak of about 299°C and" in the definition of the polyester resin.

In both requests Claims 2 to 5 are directed to elaborations of the thermoplastic moulding composition according to Claim 1 and Claims 7 to 10 concern preferred embodiments of the article according to Claim 6.

- II. The reasons for that decision were non-compliance of Claims 1 and 6 of the main request with the requirements of Article 123(2) EPC and lack of inventive step of the thermoplastic moulding composition and article as defined in these claims (Article 56 EPC).
  - (i) Concerning the wording of Claims 1 and 6 of the main request, the Examining Division argued that the feature of polyesters in general having a minimum melting point peak of about 291°C was not supported by the description of the application as originally filed. The passage referred to by the

Applicant mentioned only the commercial product "PCT-3879" which designated a poly (1,4-cyclohexane-dimethylene terephthalate) resin, thus a homopolymer. The extension of that feature to any polyester represented thus an unallowable generalization within the meaning of Article 123(2) EPC.

- (ii) That particular aspect of the wording of the claims was not, however, essential for the refusal of the application. Whether one considered the independent claims of the main request or of the auxiliary request, the claimed subject-matter appeared as the obvious combination of the teaching of EP-A-542 464 (D2 hereinafter) and EP-A-465 924 (D1 hereinafter), the former being concerned with transparent articles which could be sterilized with ionizing radiation without strong yellowing moulded from compositions based on a blend of a polycarbonate and PCT, and the latter disclosing that copolyestercarbonates within the terms of Claims 1 and 6 of the application could be advantageously blended with polyesters, including PCT, resulting in compositions having an improved processability.
- III. On 29 April 1999 a Notice of Appeal against that decision was filed by the Appellant (Applicant) together with payment of the prescribed fees. The arguments submitted in the Statement of Grounds of Appeal filed on 12 July 1999 can be summarized as follows:
  - (i) The interpretation of the critical passage in the application as originally filed was disputed. Whilst that paragraph did indeed begin

with a reference to homopolymers, the next sentence was not made in relation to a particular material, but with regard to ordering materials from Eastman Chemical Company, which provided an adequate basis for a generalization of the minimum melting point peak.

- (ii) As far as inventive step was concerned, D2 taught away from the blends as claimed in that it did not recognise transparency at all levels as indicated in the present application. Although copolyestercarbonates were known from D1, a person skilled in the art would not have replaced the polycarbonate used in D2 by the copolymer, since D2 specifically mentioned that the composition should be free of copolyestercarbonates. Moreover, the ability of the polymer ingredients to form a blend miscible in a single phase spoke in favour of the inventiveness of the compositions and articles as claimed.
- IV. In a communication issued on 22 August 2000 in preparation of the oral proceedings which had been requested by the Appellant the Board expressed the preliminary view that D1 should be regarded as the closest state of the art, since it described compositions comprising a copolyestercarbonate and polyester which were both within the terms of the application in suit. The additional feature reflected in the product-by-process formulation, that the polymer ingredients be melt blended at a temperature above 300°C, as well as the advantages to be expected from a compounding of the polymers at such high temperatures were both obvious in the light of D2.

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V. During oral proceedings held on 5 October 2000 the Board, in view of the relevance of the prior art citations, suggested to postpone the discussion of the support for polyesters having a minimum melting point peak of 291°C and to decide first the question of obviousness of the general subject-matter of the application in suit.

> The arguments presented thereafter by the Appellant regarding the interpretation of the documents, the obviousness of their combination and the advantageous effects to be expected, did not shed a new light on the issue of inventive step.

VI. The Appellant requested that the decision under appeal be set aside and that a patent be granted, as main request, on the basis of Claims 1 to 10 filed on 28 April 1998, alternatively, as auxiliary request, on the basis of these claims amended according to page 23 (Claim 1) and page 25 (Claim 6) both filed on 19 October 1998.

### Reasons for the Decision

1. The appeal is admissible.

#### Procedural matter

The present appeal has not been decided by considering successively the merits of the main request and the auxiliary request, but by examining the more general question of the obviousness of the subject-matter described in the application in suit. As explained to the Appellant at the beginning of the oral proceedings, in view of the fact (i) that this subject-matter appeared to be the mere combination of the compositional features disclosed in D1 and D2 leading to the expected combination of properties, (ii) that this combination of features had to be regarded as obvious whether one started from D1 or from D2 as representing the closest state of the art, hence whatever the definition of the technical problem underlying the application in suit, and (iii) that so far nothing in the proceedings suggested that the minimum melting point peak could be decisive for the issue of inventive step, a preliminary discussion of the adequacy of the support for this parameter as formulated in the independent claims of the main request did not seem appropriate. This approach was not disputed by the Appellant.

### State of the art

- 3. The documents referred to in the present decision were discussed during the proceedings as follows.
- 3.1 D1 describes a composition comprising an admixture of (a) 1 to 95% by weight of a copolyestercarbonate and (b) 5 to 99% by weight of an aliphatic aromatic polyester, particularly a cyclohexanedimethanol (CHDM hereinafter) containing cycloaliphatic aromatic acid polyester (cf. Claims 1 and 5; page 11, lines 1 to 5). Generally from 60 to 95% by weight of copolyestercarbonate are employed (cf. page 11, line 5). This copolymer is prepared from a dihydric phenol, a carbonate precursor and an aliphatic dicarboxylic acid or water precursor having 8 to 20 carbon atoms, the latter being present in quantities from 2 to 30 mole percent based on the dihydric

phenol in order to lower the glass transition temperature while preserving the inherent physical properties of polycarbonates (cf. page 4, line 28 to page 6, line 54). Particularly preferred aliphatic aromatic polyesters derive from an aromatic dicarboxylic acid, e.g. terephthalic acid and/or isophthalic acid, CHDM and glycols, the latter compounds being suitably used in a molar ratio between 4:1 and 1:4 (cf. page 7, lines 26 to 33; page 8, lines 4 to 10). A preferred copolyester subclass is one derived from terephthalic acid, isophthalic acid and CHDM; this product is available from Eastman Chemical Co., under the trade name KODAR A 150. Another preferred copolyester sub-class is one wherein the glycol units derive predominantly from CHDM on a molar basis, the remainder from ethylene glycol (cf. page 8, lines 15 to 20).

The polymer ingredients are extruded at 260°C (cf. examples), giving rise to compositions which exhibit not only the good properties of transparency, toughness and impact resistance of polycarbonates, but additionally lower melt viscosity, thus improved processability at lower temperature (cf. page 3, lines 3 to 8 and lines 21 to 25).

3.2 D2 describes a composition comprising an admixture of (a) 82 to 95% by weight of a polycarbonate and (b) 5 to 18% by weight of a polyester derived from terephthalic acid and CHDM, known as PCT resin (cf. Claim 1). These blends are compounded by passage through an extruder at an elevated temperature above the softening points of the polymers (cf. page 3, lines 32 to 35). The experimental results in Example 3 show the influence of the composition and the temperature on the processability of the blend;

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in particular, it appears that (i) for a given processing temperature a composition of 88% by weight of polycarbonate and 12% by weight of PCT shows far superior spiral flow than a commercial formulation known as "EKTAR", which contains 50% by weight of polycarbonate and 50% by weight of a copolyester derived from terephthalic acid, CHDM (80 mole %) and ethylene glycol (20 mole %), and (ii) for a given composition the spiral flow increases with temperature, the highest values being obtained at 329°C.

The above polycarbonate compositions, which are said to be true blends of the two polymer ingredients (cf. page 3, lines 24/25), demonstrate a combination of excellent resistance to colouration upon exposure to ionizing sterilizing radiation and ease of processability in addition to excellent heat resistance and retention of impact, better mold release characteristics and flow enhancement (cf. page 2, line 53 to page 3, line 1; Example 5). This makes them readily and consistently processible into medical objects (cf. page 4, lines 9 to 13).

#### Novelty

4. Although the objection of lack of novelty raised initially in the examination proceedings (cf. communication of 22 December 1997, point II) on the basis of the disclosure of D1 was not a ground for the refusal of the application, the Board deems it appropriate to make the following comments.

Even if, for the sake of argument, one regards D1 as describing compositions comprising a

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copolyestercarbonate and a polyester both within the terms of the application in suit, the product-byprocess formulation represents in any case a distinguishing feature in Claims 1 and 6. As will appear hereinafter, the experimental data in the application in suit provide evidence that the claimed composition and articles are characterized by a superior quality of the blends which reflects the existence of a single phase melt blend achieved by processing the polymers at the required high temperature.

#### Problem and solution

- 5. The application in suit concerns a thermoplastic moulding composition and an article of improved colour moulded therefrom.
- 5.1 The wording of Claims 1 and 6 reveals that the three features which are essential for the scope of these independent claims are (a) the definition of the copolyestercarbonate, (b) the definition of the polyester, and (c) the specific melt blending process of these polymers. The above discussion of the prior art documents shows that D1 describes feature (a) in all its aspects and feature (b) as a preferred embodiment, but only a conventional melt blending process; by contrast, D2 does not deal with polyestercarbonates at all, but teaches that resistance to colouration upon exposure to ionizing sterilizing radiation and ease of processability of blends of polycarbonate with CHDM containing polyesters (feature (b)) can be improved by melt blending the polymers at specific high temperatures (feature (c)).

5.2 Although at first sight both documents would appear to be equally relevant in that they each disclose two of the three features which determine the general properties of the blends, the description of the application clearly invites to consider D1 as representing the closest state of the art. In the introduction the object of the invention is defined as the provision of a thermoplastic moulding composition and articles moulded therefrom, this composition being based on a copolyestercarbonate resin and these articles having an improved irradiation resistance compared to that of articles moulded from that copolymer alone (cf. page 6, lines 18 to 28). The experimental data in Table 1 accordingly show the changes in yellowness index one day and one week after irradiation of various blends within the terms of the application in suit (samples B to E) and, for comparative purposes, of a copolyestercarbonate alone (sample A).

> In the Board's view, there are therefore good reasons not to depart from the approach, which means that D1 qualifies as the closest state of the art.

5.3 As stated above, D1 describes moulding compositions comprising a copolyestercarbonate and a CHDM based aromatic polyester. These compositions, which are obtained by melt extrusion of the polymer ingredients, are processable at a lower temperature and their desirable physical properties makes them suitable for the manufacture of melt articles. Although in view of these properties it would be advantageous to extend the field of applications of these compositions to the manufacture of articles useful in medicine and surgery, such as containers, prosthetics and tubing, in practice this would

require receptivity to sterilization procedures, which is commonly achieved by ionizing radiation. This in turn tends to affect the normal transparency and clarity of the article, resulting in a yellowing colouration which even increases after gamma ray exposure.

- 5.4 In the light of these shortcomings the technical problem underlying the application in suit may hence be seen in the provision of a thermoplastic moulding composition and moulded articles having an improved resistance to yellowing.
- 5.5 According to the application in suit this problem is solved by a composition comprising a copolyestercarbonate and a CHDM based aromatic polyester, in which terephthalic acid represents more than 50% by weight of the diacid component and CHDM represents more than 50% by weight of the diol component, the polymer ingredients being melt blended at a temperature higher than 300°C to generate a single phase melt blend, as specified in Claims 1 and 6.
- 5.6 In view of the experimental results in Table 1 of the application in suit, which show improved colour stability one day and one week after ã-irradiation of such compositions, it is plausible that the above defined technical problem is effectively solved by the combination of features.

### Obviousness

6. It remains to be decided whether this solution can be regarded as obvious to a person skilled in the art

having regard to teachings of D1 and D2.

6.1 D1 does not contain any information concerning a possible improvement of the properties of polyestercarbonate compositions in the direction required by the technical problem.

> As stated above, regarding feature (b) emphasis is laid on the class of aliphatic aromatic polyesters derived from (i) terephthalic acid and/or isophthalic acid and (ii) CHDM and ethylene glycol in the range of from 4:1 to 4:1 (cf. page 7, lines 26 to 33; page 8, lines 4 to 10). The three sub-classes which are explicitly envisaged (cf. page 8, lines 11 to 20), namely (1) copolyesters derived from terephthalic acid and CHDM/ethylene glycol with a molar predominance of the latter, (2) copolyesters derived terephthalic acid, isophthalic acid and CHDM known under the trade name KODAR A 150, and (3) copolyesters wherein the acid units derive presumably from terephthalic acid and/or isophthalic acid and the glycol units derive predominantly from CHDM, the remainder from ethylene glycol, are described in equivalent terms. For a skilled person there would thus be no particular reason to consider the latter sub-class (feature (b)).

As to the melt extrusion temperature (feature (c)) all the examples mention the same temperature of 260°C, whether a single screw or a twin screw extruder is used, and a possible influence of that parameter on the quality of the resulting blends is not considered.

6.2 The above discussion of D2 (cf. point 3.2) makes it however obvious that a skilled person would consider

the combination of features (b) and (c), although disclosed in connection with polycarbonates, for the solution of the technical problem.

The experimental data reported in Example 3 show, first, the advantages to be expected by (i) lowering the amount of the polyester in the moulding composition and (ii) increasing the amount of CHDM in the diol component used to prepare that polyester. This means that there was an incentive to consider a polyester component within the terms of feature (b). These data also demonstrate that the resistance to yellowing upon exposure to sterilizing irradiation increases with the melt blending temperature, so that the skilled person would consider a high temperature in accordance with feature (c).

6.3 In the Statement of Grounds of Appeal the Appellant pointed out (i) that a particular advantage obtained in D2 was the absence of any substantial transesterification reaction between the polycarbonate and the PCT; (ii) that this was to be attributed to the absence of comonomer units in the polyester, and (iii) that, consequently, D2 taught away from polymer compositions based on copolymers with additional units, such as copolyestercarbonate and copolyester. This argument cannot be accepted for the following reasons.

> The critical passage in D2 referred to by the Appellant (cf. page 3, lines 6 to 11) specifies that the admixture of (a) 82 to 95% by weight aromatic polycarbonate and (b) 5 to 18% by weight CHDM based polyester is essentially free of copolymer produced by transesterification of (a) and (b) and that the polyester (b) is essentially free of a dicarboxylic

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acid and a second diol. This statement, however, does not support a general correlation between comonomer units and transesterification. As it appears from the prior art review in D2 (cf. page 2, lines 13 to 52, in particular lines 47 to 49), compositions having 10 to 90% by weight polycarbonate and 10 to 90% by weight CHDM based polyester, hence wherein both components can be regarded as homopolymers, contain 1 to 50% by weight of these polymers in a complex form resulting from transesterification. Conversely, the same prior art review mentions documents which describe blends of a polycarbonate and a CHDM based polyester derived from a mixture of aromatic dicarboxylic acids and/or an additional glycol, without apparently any transesterification. That transesterification is not directly related to the presence of comonomer units in the polymers is also evident from D1, which discloses blends of a copolyestercarbonate and a polyester derived from terephthalic acid and/or isophthalic acid, CHDM and ethylene glycol; although both polymer components are copolymers, there is no mention of transesterification in the whole document. In fact, as explained in D2 itself (cf. page 3, lines 27 to 29), the cause of transesterification is to be found in the method of preparation of the polyester, which involves the use of a transesterification reaction catalyst, with the consequence that residual amounts of catalyst may remain in the polyester; in that case, the copolymer formation initiated by these catalytic residues may be simply inhibited by quenching the transesterification reaction catalyst by the addition of a conventional quenching agent. It is therefore not surprising the find a phosphite or phosphorous acid, both well known quenching agents, in the compositions processed in D1 (cf. Examples 1

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and 2) and in D2 (cf. Examples 1 and 2), which means that the skilled person is not only aware of the risk of transesterification, but also able to cope with that phenomenon.

For these reasons the possibility of transesterification would not deter a skilled person, aware of the increased resistance to irradiation conferred to polycarbonates by the combination of features (b) and (c), from applying that combination of copolyestercarbonates.

6.4 A final point to consider is the feature "minimum melting point peak of about 291°C" characterizing the polyester resin.

> Although this parameter is not mentioned in the above documents, it is important to note (i) that the copolyester available under the trade name KODAR A 150 is particularly recommended in D1 (cf. page 8, lines 17/18), in D2 (cf. page 4, lines 3 to 5) and in the application in suit (cf. page 10, line 32 to page 11, line 3), and (ii) that the polyester used in D2 (cf. Claim 1) is the homopolymer referred to as PCT homopolymer in the application in suit (cf. page 9, line 31 to page 10, line 2). Thus, as a general feature concerning all the polyesters, in particular KODAR A 150, it is not even different from the prior art. As a specific feature concerning PCT homopolymers commercially available under the trade name PCT-3789, the evidence of any effect related to a minimum melting point peak of 291°C has not been provided; this parameter must therefore be regarded as an arbitrary value, consequently as a non-inventive feature.

This shows that the presence or the absence of this parameter has no bearing on the issue of inventive step, so that a preliminary discussion of the adequacy of its support in the application as originally filed in order to deal separately with the main request and the auxiliary request was superfluous.

6.5 It follows from these considerations that the subject-matter of Claims 1 and 6 of both the main and the auxiliary requests does not involve an inventive step within the meaning of Article 56 EPC.

## Order

## For these reasons it is decided that:

The appeal is dismissed.

The Registrar:

The Chairman:

E. Görgmaier

C. Gérardin