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D E C I S I O N
of 16 December 1993

Case Number: T 0939/90 - 3.3.3

Application Number: 84308548.1

Publication Number: 0148594

IPC: C08L 69/00

Language of the proceedings: EN

Title of invention:
Polymer compositions

Patentee:
ARCO Chemical Technology, Inc.

Opponent:
OI) BASF Aktiengesellschaft
OII) Stamicarbon B.V.

Headword:
-

Relevant legal norms:
EPC Art. 56, 114(1)

Keyword:
"Evidence - excluded at opposition stage (too late) - admitted in appeal"
"Prior art - matter incorporated by reference to another document"

Decisions cited:
T 0270/90

Catchword:
-



Case Number: T 0939/90 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 16 December 1993

Appellant:
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Representative: -

Respondent:
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Decision under appeal: Decision of the Opposition Division of the
European Patent Office of 28 August 1990, issued
in writing on 12 October 1990 revoking European
patent No. 0 148 594 pursuant to Article 102(1)
EPC.

Composition of the Board:

Chairman: F. Antony
Members: R. Young
J.A. Stephens Ofner

Summary of Facts and Submissions

- I. The grant of European patent No. 0 148 594, relating to a polymer composition comprising a polycarbonate and a copolymer of monovinyl aromatic monomer and maleimide monomer, in respect of European patent application No. 84 308 548.1, filed on 7 December 1984 and claiming a priority of 27 December 1983 (US 566064), was announced on 10 August 1988 (cf. Bulletin 88/32).
- II. Notices of Opposition were filed on the grounds of Article 100(a) EPC, alleging lack of novelty (Opponent OII) and inventive step (Opponents OI, OII), as well as on the ground of Article 100(b) EPC (Opponent OI).

The oppositions were supported *inter alia* by the following documents:

- D1: US-A-4 160 792 and
D2: US-A-3 998 907.

- III. The patent was revoked by a decision of the Opposition Division of 28 August 1990, issued in writing on 12 October 1990.

According to the decision, in which evidence filed on 22 August 1990 was disregarded for lateness and lack of relevance, the objection based on Article 100(b) EPC was not supported by the facts, and the subject-matter of the patent as amended in both the then main and sole subsidiary requests was novel. It would, however, have been obvious for the skilled person, starting from the closest state of the art document (D1) and wanting to provide further compatible polycarbonate compositions, to have tested whether terpolymers of styrene, N-phenylmaleimide and maleic anhydride known from

document D2 would be suitable for the same purpose, D1 containing a reference to D2. To have specified arbitrarily a minimum amount of maleic anhydride in the terpolymer, i.e. 2 wt%, did not render obvious matter inventive.

IV. On 7 December 1990 a notice of appeal against the above decision was filed, together with payment of the prescribed fee.

In the Grounds of Appeal filed on 20 February 1991 and a later submission dated 27 October 1993, the Appellant (Patentee) argued essentially as follows:

The "Composition B" of D1 was not in fact the same as the copolymer described in Example I of D2. Even if "Composition B" had been the same as the copolymer described in Example I of D2, however, it was wrong to have concluded that it would have been obvious to test whether the copolymer of Example VI of D2 was suitable for the same purpose as "Composition B" in D1, because there would have been no reason to expect any success. On the contrary, the reference in D1 to there being some association of the imide groups of the copolymer with the carbonate entity of the polycarbonate chain meant that there was a disincentive to the skilled person to try to replace maleimide employed in the terpolymer of D2 with an N-substituted maleimide.

Furthermore, the Appellant took the position that it was wrong to have excluded the evidence filed on 22 August 1990, and as further evidence as to the unpredictability of compatible blends, sought to introduce two further documents:

D4: "Polymer Alloys", Chemical Week, 2 May 1983,
page 72 etc. and

D5: "Heterogeneous Polymer Systems III. Phase Separation in Styrene-Acrylonitrile Copolymers", Polymer Letters, Volume 3 (1965), pages 1007-1015.

V. The Respondents (Opponents), on the other hand, argued in essence as follows:

The definition of "Composition B" in D1 referred to Example 1 of D2. It was furthermore clear from D2 that even with an excess of amine, the imidation reaction did not go to completion, so that there was always a partial retention of the anhydride function, corresponding to the terpolymers claimed in the patent in suit. Since the actual technical problem was merely to find a further alternative polycarbonate composition which was compatible, there was no requirement for any expectation of advantage in considering further alternatives; furthermore, since there was a great similarity between terpolymers with substituted maleimide or with unsubstituted maleimide, it would indeed have been obvious to test whether a substituted maleimide known from D2, viz. (N-phenyl)maleimide, would be suitable for the same purpose as the unsubstituted maleimide in D1.

The assertion that compatibility between polymers could be lost by just changing the relative concentrations had not been proved; in particular no evidence had been brought that there was a sudden change in compatibility at the point where the claimed compositions departed from the state of the art.

The Respondent (OI) further referred for the first time to two documents in support of his submissions:

D7: "Polymer-Polymer-Miscibility", O. Olabisi et al., Academic Press, New York, 1970, page 120, and

D8: "Makromoleküle", H.-G. Elias, Hüthig & Wepf,
5th Edition, Basel, 1990, page 854 et seq.

VI. At the oral proceedings held before the Board on
16 December 1993, the Appellant filed a further set of
Claims 1 to 5. Claim 1, the only independent claim,
reads as follows:

"A polymer composition comprising polycarbonate and a
copolymer of monovinyl aromatic monomer and maleimide
monomer, characterised in that said composition is a
polymer alloy wherein the polycarbonate and the
copolymer are fully compatible with one another and
comprises in weight percent, from 1 to 99 of
thermoplastic polycarbonate based on bis-(hydroxyaryl)-
alkane and from 99 to 1 of a random terpolymer of 70 to
90% by weight of recurring units of styrene, 2 to 24% by
weight of recurring units of N-phenyl maleimide, and 2
to 24% by weight of recurring units of maleic anhydride,
the total of the weight % of the three components of the
tercopolymer adding up to 100."

VII. The Appellant requests that the decision under appeal be
set aside and that the patent be maintained on the basis
of the claims submitted in the course of the oral
proceedings. These claims, although still headed
"Claims according to 2nd subsidiary request", in fact
form the main and sole request.

The Respondents request that the appeal be dismissed.

Reasons for the Decision

1. The appeal is admissible.

2. *Late filed facts and evidence*

The documents D4, D5, D7 and D8 are excluded from the proceedings under Article 114(2) EPC for lateness and lack of relevance.

As to the evidence submitted on 22 August 1990, this was, in the Board's view, rightly excluded by the Opposition Division to the extent that its lateness practically precluded the Opponents from countering it by tests of their own (cf. the decision T 270/90 of 21 March 1991, "Polyphenylene ether compositions/ASAHI", OJ EPO 1993, 725; Reasons for the decision, point 2.2, last paragraph; omitted from the published text). On the other hand, and for obvious reasons, this no longer applies to the present appeal proceedings. Consequently, on this occasion the evidence is admitted.

3. *Admissibility of amendments*

Claim 1 is supported by Claims 1, 8 and 11 of the application as filed; Claims 2 and 3 by Claims 2 and 3 respectively as filed; and Claims 4 and 5 by Claims 9 and 10 respectively as filed. The inclusion of further features has furthermore limited Claim 1.

Consequently, there are no objections to the amended claims under Article 123(2) or 123(3) EPC.

4. *Closest state of the art*

The polymer composition to which Claim 1 of the patent in suit relates is a polymer "alloy" wherein the polycarbonate and the copolymer are fully compatible, i.e. form a single phase when mixed in any proportion. Such polymer alloys are known, for instance, from the

document D1, which represents the closest state of the art.

- 4.1 According to D1, plastic alloys which contained a plurality of thermoplastic compositions had been discovered, but represented a minority. Because incompatibility remained the "dominating rule", any discovery of a useful blend represented a "pragmatic invention" which could not have been predicted on the basis of previous publications (column 1, lines 20 to 30).
- 4.2 Outstandingly advantageous properties had been discovered for copolymers of styrene and maleimide, N-methyl maleimide, maleic diamide, bis (N-methyl) maleic diamide and related compounds, designated styrene-maleimide polymers. It had been efficient to produce such copolymers by copolymerising styrene and maleic anhydride and thereafter treating the copolymer with methylamine or ammonia to obtain such styrene-maleimide copolymers. **US-A-3 998 907** [D2] described a method of preparing maleimide-containing copolymers by reacting amine or ammonia with particles of the copolymer comprising maleic anhydride under autogenous pressure at 125° to 200°C. (column 1, lines 39 to 57).
- 4.3 Thus, a thermoplastic moulding composition comprised:
- A. from about 10% to about 90% by weight of a polycarbonate,
 - B. from about 10% to about 90% by weight of a copolymer of maleimide and styrene containing from about 5% to about 35% maleimide and from about 65% to about 95% styrene (Claim 1).

4.4 According to Examples 1 to 3, Composition A was a polycarbonate prepared from the high molecular weight carbonate ester derivative of bis(4-hydroxyphenyl)-2,2-propane; Composition B was a copolymer of styrene and about 8% maleimide prepared by the method of Example 1 of **US-A-3 998 907 [D2]**, using aqueous ammonia at a temperature of about 147°C for about 12 h. at a pressure of about 5 atm. (column 2, lines 8 to 21).

4.5 There is no mention of maleic anhydride units in the copolymers of D1.

Although it cannot be said in this latter connection that the introductory word "containing" in Claim 1 of itself excludes the presence of further monomers, or that there is any specific instruction to avoid the presence of further monomers, the presentation of the percentages of only two comonomers in complementary terms up to 100%, together with the complete silence as to any further monomer, strongly implies that no further monomers are contemplated and in any case does not amount to the disclosure of any such additional monomer.

Thus the whole tenor of the text of D1 is that the copolymers it discloses consist of a maleimide and styrene, with substantially no third monomer being present.

5. *Evaluation of the reference to D2*

A crucial question in these proceedings was whether "Composition B" as defined in D1, by virtue of the reference to Example I of D2, was to be interpreted as implying a corresponding amount of anhydride units.

5.1 According to D2, modified copolymers containing an imide derivative could be prepared directly by reacting

aqueous ammonia or amines at 125° to 200°C and under autogenous pressures of between 60 and 150 psi for 0.5 to 48 h. with a modified copolymer containing an ethylenically unsaturated dicarboxylic acid, its anhydride, or a half acid derivative of the dicarboxylic acid (column 1, lines 50 to 58).

The ammonia or amines could be used in stoichiometric amounts based on the dicarboxylic acid moiety in the copolymer. However, it was preferred to use an excess of the stoichiometric amount to ensure complete conversion of the dicarboxylic acid moiety to the desired imide or N-substituted imide. Use of less than stoichiometric amounts of the amine made it possible to prepare terpolymers (column 3, lines 34 to 45).

- 5.1.1 According to Example I of D2, to which D1 refers, pellets of a copolymer of styrene and maleic anhydride containing 10.9 mole percent anhydride, were treated with aqueous ammonium hydroxide solution, in an autoclave heated to 139° to 147°C for about 12 h., during which time the pressure varied between 63 and 86 psi. The product had about 2.0 mole percent maleic anhydride groups and 8.9 mole percent maleimide groups.
- 5.1.2 According to Example III, a series of copolymers of styrene and respective amounts of maleic anhydride from 2.0 mole-% to 33.0 mole-% were heated together with water, ammonium hydroxide and a small amount of an aqueous polyvinyl alcohol solution in individual sealed bottles for 12 h.; in all cases essentially quantitative conversion to styrene-maleimide was found.
- 5.1.3 According to Example VI, a mixture produced by refluxing styrene-maleic anhydride copolymer, polyvinyl alcohol solution, aniline and water under nitrogen in a resin kettle was heated at 140°C for 12 h. under nitrogen in a

sealed bottle. The copolymer product contained 15.9 mole % N-phenylmaleimide and 2.2 mole % maleic anhydride moieties.

A second sample, after additional heating in the presence of further aniline, had essentially all of the remaining anhydride moieties converted to the N-phenylmaleimide derivative (0.5 mole % anhydride and 17.6 mole % N-phenylmaleimide).

Thus, while terpolymers containing a proportion of unreacted maleic anhydride units are disclosed in D2, it is clearly preferred to achieve a high level of conversion to imide, by using a stoichiometric excess of amine or ammonia. Indeed, according to the decision under appeal (which was not challenged on this point), the amount of residual maleic anhydride units corresponded, in the case of Example I of D2, to only 1.9 % by weight of the polymer product (see Reasons for the Decision, paragraph 7.1). Such a small residue can only be regarded as an incidental impurity.

5.2 The argument put forward at the oral proceedings, that this reference imported the whole of the disclosure of D2 into D1, and not only Example I thereof, ignores the plain language used. It is also beside the point, since even if the whole of the content of D2 were held to be "imported" in some way by this reference, it is clear that the instruction to the skilled person for the carrying out of Examples 1 to 3 of D1, is to use the method of Example I of D2, and not the method of any other Example.

5.3 The question raised by the Respondent OII at the oral proceedings as to why, if quantitative conversion had been required, the reference was not to Example III of D2, is in the Board's view speculative and therefore

irrelevant. It was, however, refuted by the unopposed statement of the Appellant, that Example III required the presence of polyvinyl alcohol which was undesirable, whereas Example I did not.

5.4 As to what is to be understood from the reference to Example I, it is noticeable that what is mentioned is the method, rather than the product, of the Example I of D2, being used.

Furthermore, whereas certain of the relevant parameters of the method (temperature, pressure and time of reaction; use of ammonia) are already specifically stated in D1, the remainder are not mentioned at all in D1. In particular, neither the nature of the starting polymer nor the concentration of the ammonia used are specified in D1. These are, however, also crucial factors determining the extent to which conversion to the imide occurs.

5.4.1 The argument that the unstated parameters must be the same as those given in D2 is not convincing, because those parameters which are explicitly stated in D1 already fall within the ranges given for the corresponding parameters in Example I of D2. Thus, there is no particular reason for regarding the remaining features with respect to which D1 is silent as necessarily being found in D2.

On the contrary, equally strong arguments exist that the unspecified parameters of the method should be construed in the light of the aims and objects of D1, i.e. to give a copolymer consisting essentially only of recurring units of styrene and a maleimide.

All in all, the wording of the reference to D2, whilst not excluding the possibility of "Composition B" of D1

and the product of Example I of D2 being identical, is nevertheless not such as to require it.

- 5.4.2 The general tenor of D1 on the other hand is in any case such as to require substantially complete conversion (cf. section 4.5, above).

Thus, D1 evidently discloses with reference to D2 a copolymer consisting essentially only of recurring units of styrene and maleimide; if any residual maleic anhydride units are present, they represent an unintentional impurity amounting to not more than 1.9 % by weight of the copolymer.

- 5.5 In view of the above, D1 cannot be regarded as disclosing a terpolymer containing essentially recurring units of styrene, maleic anhydride in significant amounts, and a maleimide.

6. *The Technical problem and its solution*

Compared with the closest state of the art, the technical problem could be seen as the search for further fully compatible polymer alloys, based on thermoplastic polycarbonate.

The solution according to Claim 1 of the patent in suit was to replace "Composition B" of D1 with a random terpolymer of 70 to 90 % by weight of recurring units of styrene, 2 to 24 % by weight of recurring units of N-phenyl maleimide, and 2 to 24 % by weight of recurring units of maleic anhydride, the total weight of the three components adding up to 100 %.

- 6.1 From the information given in the Examples of the patent in suit, especially Examples VII and IX, it can be seen that, as the amount of styrene falls below 70 wt%

(corresponding to an excess of combined maleic anhydride and maleimide units), the full compatibility shown by the terpolymer of Example VI gives way to only partial compatibility.

On the other hand, the lower limit of 2 % by weight on each of these latter monomers evidently defines a minimum level at which their presence can be regarded as significant in the terpolymer.

- 6.2 The argument, put forward at the oral proceedings, that contents of 2 % were so low as to encounter problems of experimental detection was an assertion unsupported by any evidence, and indeed directly contradicted by the figures given in D1.

It is thus credible that the claimed product provides an effective solution of the technical problem.

7. *Novelty*

The solution of the above technical problem differs from the closest state of the art document D1 essentially in two respects.

- 7.1 Firstly, as established in section 5 above, "Composition B" of D1 cannot be regarded as a terpolymer containing significant amounts of anhydride units.
- 7.2 Secondly, Example I of D2, to which D1 refers, discloses unsubstituted maleimide as the imide monomer, whereas the derivative called for by the solution to the technical problem is N-phenyl maleimide.

Thus novelty is established over the disclosure of D1.

- 7.3 There is no disclosure in D2 of polymer alloys, let alone of such alloys containing polycarbonate.

Thus novelty is also established over the disclosure of D2.

Consequently, the subject-matter of Claim 1 is novel.

8. *Inventive step*

The next question to be decided is whether the skilled person, starting from D1, would have perceived that further fully compatible polycarbonate-based compositions could be obtained by replacing the styrene/unsubstituted maleimide copolymer ("Component B" in D1) by a terpolymer of styrene with significant amounts of N-phenyl maleimide and maleic anhydride units.

- 8.1 There is no incentive in D1 itself to make either of the modifications forming the solution to the technical problem, since D1 discloses neither N-phenyl maleimide nor, indeed, terpolymers (cf. section 5.5 above).
- 8.2 Whilst it is true that D2 discloses terpolymers in general terms (cf. section 5.1 above, second paragraph), and furthermore, in Example VI, polymer products containing styrene, N-phenyl maleimide and maleic anhydride units, there is no particular reason why the skilled person should have regarded these as suitable candidates for fully compatible blends, i.e. polymer alloys, with polycarbonate. In this connection, it was generally accepted in the art that no method was available for predicting polymer compatibility on the basis of the properties of the individual polymers (e.g. solubility in a mutual solvent).

- 8.2.1 Although this principle was strongly contested by the Respondents at the oral proceedings, they failed to substantiate their argument by concrete evidence.

On the contrary, the principle of unpredictability is clearly set out in the introductory description of D1 (column 1, lines 15 to 31) and furthermore supported by the evidence of Bi Le Khac filed on 22 August 1990, which showed that two copolymers of the same monomers (styrene with 7.9 % and 14 % by weight respectively of maleic anhydride) were not miscible (compatible) at a 50/50 mixing ratio.

- 8.2.2 The argument that unsubstituted maleimide is very similar to substituted maleimide was also an assertion unsupported by any evidence. It was in any case contradicted by the evidence of Bi Le Khac, referred to above, which shows that similarity is not an index of compatibility.
- 8.2.3 Although a number of styrene-maleimide polymers are listed in D1 as having "outstanding properties", neither N-phenyl maleimide nor, indeed, any N-aromatic substituted imide is mentioned (cf. section 4.2, above). For this reason also the reference to "related compounds" cannot be taken as a pointer to the claimed monomers.
- 8.2.4 Furthermore, the relevant Example VI requires the presence of polyvinyl alcohol, which in the case of Example III was apparently a sufficient reason for not adopting its teaching (cf. section 5.3, above).
- 8.2.5 The argument that no expectation of advantage was necessary given the statement of the technical problem in terms of "further compatible compositions" is unconvincing, since the expected advantage in this case

would be that of compatibility - the latter quality being, however, as pointed out above - unpredictable.

Thus there was no pointer in D1 or D2 which would have led the skilled person to combine their disclosures in any other way than that specifically taught in D1 - namely by using the method of Example I of D2. This would not, however, have resulted in a solution of the technical problem.

8.3 Even if, in spite of the lack of pointers, the skilled person were nevertheless to have combined other parts of the disclosure of D2 with that of D1, the question arises as to what the result would have been.

8.3.1 It is to be noted in this connection that the relevant Examples of D2 (I, III, VI) all disclose an amount of residual maleic anhydride which varies in the range **below 2 %** by weight. This applies particularly in the case of Example VI where two stages of the reaction are given. In the first, the amount of residual maleic anhydride in the product was 2.2 mole %. This corresponds, according to the decision under appeal, to 1.8 % by weight (see Reasons for the Decision, paragraph 7.1, penultimate section). In the second stage, in which essentially all the remaining anhydride moieties had been converted to the N-phenyl maleimide derivative, the amount was 0.5 mole %.

Thus even if it had been obvious to the skilled person to try to blend polycarbonate with a N-phenyl derivative of styrene-maleimide copolymer according to Example VI, rather than according to Example I of D2, and even if the conditions of reaction had been exactly as disclosed in D2, the amount of residual maleic anhydride would still not have reached the 2 wt% threshold required by Claim 1 of the patent in suit.

8.3.2 Since furthermore there was no teaching in D1 that the presence of the third monomer was desirable, the skilled person would have had no incentive to do anything other than to try to ensure complete conversion to the maleic anhydride derivative. This would mean that he would have tended to choose the second stage product of Example VI, i.e. the one in which "essentially all" the remaining anhydride moieties had been converted to the N-phenyl maleic anhydride derivative, the amount of residual maleic anhydride then being far below the minimum required by the solution of the technical problem.

Consequently, the result of using the product of Example VI of D2 in "Composition B" of D1 would not have been something falling within the scope of Claim 1 of the patent in suit.

8.4 The argument of the Respondents that no evidence had been brought to show that there was a sudden change in compatibility at the point where the claimed compositions departed from the prior art ignores the fact that they are not a selection from the latter. All that is necessary, therefore, is for it to be credible that the technical problem is solved over the whole of the range claimed. This has, however, been established on the basis of the information in the patent itself (cf. section 6.2, above, last sentence).

The onus of proof was in any case on the Respondents at this stage which they have not discharged by evidence.

8.5 Neither can the Board concur with the argument in the decision under appeal, according to which "To specify an arbitrary minimum amount of MAA [maleic anhydride] in the terpolymer, i.e. 2 w%, does not render obvious matter inventive" (see Reasons for the Decision, paragraph 7.1, last sentence).

Not only was the argument based on a premise that had yet to be proved (namely that the limit was arbitrary) but the latter was unsupported by any evidence, or indeed any reasoning at all.

On the contrary, the limit of 2 %, far from being arbitrary, expresses an important aspect of the invention distinguishing it from the prior art, namely the essential presence of significant amounts of a third monomer (cf. sections 5.5, 6.1 and 7.1 above).

Consequently, the solution of the technical problem does not arise in an obvious way from the state of the art.

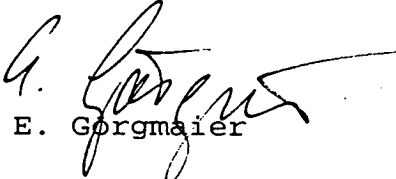
9. The subject-matter of Claim 1 is therefore not only novel but also involves an inventive step. The remaining claims, which are all directly or indirectly dependent on Claim 1, are by the same token directed to subject-matter which is both novel and based on an inventive step.

Order

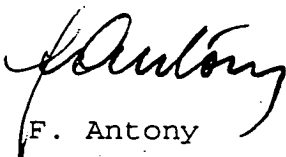
For these reasons, it is decided that:

1. The Opposition Division's decision is set aside.
2. The case is remitted to the Opposition Division with the order to maintain the patent with the claims as submitted in the course of oral proceedings with consequential amendments to the description.

The Registrar:


E. Gorgmaier

The Chairman:


F. Antony