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D E C I S I O N
of 29 September 2000

Case Number: T 0394/98 - 3.3.3

Application Number: 92311455.7

Publication Number: 0550206

IPC: C08L 71/12

Language of the proceedings: EN

Title of invention:

Polyphenylene ether/polyamide compositions

Applicant:

GENERAL ELECTRIC COMPANY

Opponent:

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Headword:

-

Relevant legal provisions:

EPC Art. 56

Keyword:

"Inventive step - obvious selection"

Decisions cited:

-

Catchword:

-



Case Number: T 0394/98 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 29 September 2000

Appellant: GENERAL ELECTRIC COMPANY
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Decision under appeal: Decision of the Examining Division of the
European Patent Office posted 31 October 1997
refusing European patent application
No. 92 311 455.7 pursuant to Article 97(1) EPC.

Composition of the Board:

Chairman: C. Gérardin
Members: P. Kitzmantel
A. Lindqvist

Summary of Facts and Submissions

- I. This appeal, which was filed on 4 December 1997, lies against the decision of the Examining Division dated 31 October 1997, refusing European patent application No. 92 311 455.7 filed on 15 December 1992 in the name of GENERAL ELECTRIC COMPANY, and published under No. 0 550 206. The appeal fee was paid together with the Notice of Appeal and the Statement of Grounds of Appeal was filed on 6 March 1998.
- II. The decision under appeal was based on Claims 1 and 12 to 25 filed with a submission dated 12 December 1995 and on Claims 2 to 11 as originally filed.

Independent Claims 1, 22 and 25 read as follows:

"1. A thermoplastic composition comprising:

- a) polyphenylene ether resin;
- b) polyamide resin, the weight ratio of said polyphenylene ether resin to said polyamide resin being from 20:80 to 80:20;
- c) an effective amount of a compatibilizing agent for components (a) and (b); and
- d) a radial block copolymer which comprises 60% to 95% by weight polymerized vinyl aromatic material, and 40% to 5% by weight polymerized conjugated diene monomer, said copolymer having at least three polymer

chains which form the radial configuration, said radial block copolymer being present at a level of from 1% by weight to 30% by weight based on the weight of the entire composition."

"22. A thermoplastic composition comprising:

- I) a compatibilized blend of polyphenylene ether resin and polyamide resin the weight ratio of said polyphenylene ether resin to said polyamide resin being from 20:80 to 80:20, and

- II) a radial block copolymer which itself comprises 60% to 95% by weight polymerized vinyl aromatic material, and 40% to 5% by weight polymerized conjugated diene monomer, said radial block copolymer having at least three polymer chains which form the radial configuration, each chain terminating with a substantially non-elastomeric segment, said radial block copolymer being present at 1% by weight to 30% by weight based on the weight of the entire composition."

"25. A thermoplastic composition consisting essentially of:

- a) polyphenylene ether resin;

- b) polyamide resin, the weight ratio of said polyphenylene ether resin to said polyamide resin being from 20:80 to 80:20,

- c) an effective amount of a compatibilizing agent for components (a) and (b); and
- d) a radial block copolymer which comprises 60% to 95% by weight polymerized vinyl aromatic material, and 40% to 5% by weight polymerized conjugated diene monomer, said copolymer having at least 3 polymer chains which form the radial configuration, said radial block copolymer being present at a level of from 1% by weight to 30% by weight based on the weight of the entire composition."

The further claims were, respectively, dependent on Claim 1 (Claims 2 to 21) and on Claim 22 (Claims 23 and 24).

III. The decision under appeal held that the subject-matter of Claims 1 to 25 did not meet the requirement of Article 56 EPC, because it was an arbitrary and/or obvious selection from the prior art as represented by

D1: WO-A-86/02086 and

D2: WO-A-88/06173.

Since it was known from these documents that the physical properties, including impact strength and processability, of blends of polyphenylene ether resin and polyamide resin (hereinafter "PPE-PA blend") could be improved by the addition of rubbery radial (tele)block copolymers (hereinafter "RB copolymers"), which are unspecified with regard to their content of polystyrene and rubber, it did not, in the Examining

Division's opinion, require an inventive effort to use RB copolymers having a relatively low rubber content as specified in Claim 1 of the application in suit.

This conclusion was *inter alia* based on the view that the evidence filed by the Applicant with its submission dated 12 December 1995 (Supplemental Declaration under 37 C.F.R. 1.132 of John B. Yates before the USPTO, hereinafter "Supplemental Declaration") did not show any unexpected improvement of the "inventive" compositions over prior art compositions containing RB copolymers having a higher rubber content. The higher stiffness of the "inventive" compositions and the special morphology, characterized by the presence of RB copolymer domains in the PPE phase, were to be expected.

IV. In an annex, dated 28 April 2000, to the summons to attend oral proceedings on 27 September 2000, which had been requested by the Appellant, the Rapporteur concurred with the objections of obviousness raised in the decision under appeal and also raised objections of lack of clarity against the statements in Claim 1 "an effective amount of a compatibilizing agent" and in Claim 22 "a compatibilized blend of polyphenylene ether and polyamide resin".

V. The arguments of the Appellant in the Statement of Grounds for Appeal and in the facsimile submission of 26 September 2000 (i.e. the day before the scheduled oral proceedings, received at 16:50) may be summarized as follows:

(i) Neither D1 nor D2 suggested that compositions having suitable impact strength, including at low

temperatures, good ductility, high flexural modulus and flexural strength as well as an enhanced melt flow could be produced by using RB copolymers having a low rubber content. The feasibility of these copolymers was particularly unlikely in view of the low impact strength of analogous compositions comprising high impact strength polystyrene (HIPS) having a low rubber content . The Appellant pointed in particular at the comparable high Dynatup impact strength values evidenced in the Supplemental Declaration for the compositions according to "inventive" sample 4, which used a low rubber RB copolymer, and according to comparative samples 2 and 6, which used high rubber RB copolymers.

(ii) Concerning the Rapporteur's objections of lack of clarity, the Appellant stressed that both the terms "effective amount" and "compatibilizing agent" presented no problem to the person skilled in the art.

VI. With the afore-mentioned submission of 26 September 2000 the Appellant withdrew its request for oral proceedings and asked that the proceedings be continued in writing.

VII. The Appellant requested that the decision under appeal be set aside and a patent be granted on the basis of Claims 1 and 12 to 25 filed with a submission dated 12 December 1995 and Claims 2 to 11 as originally filed.

Reasons for the Decision

1. The appeal is admissible.

2. *Procedural matters*

In view of the fact that the Appellant withdrew its request for oral proceedings (cf. point VI supra), there was no need to hold such proceedings.

Although the Appellant's conduct of the appeal proceedings has no impact on their outcome, the Board remarks that fruitful proceedings have not been favoured by the Appellant's very late reaction to the Rapporteur's extensive communication of 28 April 2000 (cf. points V and VI supra), leading to the undesirable situation that the Board came to know the Appellant's abandonment of its request for oral proceedings only on the very day for which such proceedings had been arranged at the request of the Appellant.

3. *Amendments*

Claims 1 and 22 are based on their respective original versions as well as on the statements on page 6, 4th paragraph and on page 13, lines 1 to 3 of the application as filed.

Claim 25 only differs from Claim 1 by the substitution of the term "composition consisting essentially of" for the term "composition comprising".

Claims 2 to 13, 17 to 21, 23 and 24 are as originally filed. Claims 14 to 16 differ from their original version only by deletion of the qualifications "about" used in connection with the definition of weight percentages.

The requirement of Article 123(2) EPC is therefore complied with by all claims.

4. *Clarity*

In view of the outcome of this appeal, i.e. its rejection and the consequential maintenance of the impugned decision to refuse the application because of the obviousness of the claimed subject-matter (cf. point 10 below), there is no need to decide the issue of clarity raised in the Rapporteur's communication of 28 April 2000 (cf. points IV and V(ii) supra).

5. *Prior art*

5.1 Document D1

Claim 29 of this document relates to a thermoplastic composition comprising (1) 5 to 95% by weight of a polyamide resin and (2) 95 to 5% by weight of a functionalised-polyphenylene ether compound which is the reaction product of a polyphenylene ether polymer and a compound having the general formula (i)--Z--(ii) wherein (i) is at least one polyphenylene ether-philic acyl-functional moiety, (ii) is at least one polyamide-philic moiety and Z is a divalent hydrocarbon radical linking group. Compounds having the general formula (i)--Z--(ii) may be chloroethanoylsuccinic anhydride, trimellitic anhydride acid chloride, chloroformylsuccinic anhydride and 1-acetoxyacetyl-3,4-dibenzoic acid anhydride (page 16, line 9 to page 17, line 11; Claims 5, 16, 17).

According to page 18, lines 30 to 34 rubbery-high molecular weight polymers may be added "to further

improve the physical properties ... such as impact strength, and processability". Among the many examples of such rubbery polymers a "radial teleblock copolymer of styrene and a conjugated diene" is mentioned (page 18, line 34 to page 20, line 7, especially page 19, lines 20 to 31).

According to page 20, lines 12 to 20 the rubbery polymer is preferably used in amounts of from about 5 to about 50 parts by weight based on 100 parts by weight of the PPE-PA blend.

5.2 Document D2

Claim 1 of this document relates to PPE-PA blends comprising

- (A) 100 parts by weight of a combination of (i) 5 to 95% by weight of a polyphenylene ether component and (ii) 5 to 95% by weight of a polyamide component, being a combination of an amorphous and of a crystalline polyamide,
- (B) 0.01 to 30 parts by weight of a compatibilizing agent, and
- (C) 5 to 100 parts by weight of a rubbery polymer or copolymer, or the reaction product thereof.

According to page 3, lines 5 to 37 the compatibilizing agent may be selected from the group of (i) liquid diene polymers, (ii) epoxy compounds, (iii) quinones, (iv) oxidized polyolefin wax, (v) certain organosilane compounds and (vi) certain polyfunctional compounds.

The rubbery polymers used to improve the physical properties, particularly the impact strength and/or processability, may, among other alternatives, be radial block copolymers (cf. page 21, lines 7 to 23; page 22, lines 5 to 10).

6. Novelty

The claimed subject-matter is novel over the available prior art, because neither D1 nor D2 disclose PPE-PA blends comprising a RB copolymer having the weight proportions of vinyl aromatic monomer and conjugated diene monomer specified in independent Claims 1, 22 and 25.

7. *Closest prior art*

Since D1 and D2 both relate to compatibilized PPE-PA blends, which contain rubbery radial block copolymers comprising vinyl aromatic units (cf. points 5.1 and 5.2 supra) in order to improve the physical properties of the blends such as impact strength and processability, these documents are equally suitable as starting point for the assessment of inventive step.

8. *Problem to be solved*

According to the original description (page 2, second paragraph; page 11, second paragraph; page 16, last paragraph) the problem underlying the claimed invention is the provision of a compatibilized PPE-PA composition having excellent impact strength also at low temperature, high melt flow, high tensile strength, ductility and chemical resistance.

Since there is no evidence on file allowing any conclusion regarding the ductility and chemical resistance of these compositions, the latter two properties have to be disregarded for the assessment of an inventive step.

9. *Solution of the technical problem; evidence*

9.1 As compared to the closest prior art disclosed in D1 and D2, the solution of the existing technical problem is to be seen in the use as a modifier for the compatibilized PPE-PA blend of a RB copolymer as defined in present Claim 1, which has a content of 5 to 40 weight percent of polymerized conjugated diene monomer (rubber).

9.2 Examples 1 to 3 of the original description demonstrate that compatibilized PPE-PA blends comprising RB copolymers having a content of butadiene of 27 % (cf. page 17: K-resins^(R) KR-01 and KR-04) exhibit i.a. enhanced Notched Izod Impact Strength and Tensile Elongation as compared with analogous PPE-PA blends comprising HIPS having a rubber content of 10.5 % (cf. page 17; Tables 1, 2 and 3 on pages 20, 23 and 26).

Since the PPE-PA base compositions used according to these examples also comprise 9 parts by weight of Kraton^(R)D 1102, an unsaturated styrene-butadiene-styrene linear block copolymer which necessarily has an impact on the physical properties of the blends, these examples are unable to demonstrate to what extent the RB copolymers alone contribute to the property changes of the compatibilized PPE-PA blends (cf. original description pages 17 to 26, especially page 17 last paragraph; page 18, line 17; page 21, lines 8 to 14;

page 25, line 5).

9.3 According to the Supplemental Declaration the compositions of the relevant samples ("inventive" samples 4 and 12, comparative samples 3, 6, 11, 14) only comprise one modifier component; accordingly, the impact of this single component on the properties of the compositions can be appreciated.

The relevant results can be summarized as follows:

sample	PPO	Fumaric Acid	PA66	RB copolymer
4*	45	0.7	45	KR-03 (27%rubber)
6	45	0.7	45	F-411 (72% rubber)
3	45	0.7	45	HIPS,(10% rubber)
12*	34	0.5	66	KR-03 (27%rubber)
14	34	0.5	66	F-411 (72% rubber)
11	34	0.5	66	HIPS,(10% rubber)

sample	flow in	NI ft-lb/in	NI -20F	DYN RT ft-lb/in	DYN LT	TEN% kpsi	TYS kpsi	TUS kpsi
4*	27.5	1.1	0.6	41	4	37	10.2	8.4
6	26	2.7	2.7	42	25	112	7.6	7.8
3	29	0.9	0.7	5	2	40	9.9	8.4
12*	30	2.1	1	44	7	41	10	8.2
14	29	4.1	1.8	40	23	87	7.5	8
11	30.5	1.8	1.1	10	2	31	9.9	8.5

* "inventive"

9.4 From the evidence summarized in point 9.3 supra the following conclusions may be drawn with respect to the solution of the existing technical problem, i.e. the provision of a compatibilized PPE-PA composition having excellent impact strength also at low temperature, high melt flow and high tensile strength (cf. point 8 supra):

9.4.1 The use of (low) rubber RB copolymers comprising 27 % rubber (samples 4, 12) instead of (high) rubber RB copolymers comprising 72 % rubber (samples 6, 14) causes

- no significant change of the melt flow,
- a considerable reduction of the Notched Izod Impact Strength at room temperature and at -20°F,
- no significant change of the Dynatup Impact Strength at room temperature (RT), but a significant lowering of this property at low temperature (LT),
- a significant reduction of tensile elongation, and
- a certain enhancement of tensile yield (TYS) and tensile break (TUS).

9.4.2 The use of (low) rubber RB copolymers comprising 27 % rubber (samples 4, 12) instead of HIPS comprising 10 % rubber (samples 3, 11) causes

- no significant changes of melt flow, Notched Izod Impact Strength at room temperature and at -20°F, Dynatup Impact Strength at low temperature (LT), tensile elongation, tensile yield (TYS) and tensile break (TUS), and
- a significant enhancement of the Dynatup Impact Strength at room temperature (RT).

10. *Obviousness*

- 10.1 The subject-matter of Claim 1 of the application in suit may be considered as a selection from the compatibilized PPE-PA blends according to D1 and D2, which already comprise RB copolymers from polymerized vinyl aromatic material (e.g. styrene) and polymerized conjugated diene monomer (e.g. butadiene) in unspecified amounts.
- 10.2 In view of the fact that according to D1 and D2 the object of the addition of RB copolymers was an improvement of some physical properties, including impact strength and processability, it is *prima facie* obvious to the skilled person seeking an improvement of these properties to use for this purpose any RB copolymer having any monomer ratio vinyl aromatic material/diene.
- 10.3 In this situation an invention step could only be recognized if the selection of the specific monomer ratio of the RB copolymers, which is specified in present Claim 1, is purposive, i.e. leads to an unexpected effect, not foreseeable for the skilled person.
- 10.4 However, the evidence discussed in point 9 supra is unable to demonstrate the existence of any unexpected effect.
- (i) In the first place, any such effect has to be demonstrated with respect to the closest prior art, i.e. compatibilized PPE-PA blends, which comprise RB copolymer, because, undoubtedly, the structure of any (rubbery) modifier has an impact on the properties of the ultimate blend. Thus, in order to compare like with like,

radially structured copolymers ("star polymers") should be compared.

- (ii) Furthermore, in order to appreciate the alleged effect of the choice of RB copolymers having a low rubber content, as specified in present Claim 1, they must be compared with compositions comprising RB copolymers having a higher rubber content. As set out in point 9.3 supra, such a comparison is, indeed, provided by the Supplemental Declaration.
- (iii) However, the conclusions which may be drawn from this Declaration according to point 9.3.1 supra are not supportive of the Appellant's assertion of the existence of an unexpected technical effect.
- (iv) Rather, for the skilled person, the effects achieved by the replacement of RB copolymers having a high rubber content by RB copolymers having a low rubber content are exactly those, which are to be expected by the lowering of the rubber content.

This is particularly conspicuous with regard to the impact strength data, which demonstrate a deterioration of this property at low temperatures according to the Notched Izod and the Dynatup measurement and also according to the Notched Izod measurement at room temperature. The Dynatup measurement at room temperature remains unchanged.

The higher rigidity and thus greater resistance

to deformation of the PPE-PA blends comprising a RB copolymer having a low rubber content is also reflected by the tensile data (elongation, yield and break).

Furthermore, the Appellant's assertion of an enhanced melt flow (page 2, second paragraph of the Statement of Grounds for Appeal) is at variance with the results reported in point 9.3 supra.

- (v) In the face of the considerable deterioration of the impact strength according to three measurements, the similar Dynatup impact strength data at room temperature obtained in the presence of RB copolymers having high and low rubber content cannot be accepted as evidence for the existence of an inventive step, a conclusion which was drawn by the Appellant on the contention that this similar behaviour was unexpected to the skilled person. This single deviating impact strength result rather shows that, for the measurement according to this drop weight method at room temperature, the amount of rubber in the RB copolymer is not critical.

(vi) As set out in sub-point (i) supra, the appropriate comparative prior art for the assessment of any unexpected effect resulting from the choice of low rubber RB copolymers is represented by PPE-PA blends comprising RB copolymers having a higher rubber content. The Appellant's stance, that a comparison with blends comprising low rubber modified HIPS was also appropriate, is not acceptable, because of the important structural differences between RB copolymers, in which the rubber portions are chemically bonded within the copolymer, and HIPS, in which the rubber domains are mainly dispersed in the polystyrene matrix.

However, even if accepted, this comparison would not provide evidence of an unexpected effect capable of substantiating an inventive step.

As set out in point 9.2 supra, the data in the original description are not appropriate for the demonstration of such an effect, because the tested "inventive" samples comprise a second rubber component, which prohibits a clear interpretation of the impact of the RB copolymer on the properties of the compatibilized PPE-PA blends.

Furthermore, the data afforded by the Supplemental Declaration (cf. point 9.4.2 supra), which are based on compositions not comprising such a second rubber component, do not exhibit any significant changes of melt flow, Notched Izod Impact Strength at room temperature and at -20°F, Dynatup Impact

Strength at low temperature (LT), tensile elongation, tensile yield (TYS) and tensile break (TUS). The only property, which is improved according to these data, is the Dynatup Impact Strength at room temperature (RT).

However, in view of the fact that the majority of the impact strength results shows that the replacement of low rubber HIPS by low rubber RB copolymer has little influence on this property, as is as well the case with regard to the other tested properties, the Appellant's contention of an unexpected balance of properties (submission of 26 September 2000, last paragraph) cannot be accepted.

- (vii) The Appellant's further argument that an inventive step should be recognized for the subject-matter of present Claim 1, because of the fact that the Dynatup Impact Strength at room temperature of "inventive" sample 4 is similar to that of (comparative) sample 2 can also not be accepted. The compositions according to this sample comprise the linear block copolymer Kraton^(R) D-1102, which is structurally different from the branched RB copolymers to be used according to present Claim 1 (cf. Table 1 of the Supplemental Declaration). It is not possible, therefore, to make any valid conclusions with respect to the influence on the physical properties of the PPE-PA blend of the amount of rubber in these two structurally different modifiers, because this comparison would be hampered by the overlapping influence of the different molecular configuration.

(viii) Neither can the alleged unexpected morphology of the claimed compositions (cf. point 8 of the Supplemental Declaration; point 4.3 of the decision under appeal; page 1, 2nd and 3rd paragraphs of the Statement of Grounds of Appeal; paragraph bridging pages 1 and 2 of the submission of 26 September 2000) contribute an element of unobviousness, because, in practical terms, the morphology is of no relevance for the assessment of inventive step. Rather, the morphology must be regarded as a mere explanation for the properties of the tested compositions and it is the obviousness or not of the achievement of these properties, which determines this issue.

10.5 Thus, the subject-matter of Claim 1 does not comply with the requirements of Article 56 EPC.

10.6 The same conclusion applies to the subject-matter of the further independent Claims 22 and 25, which relate to compositions comprising the same components, and a *fortiori* to the subject-matter of the dependent Claims 1 to 21, 23 and 24.

Order

For these reasons it is decided that:

The appeal is rejected.

The Registrar:

The Chairman:

E. Görgmaier

C. Gérardin