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D E C I S I O N
of 11 May 1999

Case Number: T 0038/96 - 3.3.3

Application Number: 88117949.3

Publication Number: 0314146

IPC: C08L 25/02

Language of the proceedings: EN

Title of invention:

Thermoplastic compositions based on syndiotactic polymers of styrene and polyphenylene ethers

Patentee:

ECP Enichem Polimeri S.r.l.

Opponent:

Idemitsu Kosan Co. Ltd.

Headword:

-

Relevant legal provisions:

EPC Art. 54, 56

Keyword:

"Novelty - prior generic disclosure - availability (no)"
"Inventive step - unobvious combination of compounds only recently available with unexplored properties"

Decisions cited:

T 0192/82, T 0666/89, T 0651/91

Catchword:

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Boards of Appeal

Chambres de recours

Case Number: T 0038/96 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 11 May 1999

Appellant:
(Opponent)

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Respondent:
(Proprietor of the patent)

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Decision under appeal:

Decision of the Opposition Division of the
European Patent Office posted 13 November 1995
rejecting the opposition filed against European
patent No. 0 314 146 pursuant to Article 102(2)
EPC.

Composition of the Board:

Chairman: C. Gérardin
Members: B. ter Laan
W. Moser

Summary of Facts and Submissions

I. European patent No. 0 314 146 in respect of European patent application No. 88 117 949.3, filed on 27 October 1988, claiming priority from an earlier application in Italy (2242287), was granted on 9 September 1992, on the basis of ten claims, Claim 1 reading:

"Compositions comprising:

- from 50 to 95% by weight of at least one styrene polymer having an essentially syndiotactic structure; and
- from 50 to 5% by weight of at least one polyphenylene ether,

with respect to the sum of the weights of the styrene polymer plus the polyphenylene ether."

Claims 2 to 10 referred to preferred embodiments of the compositions according to Claim 1.

On 9 June 1993 a Notice of Opposition against the granted patent was filed, in which, on the grounds set out in Article 100(a) EPC, the revocation of the patent in its entirety was requested. The opposition was, *inter alia*, supported by the following documents:

D1: EP-B-0 307 488,

D3: Figure A showing the variation of elastic modulus of various blends as a function of temperature,

D9: Polym. Prep: Amer. Chem. Soc. Div. Polym. Chem. **17**

(1), 145-159 (1976),

D10: Polymer Blends, vol. 1, pp. 215-241, Academic Press (1978),

D13: Pap. Meet. - Am.Chem.Soc. Div. Coat.Plast.Chem. **36** (1), 140-145 (1976) and

D16: EP-A-0 224 097, which was cited after the opposition time limit of nine months.

II. By a decision announced orally on 17 October 1995 and issued in writing on 13 November 1995, the Opposition Division rejected the opposition, since the arguments and documents upon which the opposition was based did not prejudice the maintenance of the patent in suit in unamended form (Article 102(2) EPC). It was held that:

(a) Regarding novelty, in D1, a prior art document under Article 54(3) EPC, which described compositions of 5-95% by weight of at least one styrene polymer having an essentially syndiotactic structure and from 5-95% by weight of specified thermoplastic resins including "thermoplastic polyethers", that generic term did not disclose the specific compound polyphenylene ether. Therefore, the claimed subject-matter was novel.

(b) As to inventive step, D9, which disclosed blends of atactic polystyrene/polyphenylene oxide in proportions corresponding to those required in the patent in suit, was considered to be the closest prior art document and taken as the starting point for assessing inventive step. The problem to be

solved was to find compositions which did not show the sharp decrease of the elastic modulus just above the glass transition temperature of known compositions, or, in other words, which had an improved E/T behaviour (elastic modulus behaviour in relation to temperature). Although D16 described syndiotactic polystyrene as well as its properties and contained a general statement of the suitability of syndiotactic polystyrene in combination with other resins for thermal and chemical resistance, there was no mention of any specific blends with polyphenylene ether. D10 disclosed compositions comprising isotactic polystyrene and polyphenylene ether, which were fully compatible and provided clear films. None of those documents, nor any of the other cited documents, taught to use syndiotactic instead of atactic polystyrene in blends with polyphenylene oxide in order to improve the E/T behaviour. Hence the claimed subject-matter was inventive.

III. On 10 January 1996 the Appellant (Opponent) lodged an appeal against the above decision and paid the prescribed fee simultaneously. The Statement of Grounds of Appeal, filed on 15 March 1996, referred to five new documents and contained, in addition to arguments concerning the issues dealt with in the decision under appeal, a Declaration by Prof. Koyama about the elastic modulus behaviour in relation to temperature of crystalline and non-crystalline polymers. In the Declaration, a document not previously cited during the opposition proceedings was also relied upon.

The Appellant, in writing and during the oral

proceedings held on 11 May 1999, argued essentially as follows:

- (a) Regarding novelty, the terms used in the patent in suit, in the light of decision T 666/89 (OJ EPO 1993, 495), did not exclude the compounds disclosed in D1, so that the claimed subject-matter was not novel. Even if, in line with the standing jurisprudence of the boards of appeal, the compositions as claimed were deemed not to be explicitly disclosed by D1, they had been made available by that generic disclosure. This resulted in double patenting, which was not in conformity with Article 125 EPC.

- (b) As regards inventive step, starting from D9, which disclosed blends of a non-crystalline atactic polystyrene with polyphenylene ether, the problem was to overcome the drawback of non-crystalline atactic polystyrene/polyphenylene ether blends, which showed a sharp decrease of the elastic modulus at a temperature just above the glass transition temperature. The behaviour of polymers in relation to their crystallinity was well-known, in particular the fact that crystalline polymers did not show the sharp decrease in elastic modulus above the glass transition temperature of non-crystalline polymers. Therefore, the E/T behaviour of crystalline polymers belonged to the common general knowledge of the skilled person. In support of that argument, reference was also made to D3 and to several documents not mentioned before in the proceedings, as well as to the Declaration by Prof. Koyama. In view of that

common general knowledge, the E/T behaviour of syndiotactic polystyrene was implicitly disclosed in these documents which described that polymer, in particular D16. In order to predict the E/T behaviour of syndiotactic polystyrene it was sufficient to be aware that it was crystalline and to know the values of its melting point and glass transition temperature. Therefore, it was obvious to replace the thermally inferior atactic polystyrene by syndiotactic polystyrene in a blend of polystyrene with polyphenylene ether as described in D9.

Starting from another document, D16, which disclosed syndiotactic polystyrene, it was not clear which was the problem solved by the patent in suit. Adding polyphenylene ether to the syndiotactic polypropylene of D16 was not excluded as a possibility by that document. The Appellant also referred to Decision T 192/82 (OJ EPO 1984, 415). Therefore, from that viewpoint too, the claimed subject-matter did not involve an inventive step.

IV. The Respondent (Proprietor), in its written and oral submissions, gave essentially the following arguments:

- (a) The Respondent protested against the late filing of a significant number of documents without any apparent necessity and requested that these documents should not be admitted to the proceedings. Also, the reference to D1 was inappropriate, since it was late published. Instead, the corresponding A-document (D26) should

be considered.

- (b) The compounds disclosed in D1 were only a general group of polymers, the polyethers, of which aromatic polyethers were a subclass, of which polyphenylene ethers again were a subclass. Therefore, the choice of polyphenylene ether amounted to a double selection. It was in accordance with standing jurisprudence of the Boards of Appeal that in such a case novelty was recognized. Decision T 666/89 (*supra*) did not apply since it referred to numerical ranges, not to groups of compounds as in the present case.
- (c) D9 was the closest prior art document. It disclosed mixtures of a non-crystalline atactic polystyrene with polyphenylene ether. The problem underlying the patent in suit was to avoid the sharp decrease in elastic modulus at a temperature just above the glass transition temperature as was the case with the mixture of D9. Although the melting point and glass transition temperature of syndiotactic polystyrene were known, its E-modulus and its shift with temperature (E/T curve) were not, let alone the properties of a mixture with polyphenylene ether, since syndiotactic polystyrene was a new polymer, the properties of which were not yet completely known at the priority date of the patent in suit. From D10 it was known to mix isotactic polystyrene with polyphenylene ether, but the mixture had to be annealed before compatibility was achieved. Therefore, none of the cited documents provided an incentive for the skilled person to substitute a

syndiotactic polystyrene for the atactic polystyrene according to D9.

- V. The Appellant requested that the decision under appeal be set aside and the patent be revoked.

The Respondent requested that the appeal be dismissed.

Reasons for the Decision

Admissibility of the appeal

1. The appeal is admissible.

Late filed documents

2. Originally, the Appellant based its opposition on 14 documents filed within the opposition period of nine months pursuant to Article 99(1) EPC. Later, three more documents were cited by the Appellant (then Opponent) and one by the Respondent (then Proprietor). Since the Opposition Division made no statement as to the contrary, and since D16 was specifically considered, those additional documents were apparently admitted into the proceedings. With its Statement of Grounds of Appeal the Appellant filed a Declaration by Prof. Koyama, which referred to yet another document, and, in addition, cited five further documents. In response, the Respondent submitted D26 and referred to three additional documents as well.

The Board duly studied the late filed documents and

came to the conclusion that they would not influence the outcome of the decision and hence are not more relevant than the documents already in the proceedings. Moreover, with the exception of D16, no specific arguments would appear to be based on any of those late filed documents, since they were only mentioned as background reference. However, in the Opposition Division's decision, D16 was considered as the document to be combined with the closest prior art for assessing the presence of an inventive step, and the Appellant based a significant part of its argumentation in appeal upon it; thus, the Respondent had, and in fact used, the opportunity to react to the arguments based on it.

D26, the corresponding A-document of the B-document cited as D1, was also late filed. Whereas D1 (publication date 22 July 1992) does not belong to the state of the art and therefore cannot be considered, the contents of D26 in its filed version (filing date 15 September 1987; published on 22 March 1989) are prior art pursuant to Article 54(3)(4) EPC for the Contracting States BE CH DE FR GB LI NL SE. Since the published version of D26 is identical to the corresponding application as filed, that instead of D1 should be considered when assessing novelty.

Therefore, of the late filed documents, only D16 and D26 are admitted to the proceedings (Article 114(2) EPC).

Novelty

3. The novelty objection was solely based upon D1, which was late published. Instead, the Board takes D26 into

consideration (see point 2 above).

3.1 D26 describes a polystyrene-based resin composition which comprises

- (a) a styrene-based polymer of which the molecular structure relative to the stereospecificity is mainly syndiotactic; and
- (b) an additive selected from the group consisting of thermoplastic resins and inorganic fillers (Claim 1).

The amount of syndiotactic polystyrene in such a composition is from 1 to 99% by weight, preferably from 5 to 95% by weight (page 3, lines 4 to 9). The thermoplastic resin can be any other resin than syndiotactic polystyrene (page 3, lines 10 to 11) and a great number of very different resins are exemplified on page 3, lines 13 to 20, amongst which also polyethers are mentioned.

The examples mention polycarbonate, polyethylene terephthalate resin, ABS resin and polysulfone resin (Examples 8 and 9). In Example 8 the syndiotactic polystyrene:polysulfone ratio is 20:80, in Example 9 it is 50:50. Therefore, the only amounts of the mixture constituents which correspond to the ones now required are those of Example 9.

3.2 The Appellant's argument was that polysulfone resins were included in the generic definition of "polyphenylene ethers" (which was a broader concept than polyphenylene oxides) of present Claim 1, so that

Example 9 of D26 disclosed the claimed subject-matter. However, the Board cannot follow that view for the following reasons:

First, according to standard nomenclature, polysulfones contain $-SO_2-$ as linkage groups, whereas polyethers are linked by $-O-$ groups. Polymers containing both groups as linkages are called polysulfonethers. That distinction in itself is sufficient indication that the polyphenylene **ethers** mentioned in Claim 1 do not encompass polysulfones. In case any doubt would remain, the reference in the patent in suit to the formula on page 3, lines 2 to 12, and the specification of which compounds are understood to fall under the definition of polyphenylene ethers on page 3, lines 28 to 37, make it clear that no polysulfone resins are envisaged.

Secondly, the listing of thermoplastic resins on page 3, lines 13 to 20 of D26 is clearly not restricted to only those polymers explicitly mentioned ("Exemplary of the thermoplastic resin ...": line 13; "... including...": lines 14, 16, 17, 18, 19; "... and the like;": lines 15, 17, 18, 19, 20; "... and so on.": line 20). Even if polysulfones as such are not mentioned, it cannot therefore be concluded that the polysulfones used in Examples 8 and 9 should be interpreted to fall under the generic term "polyethers" (page 3, line 16). Any such construction would go against both the information contained in D26 as well as standard nomenclature.

3.3 In Decision T 666/89 (supra) the question of overlapping numerical ranges with regard to novelty was dealt with. The Appellant, referring to that decision,

argued that the polyethers mentioned in D26 included the polyphenylene ethers now required. However, that statement, though in itself true, is not relevant for the issue of novelty.

In the present case, there is no question of overlap, as with the numerical ranges to which T 666/89 (supra) refers, but rather the question of whether a generic term (polyethers) discloses a more specific class of compounds (polyphenylene ethers). Since in the Board's judgement a generic term does not normally take away the novelty of any specific compound falling within that generic term (see also T 651/91 of 18 February 1993; not published in OJ EPO), and since D26 contains no clear and unambiguous indication of polyphenylene ethers as now required (see above), the Board concludes that D26 does not disclose that specific class of compounds.

3.4 Regarding the issue of "double patenting" raised by the Appellant, it is not clear upon which basis that opinion is founded since no evidence was submitted that such a situation, if it would occur, would contravene the principles of procedural law generally recognised in the Contracting States. Therefore, the Appellant's reference to Article 125 EPC is not convincing.

3.5 In the light of the above, D26 does not disclose the subject-matter of Claim 1 of the patent in suit, which is therefore novel with respect to D26.

4. The Appellant did not base its novelty objection on any other document than D1 (D26), and the Opposition Division considered the claimed subject-matter to be

novel also in view of the other documents on file. The Board concurs with that view.

Inventive step

5. The patent in suit concerns thermoplastic compositions based on syndiotactic polymers of styrene and polyphenylene ethers.

5.1 Compositions of polystyrene and polyphenylene ethers have been disclosed in D9, which the Board, like the parties and the Opposition Division, regards as the closest state of the art. D9 is a study of the mechanical properties of blends of polyphenylene oxide and polystyrene. The polystyrene is not indicated as being atactic, isotactic or syndiotactic, but is identified as Dylene 8G from Sinclair-Koppers Co (page 146, chapter "Specimen Preparation"). Both parties agreed that the polystyrene of D9 was atactic, i.e. amorphous, and the Board sees no reason to take another view. One of the mechanical properties which were actually studied was the dynamic tensile modulus, which was measured for different ratios of polyphenylene ether/polystyrene, varying from 0/100 to 100/0 (Figure 1). All these blends demonstrate a sharp decline in the modulus at a temperature just above the glass transition temperature.

5.2 Therefore, in accordance with the patent specification (page 2, lines 16 to 18), the technical problem underlying the patent in suit is to be seen in providing polystyrene/polyphenylene ether compositions having improved mechanical properties, in particular without the sharp decrease of the elastic modulus at a

temperature just above the glass transition temperature.

- 5.3 According to the patent in suit this problem is to be solved by compositions comprising specified amounts of syndiotactic polystyrene and polyphenylene ether, as defined in Claim 1.

- 5.4 The examples and comparisons with the prior art in the patent specification (Figures 1 to 3) as well as the examples filed later by both the Respondent and the Appellant (D3) show that the various aspects of the above-defined problem are effectively solved. In particular, it has been shown that the claimed syndiotactic polystyrene/polyphenylene ether compositions do not have a sharp decline in the elastic modulus at a temperature just above the glass transition temperature.

- 6. The issue to be decided, therefore, is whether the claimed subject-matter is obvious having regard to the documents on file.

- 6.1 D9 describes the basic properties of two important categories of polymer blends: incompatible ones, where the components separate in the bulk phase, and compatible ones, which are usually defined as having one single glass transition temperature and producing a clear film. The degree of mixing and the presence or absence of cosolvents play a role in compatibility (page 145, second full paragraph).

The characteristics of the polymer mixture strongly influence the dynamic-mechanical properties,

incompatible systems having multistep changes in the storage modulus in relation to different temperatures, whereas compatible systems show a drop in the storage modulus at a temperature intermediate between the glass transition temperatures of the components (page 145 and 146, chapter "The Mechanical Properties of Blends"). Mixtures of polystyrene and poly(2,6-dimethyl p-phenylene oxide) were chosen for several reasons, in particular because they are compatible over the whole range of possible compositions (page 146, second full paragraph).

Thus, the general teaching of D9 relates to the mechanical properties of the specifically defined polystyrene/polyphenylene oxide blends in relation to temperature, their compatibility and what effects compatibility has on the deformation behaviour of those blends. It is however completely silent regarding the properties of other types of blends, in particular those containing crystalline or syndiotactic polystyrene, so that a skilled person would have no information whatsoever about the features to be modified in order to solve the above-defined technical problem. Therefore, the claimed subject-matter could not be derived from D9 by itself.

- 6.2 D16 discloses a process for producing styrene polymers in which the polymer side chains are mainly in the syndiotactic configuration (page 1, first full paragraph). Until then, either mainly atactic polystyrene was produced by free radical polymerization, or mainly isotactic polystyrene by means of Ziegler catalysts (page 1, third full paragraph). D16 describes how it is possible to obtain

mainly syndiotactic polystyrenes by using a specific type of catalyst. According to the Respondent, D16 was the first patent application describing syndiotactic polystyrene and the Appellant did not oppose that statement. The products of D16 are examined by X-ray diffraction and ¹³C-NMR techniques (pages 2 to 3, chapter "Brief Description of the Drawings" in conjunction with Figures 1 to 5). Their properties are described on page 9, second full paragraph: they are either crystalline or amorphous and the crystalline styrene polymers have higher thermal resistance and better solvent resistance than atactic polystyrene and, therefore, are useful in fields where such properties are required or as modifiers blended with other resins. From the examples it appears that the melting point is higher than that of a comparable isotactic polystyrene (Example 1: 270°C vs. 220°C). However, no specific examples of resins to be blended are given, nor is there any indication of the E/T behaviour either of the syndiotactic polystyrene itself or of any blend of it.

The Board cannot accept the Appellant's argument that once a compound is described, all its properties are implicitly disclosed. Such can only be the case where known compounds - with known properties- are mentioned. The parties agreed that D16 was one of the first documents to describe syndiotactic polystyrene, and it can therefore not be supposed that all its properties were known at that time. Since D16 gives no clue regarding the properties sought to be improved in the patent in suit, the skilled person could not know what was to be expected when syndiotactic polystyrene would replace the atactic polystyrene in D9 in general, and even less so for the specific amounts required by

present Claim 1 in particular (see also T 192/82 supra).

Hence, the skilled person would not have combined D16 with D9 with a view to improving of the E/T behaviour.

6.3 A possible combination of D16 with D10 would not lead to any other conclusion. D10, which deals with blends of polyphenylene ether and isotactic polystyrene, describes such blends containing a crystalline isotactic polystyrene phase and an amorphous mixed isotactic polystyrene-polyphenylene ether phase (page 214, last paragraph). It reveals structural parameters for several of those blends, varying from 100/0 to 70/30 isotactic polystyrene/polyphenylene ether (Table III). However, although reference is made to prior art literature mentioning dynamic mechanical properties of polyphenylene ether/isotactic polystyrene blends in which the isotactic polystyrene was quenched to the amorphous state in which it resembles atactic polystyrene (page 207, third full paragraph), no reference is made to the E/T behaviour of those blends, and D10 itself does not deal with those properties. Therefore, D10 contains no teaching regarding the E/T behaviour of isotactic polystyrene/polyphenylene ether blends. Even if that property had been described, D16 contained no information regarding the properties of syndiotactic polystyrene (see point 8.3 above) that could have suggested to the skilled person to replace the isotactic polystyrene of D10 by syndiotactic polystyrene.

6.4 D13, which was relied upon by the Appellant during oral proceedings, is a study of the plastic deformation,

i.e. crazing and shear banding, of amorphous polymers below their glass transition temperature, in particular of blends of isotactic polystyrene and 2,6-dimethylpolyphenylene ether (page 142, chapter "Conclusions").

Figure 8 on page 145 shows the dynamic mechanical spectra of thick films of such blends as a function of the temperature. As pointed out by the Respondent in opposition proceedings (cf. submission of 20 July 1994, page 3, discussion of D13), this investigation is limited to a temperature range of 90 to 360 K, corresponding to a range of -183 to +87°C. Hence there is no information about the behaviour of the blends at temperatures higher than 87°C, which is the critical range of temperatures in the patent in suit, as can be seen from the examples and the figures.

A further point to consider is the behaviour of the shear to craze transition in polyphenylene ether/polystyrene thin films. As it appears from Figure 7, the most pronounced change in shear properties occurs in the 15 to 30 weight percent range of polyphenylene ether; this transition in behaviour is little affected by the tacticity of the polystyrene component - whether atactic polystyrene or isotactic polystyrene is used in the blend (page 141, fourth full paragraph). It follows that there was no incentive to consider a structural parameter of polystyrene, in particular a polystyrene with syndiotactic structure, for the solution of the above-identified technical problem.

In fact, the teaching of D13 is explicitly limited to unsubstituted polyphenylene ether and to symmetric 2,6-disubstituted polyphenylene ether (page 140, last two paragraphs to page 144, first full paragraph), which would exclude the unsymmetrically substituted polymers, contrary to the patent in suit, in which the two substituents R_1 and R_2 may be the same or different (cf. Claim 8). Furthermore, the exploration of the compatibility of the blends brought to light that the latter are amorphous (page 141, fifth full paragraph), whereas the patent in suit refers to the preparation of crystalline compositions (cf. patent specification, page 2, line 19).

For these various reasons it must be concluded that D13 does not provide an incentive to consider a solution along the lines of the patent in suit.

6.5 As to D3, to the extent that such a figure may be regarded as state of the art suitable to support an objection of lack of inventive step, even if the effect resulting from the addition of 5% by weight could be called marginal, the addition of 30% by weight has the beneficial effect that the temperature at which the elastic modulus starts to decrease is increased by more than 30°C (cf. point 5.4 above). The fact that this improvement is more pronounced for the intermediate values than for the extreme values of the range defining the weight ratio of the polymers can hardly be regarded as surprising and cannot, in the Board's view, speak against the inventiveness of the claimed subject-matter.

6.6 From the above it is clear that none of the cited

documents, taken alone or in combination, actually contains an incentive for the skilled person to use syndiotactic polystyrene for the solution of the above-defined technical problem. Even if the various elements of the present composition were all known, their properties were not, so that there was no reason for the skilled person to select the specific combination of features as now claimed with a view to improving the E/T behaviour of the composition.

Therefore, the Board comes to the conclusion that the combination of features required in Claim 1 in order to provide an improved E/T behaviour in polystyrene/polyphenylene ether blends in accordance with the object underlying the present invention, was not obvious in the light of the available prior art, and, therefore, involves an inventive step.

7. As Claim 1 is allowable, the same applies to Claims 2 to 10, which are directed to preferred embodiments of Claim 1 and derive their patentability from that of Claim 1.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar:

The Chairman:

E. Görgmaier

C. Gérardin