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D E C I S I O N
of 11 March 1994

Case Number: T 0219/91 - 3.3.3

Application Number: 81 105 454.3

Publication Number: 0 069 792

IPC: C08F 287/00

Language of the proceedings: EN

Title of invention:
Transparent impact resin and process for the preparation thereof

Patentee:
The Dow Chemical Company

Opponent:
BASF Aktiengesellschaft, Ludwigshafen

Headword:
-

Relevant legal norms:
EPC Art. 83

Keyword:
"Sufficiency of disclosure (affirmed)"
"Counter-experiments not *bona fide* attempt to reproduce the teaching of the patent"

Decisions cited:
-

Catchword:
-

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Beschwerdekammern | Boards of Appeal | Chambres de recours

Case Number: T 0219/91 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 11 March 1994

Appellant: (Opponent)
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Representative:
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Decision under appeal:
Interlocutory decision of the Opposition Division of the European Patent Office of 18 December 1990, issued in writing on 1 February 1991 concerning maintenance of European patent No. 0 069 792 in amended form.

Composition of the Board:

Chairman: F. Antony
Members: R. Young
W.M. Schar

Summary of Facts and Submissions

I. The mention of the grant of European patent No. 0 069 792 in respect of European patent application No. 81 105 454.3, filed on 13 July 1981, was announced on 21 September 1988 (cf. Bulletin 88/38).

II. Notice of Opposition was filed on 15 June 1989 on the grounds of Article 100 (a) and (b) EPC. The Opposition was supported by the document:

D1: A. Echte *et al.*, Angew. Makromol. Chem. 90, 95-108 (1980).

III. By a decision which was given at the end of oral proceedings held on 18 December 1990 and issued in writing on 1 February 1991, the Opposition Division found that the patent could be maintained in amended form on the basis of the text filed with the letter dated 23 October 1990, and comprising two claims, which read as follows:

"1. An impact resistant alkenyl aromatic resinous composition in the form of a matrix of a polyalkenyl aromatic monomer having therein a block copolymer, said resinous composition being obtained by free radical polymerization of the alkenyl aromatic monomer in the presence of the block copolymer, the block copolymer having a weight average molecular weight of at least 40 000 and having the AB or $AB(BA)_n$ configuration, wherein A represents a block of polymerized alkenyl aromatic monomer and B represents a block of polymerized conjugated diene monomer and n is an integer from 1 to 5, characterized in that,

a) the composition is transparent and contains from 70 to 85 weight percent of alkenyl aromatic units and from 30 to 15 weight percent of conjugated diene units polymerized therein,

b) the weight average molecular weight of the polyalkenyl aromatic monomer matrix lies in the range of from 75 000 to 350 000,

c) the block copolymer is contained in the matrix as a plurality of domains in the form of rods and spheres having diameters less than 70 nanometers and no occlusions being observable by electron microscopy,

d1) the block copolymer contains from 20 to 70 weight percent of polymerized alkenyl aromatic monomer and from 80 to 30 weight percent of polymerized butadiene therein or

d2) the block copolymer contains from 35 to 50 weight percent of polymerized alkenyl aromatic monomer and from 65 to 50 weight percent of polymerized isoprene, which optionally can be partially replaced by butadiene, and

e) when the block copolymer used is a copolymer comprising butadiene, crosslinking between the polymerized diene blocks has been prevented by addition of an antioxidant at or near the end of the polymerization.

2. A method for the preparation of an impact resistant alkenyl aromatic resinous composition by the free radical polymerisation of at least one alkenyl aromatic monomer in the presence of a block copolymer and forming a matrix of polyalkenyl aromatic monomer having therein the block copolymer having a weight average molecular weight of at least 40 000 and having the AB or $AB(BA)_n$ configuration, wherein A represents a block of polymerized alkenyl aromatic monomer and B represents a block of polymerized conjugated diene monomer and n is

an integer from 1 to 5, characterized by forming a transparent composition containing from 70 to 85 weight percent of alkenyl aromatic unit and from 30 to 15 weight percent of conjugated diene units polymerized therein, providing a weight average molecular weight of the polyalkenyl aromatic monomer matrix of from 75 000 to 350 000 having therein the block copolymer as a plurality of domains in the form of rods and spheres having diameters less than 70 nanometers and no occlusions of polymerized alkenyl aromatic monomers being observable by electron microscopy and

i) the block copolymer contains from 20 to 70 weight percent of polymerized alkenyl aromatic monomer and from 80 to 30 weight percent of polybutadiene therein or

ii) the block copolymer contains from 35 to 50 weight percent of polymerized isoprene, which optionally can partially be replaced by butadiene, and when the block copolymer used is a copolymer comprising butadiene, preventing crosslinking between the polymerized diene blocks by adding an antioxidant at or near the end of the polymerisation."

According to the decision, the subject matter claimed was sufficiently described because data were given on all the essential variables (the microstructure and transparency of the composition being technical signals contributing to a functional definition), and the skilled person would be capable, on the basis of the worked examples, of producing further, different compositions falling within the teaching of the patent. The claimed subject matter was novel, since the features (i) that the composition contained 15 to 30 wt% of conjugated diene units, and (ii) the diene was isoprene, or (iii) was butadiene and an antioxidant added at or near the end to avoid cross-linking of butadiene units, had not been identified in the state of the art. Since,

furthermore, these evidently essential features could not be readily derived from the state of the art, the claimed subject matter also involved an inventive step.

IV. On 16 March 1991 a Notice of Appeal against the above decision was filed, together with payment of the prescribed fee. The Statement of Grounds was filed on 4 June 1991.

The arguments of the Appellant (Opponent) presented in writing and during the oral proceedings of 11 March 1994 can be summarised as follows:

(i) The transparency characterising feature (a) of Claim 1 did not correspond to any technical teaching and therefore was not appropriate for distinguishing from the state of the art.

(ii) The block copolymers used in the examples were not further characterised, even though it was common knowledge that their composition and structure had a considerable influence on the transparency. In particular there was no information given as to the length of the blocks. Regarding the influence of block length on transparency, reference was made for the first time to the document: DE-B-1 959 922 (D2).

(iii) Comparative tests showed that greatly differing transparencies could be obtained:

According to an experimental report and photomicrographs filed with the submission dated 22 May 1992, the product of polymerising styrene in the presence of a styrene/butadiene (40/60) block copolymer using a peroxide initiator, although showing the rod and sphere morphology

illustrated in the patent in suit, was not transparent but at best translucent. It could not be cast into a layer 0.25 mm thick because of the high level of specks. It was furthermore exceptionally brittle, so much so that no tensile or impact measurements could be made.

As foreshadowed in the letter accompanying the report (last para.), a sample was produced at the oral proceedings, which, although only 0.1 mm thick, was not transparent. Since, however, the experimental work had been done with the necessary skill, it was evident that the promise of the patent was not fulfilled by the claimed measures.

Thus the patent in suit either described something that did not work, or did not sufficiently describe what had to be done to get a simultaneously impact resistant and transparent modified polystyrene.

V. The Respondent (Patentee) on the other hand argued essentially that:

(i) The term "transparent" was well known and correctly used in comparison with prior art impact resistant polystyrenes, which did not have sufficient transparency. The problem underlying the invention was thus to provide a more transparent impact resistant styrenic resin. That the problem was indeed solved was evident from a comparison of the examples with the comparative tests of the patent in suit.

(ii) As to objection to the missing data on the block lengths, sufficient information was given in the claims, description and 17 examples of the

patent in suit which would enable the skilled person without difficulty to duplicate the subject matter of the patent in suit.

(iii) The cited document D2 differed in many respects from the claimed subject matter; it could not serve as prior art relevant to the questions of novelty or inventive step in the present case.

(iv) The fact that the Appellant's experimental report disclosed the claimed morphology was itself evidence for the sufficiency of the description, since transparency was dictated by the refractive indices of the phases and the dimensions of the phase domains. Any deficiency must therefore be traceable to inhomogeneities arising from the Appellant's experimental technique. In this connection, differences were pointed out, in particular the employment, in the Appellant's experiment, of t-dodecylmercaptan, which was a very powerful chain transfer agent, in quantities similar to those of the much weaker agents used in the Examples of the patent in suit.

VI. At the oral proceedings, the Respondent filed amendments to the claims and description. The documents submitted comprised five amended sheets corresponding to pages 2 to 5 and 7 respectively of the printed specification (the latter page including Claims 1 and 2) and page 6 (Table II) which by implication remained unamended.

The amended claims differ from those underlying the decision under appeal in the following way:

Features d1) and e) of Claim 1 now read respectively as follows, the deleted items being shown in square brackets and the newly inserted ones by underlining:

"d1) the block copolymer contains from [20] 39 to 70 weight percent of polymerized alkenyl aromatic monomer and from [80] 61 to 30 weight percent of polymerized butadiene therein or...."

"e) when the block copolymer used is a copolymer comprising butadiene, the block copolymer having a weight average molecular weight from 40,000 to 150,000, crosslinking between the polymerized diene blocks has been prevented by addition of an antioxidant at or near the end of the polymerization."

Similarly, features i) and ii) of Claim 2 read respectively as follows:

"i) the block copolymer having a weight average molecular weight from 40,000 to 150,000, [contains] containing from [20] 39 to 70 weight percent of polymerized alkenyl aromatic monomer and from [80] 61 to 30 weight percent of polybutadiene therein or...."

"ii) the block copolymer contains from 35 to 50 weight percent of polymerized alkenyl aromatic monomer and from 65 to 50 weight percent of polymerized isoprene, which optionally can partially be replaced by butadiene, and when the block copolymer used is a copolymer comprising butadiene, preventing crosslinking between the polymerized diene blocks by adding an antioxidant at or near the end of the polymerisation."

VII. The Appellant requests that the decision under appeal be set aside and the patent in suit be revoked.

The Respondent requests that the appeal be dismissed and that the patent be maintained on the basis of the amended claims and description filed during the oral proceedings.

Reasons for the Decision

1. The appeal is admissible.

2. *Formal admissibility of amendments*

2.1 Claim 1 is supported in its precharacterising portion by original Claims 1, 2 and 7 in conjunction with page 2, lines 11 to 17 (polymerization in the presence of block copolymer) and page 4, line 10 (matrix component). In the characterising portion, support is to be found in the original version for the features (a) to (e) as follows:-
(a): Claim 1 (transparent) and page 3, lines 6 to 9;
(b): page 4, lines 10 to 13;
(c): Claim 1 in conjunction with Example 2, last but one sentence (electron microscopy);
(d1): Claim 4 in conjunction with Example 2, Table I (second vertical column, third run);
(d2): Claims 5 and 6 in conjunction with Example 3, Table II (first and last runs);
(e): Claim 8 in conjunction with page 5, lines 2 to 5, and page 3, lines 4 to 6.

2.2 The features of Claim 2 find the same support in the application as filed as do those of Claim 1.

2.3 The amendments to the description reflect those made in the claims.

2.4 No objections therefore arise under Article 123(2) or 123(3) EPC to the claims or description of the sole request.

Neither, incidentally, were there any objections to the formal admissibility of the amendments on the part of the Appellant.

3. *Sufficiency*

The patent in suit is concerned with transparent impact resistant resins and processes for the preparation thereof, in particular impact resistant polystyrene having a two phase system of a polystyrene matrix with embedded particles of a styrene-butadiene block copolymer, obtained by free radical polymerisation of styrene monomer in the presence of the block copolymer.

3.1 Although it was not disputed that it was possible to obtain transparent resins by free radical polymerisation, it was disputed that it would always be possible to achieve transparency when working according to the claims of the patent in suit. The sample shown by the Respondent at the oral proceedings was cited as evidence of such a deficiency.

3.1.1 Since, however, it turned out on further enquiry that the sample displayed had not in fact been produced by the method set out in the experimental report filed with the submission dated 22 May 1992, but was a "technical product" typically in use twenty years before the advent of the patent in suit, the Board has not had an opportunity to inspect any relevant sample.

It is therefore limited in its assessment to the description in the report itself.

3.1.2 According to this description, the experimental procedure used differed in a number of respects from that adopted in the Examples of the patent in suit. For instance, two stages of heating for removal of volatiles were reported instead of one. No details were given, however, of how much volatiles were removed, whereas the Examples of the patent in suit specified how much butadiene remained. Furthermore, and most significant for the Board, a different chain transfer agent (t-dodecylmercaptan) was used, which, in spite of being five times more powerful than that used in the Examples of the patent in suit (alphamethylstyrene dimer), was nevertheless used in quantities similar to those of the much weaker agent.

In spite of all these differences, however, it was admitted in the report that the rod and sphere morphology described in the patent in suit had nevertheless been obtained (cf. report, page 2, lines 1 to 3).

3.1.3 Since, on the one hand, polystyrene homopolymer is itself transparent, and on the other, the morphology of the block copolymer obtained (rod and sphere diameters less than 70 nm) corresponded to a particle size less than half the wavelength of visible light (the latter being of the order of 400 nm), it is clear that the prerequisites for transparency were already present.

3.2 The question thus arises of why the product obtained according to the report was not in fact transparent.

3.2.1 In the Board's view, it belongs to the general knowledge of the skilled person that, when attempting to reproduce the teaching of a patent specification, the most appropriate starting point is the illustrative Example or Examples. In the present case, there are over a dozen such examples. Nevertheless, the Appellant did not choose to follow any of them.

3.2.2 On the contrary, the Board is struck by the divergences in the experimental technique described in the experimental report of the Appellant from that of the Examples of the patent in suit, in particular as regards the number of heating steps and the choice and amount of chain transfer agent (cf. section 3.1.2, above).

3.2.3 The latter, according to the uncontested submission of the Respondent during the oral proceedings, would have led to the formation of a very low molecular weight - and therefore very weak - polystyrene matrix. Furthermore, overheating or the use of impure components or less than clean apparatus could itself result in the formation of specks. The fact that there were two heating steps instead of one would thus have increased the risk of loss of transparency.

Hence, the Appellant not only failed to make full use of the information disclosed in the Examples of the patent in suit, but instead diverged, evidently gratuitously, from their teaching in a number of respects, each of which had a potentially negative significance for the quality of the product obtained, in particular as regards its strength and transparency.

3.2.4 Furthermore, since, in the Board's view, the skilled person would inevitably have been aware of these negative tendencies, the Board is not satisfied that a *bona fide* attempt was made to reproduce the teaching of

the patent in suit. Consequently, no criticism can attach to the Respondent for not having repeated such a procedure.

3.2.5 On the other hand, considering that in spite of the divergencies, the essential precondition for transparency (the rod and sphere morphology) was nevertheless met, the Board cannot regard the results obtained, even if less than optimal, as evidence of insufficient disclosure or of a failure of the claimed measures to fulfil the promise of the patent in suit. On the contrary, the Patentee is entitled to protect less preferred embodiments.

3.2.6 Since, furthermore, the large number of illustrative examples given in the description of the patent in suit all show that the level of light transmission through samples 0.254 mm thick was of the order of 99.9%, corresponding to a very high level of transparency, as well as exhibiting the required rod and sphere morphology, the Board has no reason to doubt that the skilled person is provided with sufficient instructions for obtaining a transparent polystyrene according to the patent in suit.

3.2.7 In this connection the argument concerning the use of the term "transparent" (cf. section IV (i), above) cannot succeed because it has neither been shown that the whole claim when read in the light of the description is deficient in technical teaching nor that the claim depends for its distinction from the prior art on the transparency feature alone (cf. section IV, below). In any case no objectionable unclarity arises.

3.2.8 As regards the question of block length (section IV(ii) above), not only the block configuration and overall molecular weight, but also the manner of preparation of

the block copolymers is specified in the Examples of the patent in suit (cf. especially Tables I and II). Consequently, it is evident that all the necessary details have been disclosed.

3.2.9 Finally, the argument put at the oral proceedings that a more sophisticated manner of adding the particulate phase was necessary to ensure that the refractive indices of the polybutadiene and the matrix phase were the same, and that this had not been described in the patent in suit, is unconvincing. The fact that the claimed morphology corresponds to a particle size of less than half the wavelength of visible light means that incident radiation will not be scattered by the particles whatever their refractive index in bulk.

Consequently the requirements of sufficiency are met, and the ground of opposition under Article 100(b) must fail.

4. *Novelty and inventive step*

Although a brief reference appeared at the end of the Grounds of Appeal that the submissions made in the opposition proceedings had been maintained, no further details were given. In essence, thus, no arguments relating to the novelty or inventive step of the claimed subject matter were put forward, the appeal being essentially based on Article 100(b) EPC arguments.

The decision under appeal found that the two claims then under consideration, which were rather broader in scope than the present claims, related to subject matter which was both novel and involved an inventive step (see Reasons for the decision, paragraphs 4 and 5). The Board

sees no reason to differ from this conclusion, which is furthermore regarded as applying *a fortiori* to the present (narrower) claims.

Consequently, the subject matter of Claims 1 and 2 is both novel and involves an inventive step.

Order

For these reasons, it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the first instance with the order to maintain the patent on the basis of the documents submitted during the oral proceedings (see section VI above).

The Registrar: The Chairman:

E. Görgmaier F. Antony