BESCHWERDEKAMMERN DES EUROPÄISCHEN PATENTAMTS

BOARDS OF APPEAL OF THE EUROPEAN PATENT OFFICE

CHAMBRES DE RECOURS DE L'OFFICE EUROPEEN DES BREVETS

A		B		C	X

File Number: T 432/91 - 3.3.3

Application No.: 84 114 478.5

Publication No.: 0 144 081

Title of invention: Thermoplastic resin composition having excellent impact resistance and heat resistance

Classification: C08L 25/16

D E C I S I O N
of 15 July 1992

Proprietor of the patent: Mitsubishi Rayon Co., Ltd.

Opponent: Bayer AG

Headword:

EPC Article 56

Keyword: "Inventive step (no) - promising prior art pointing to the solution claimed - advantages corresponding to the expectations"

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Beschwerdekammern | Boards of Appeal | Chambres de recours

Case Number : T 432/91 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 15 July 1992

Appellant : (Opponent)
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Decision under appeal :
Interlocutory decision of the Opposition Division of the European Patent Office dated 20 March 1991, issued on 15 May 1991 concerning maintenance of European patent No. 0 144 081 in amended form.

Composition of the Board :

Chairman : F. Antony
Members : C. Gérardin
M. Aúz Castro

Summary of Facts and Submissions

I. The mention of the grant of the patent No. 0 144 081 in respect of European patent application No. 84 114 478.5 filed on 29 November 1984 and claiming priorities of 1 December 1983 and 2 December 1983 of two earlier applications in Japan, was published on 22 February 1989 on the basis of 11 claims.

Claim 1 read as follows:

"A thermoplastic resin composition having excellent impact resistance and heat resistance which comprises 5 to 95% by weight of a graft copolymer (I) obtained by polymerizing 93 to 30 parts by weight of a monomer or a monomer mixture for grafting comprising 30 to 100% by weight of at least one monomer selected from the group consisting of styrene, α-methylstyrene, acrylonitrile and methyl methacrylate and 0 to 70% by weight of at least one copolymerizable monomer having a $CH_2 = C<$ group in the presence of 7 to 70 parts by weight (in terms of solid) of a rubber latex of agglomerated particles having a particle diameter of at least 0.2 μm obtained by adding 0.1 to 5 parts by weight (in terms of solid) of an acid-group containing copolymer (B) latex obtained from 3 to 30% by weight of an acid-group containing monomer selected from the group consisting of acrylic acid, methacrylic acid, itaconic acid and crotonic acid, 97 to 35% by weight of at least one alkyl acrylate having 1 to 12 carbon atoms in the alkyl group, and 0 to 48% by weight of at least one monovinyl monomer copolymerizable therewith (100% by weight in total) to 100 parts by weight (in terms of solid) of a rubber (A) latex having an average particle diameter of 0.04 to 0.2 μm and having a pH of 7 or above obtained from 100 to 50% by weight of 1,3-butadiene and 0 to 50% by weight of other copolymerizable monomer having a

CH_2 = C< group (100% by weight in total); 95 to 5% by weight of a thermoplastic resin (II) obtained by polymerizing α-methylstyrene and at least one of other copolymerizable monomer having CH_2 = C< group; and 0 to 50% by weight of another thermoplastic resin (III) containing no α-methylstyrene unit, said components (I), (II) and (III) being compounded so as to give a content of said butadiene rubber (A) in the whole composition of 3 to 40% by weight and a content of α-methylstyrene unit in the whole composition of 10 to 75% by weight."

Claims 2 to 11 were dependent claims directed to preferred embodiments of the thermoplastic resin composition according to the main claim.

II. On 11 July 1989 the Opponent filed a Notice of Opposition against the grant of the patent and requested revocation thereof for non-compliance with the requirements of Article 100(a) EPC. More specifically, it was stated that the subject-matter as defined in the main claim was not novel and, in any case, would not involve any inventive step with regard to the teaching of DE-B-2 427 960 (document (1)). Following an amendment of the main claim in the course of the opposition procedure GB-A-2 002 398 (document (2)) was cited additionally in support of the latter objection.

III. By decision delivered orally on 20 March 1991, with written reasons posted on 15 May 1991, the Opposition Division held that there were no grounds of opposition to the maintenance of the patent in amended form on the basis of a new Claim 1, the amendments consisting in specifying that (a) the monomer mixture for grafting comprised "a mixture of styrene, α-methylstyrene and acrylonitrile or a mixture of α-methylstyrene and acrylonitrile", and (b) the copolymer (B) latex was a n-butylacrylate-methacrylic acid

"obtained from 3 to 30% by weight of methacrylic acid and 97 to 35% by weight of n-butylacrylate".

It was stated in this decision that the documents relied upon by the Opponent described two different methods to improve the impact strength of ABS graft polymers which were actually incompatible. Whereas document (1) taught agglomerating the rubber before grafting and without using α-methylstyrene in the graft polymer, document (2) disclosed that styrene should be replaced by α-methylstyrene and that a small particle size rubber spine should be used.

IV. The Appellant (Opponent) thereafter lodged a Notice of Appeal on 11 June 1991 and paid the prescribed fee at the same time. In the Statement of Grounds of Appeal filed simultaneously the Appellant underlined that the teaching of both documents was actually directed to polymer compositions having improved impact resistance properties and that it would self-evidently occur to the skilled man to combine the features of the two disclosures. The argument of a prejudice as acknowledged by the Opposition Division could not therefore be accepted.

In its subsequent submissions as well as during oral proceedings on 15 July 1992, which at both parties' request were conducted in the German language, the Appellant further argued that, in contradistinction to the introductory statement in the patent in suit, the substitution of α-methylstyrene for styrene generally improved both the heat resistance and the impact strength of ABS resins, as illustrated in US-A-3 010 936 (document (4)) and in US-A-3 111 501 (document (5)). Moreover, in the absence of appropriate comparative examples, the selection of n-butylacrylate-methacrylic acid copolymers as latex (B) could not be regarded as inventive; in

particular, the comparison with the prior art agglomerating agents should not be based on the same ponderal amounts of latex, but on equivalent amounts of the active groups. In fact, nothing distinguished the agglomerated rubber latex particles mentioned in document (1) and in the patent in suit, as evident from DE-A-2 101 650 (document (6)), to which there was a reference in document (1).

V. During oral proceedings the Respondent filed a new set of Claims 1 to 7 wherein the main claim had been slightly amended so as to read that the latex was "obtained from 3 to 30% by weight of methacrylic acid and n-butylacrylate (100% by weight in total) ...".

In support of the inventiveness of the claimed subject-matter the Respondent argued that a prejudice against the use of α-methylstyrene as a graft monomer to improve the impact strength resistance while retaining the heat resistance had to be overcome, and that there was an inventive selection invention as far as the composition of copolymer (B) was concerned. On the one hand, the simultaneous achievement of improved impact strength resistance and improved heat resistance must be regarded as surprising, since hitherto an improved heat resistance could only be obtained at the expense of a lower impact strength resistance; on the other hand, the only agglomerating agent actually used in document (1) was a copolymer of ethylacrylate and methacrylamide and nothing in that disclosure pointed towards the specific copolymer required in the patent in suit. The specificity of n-butylacrylate/methacrylic acid copolymer as an agglomerating agent was further supported by a technical report submitted on 16 June 1992, wherein the number average particle diameter distribution of agglomerated rubber latex was illustrated.

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

The Respondent requested that the appeal be dismissed with the proviso that the patent be maintained on the basis of Claims 1 to 7 filed during oral proceedings.

Reasons for the Decision

1. The appeal complies with Articles 106 to 108 and Rule 64 EPC and is admissible.

2. From points IV and V above it appears that both parties relied on late-filed technical evidence and documents in their written submissions as well as during oral proceedings.

At the beginning of oral proceedings the Chairman informed the parties that the experimental report announced by letter of 3 April 1992, but only submitted by the Respondent on 16 June 1992, thus barely one month before the oral proceedings, concerned a feature - the rubber latex particle size distribution - which was not essential for the issue of inventive step; in particular the Board took the view that these tests did not overcome the shortcoming of the information made hitherto available regarding the selection of the agglomerating agent (cf. point 6.1 hereinbelow). Both parties eventually agreed not to consider that feature in their subsequent argumentations.

Regarding the documents introduced by the Appellant, the Board decided to admit into the proceedings document (4) first cited in the submission dated 14 January 1992, for

it shows the influence of a thermoplastic resin (II) containing α-methylstyrene on the properties of the polymer blend, which feature had not been considered during the opposition procedure (cf. point 6.3 hereinbelow). By contrast, document (6) first mentioned in the submission dated 6 July 1992, which relates to the agglomeration of rubber latex particles and the resulting particle size distribution, and document (5) referred to for the first time during oral proceedings, which basically confirms the teaching of document (4), have been disregarded (Article 114(2) EPC).

3. The current wording of the claims does not give rise to any objections under Article 123 EPC.

Claim 1 differs in substance from the main claim as granted by (i) the limitation to two particular combinations of grafting monomer mixtures for the preparation of the graft copolymer (I), (ii) the selection of a specific copolymer (B) as agglomerating agent, and (iii) the deletion of the range of 97 to 35% by weight of n-butylacrylate in the compositional definition of copolymer (B).

More specifically, feature (i), i.e. the two grafting monomer combinations styrene/α-methylstyrene/acrylonitrile and α-methylstyrene/acrylonitrile, is supported by Claims 6 and 7 as granted, which themselves are identical with Claims 9 and 10 as originally filed. Further, feature (ii), i.e. the selection of n-butylacrylate and methacrylic acid as monomers for the preparation of copolymer (B), is the subject-matter of Claim 3 as granted, which itself is identical with Claim 5 as originally filed. Last, feature (iii), i.e. the deletion of a quantitative requirement which was obviously incompatible with the primary condition that the

constituting proportion of methacrylic acid in the copolymer is 3 to 30% by weight, overcomes an inconsistency without modifying the actual definition of copolymer (B) in view of the term "100% by weight in total" following "n-butylacrylate".

Claim 1 as granted differed itself from Claim 1 as originally filed by (iv) the indication of a specific range for the average particle diameter of the rubber (A) latex, which was originally disclosed in Claim 3, and (v) the correction of an obvious misspelling of the word "α-methylstyrene" at the end of the claim, which had self-evidently no impact on the scope of the claim.

As to the dependent Claims 2 to 7, they correspond respectively to Claims 2, 4, 5, 9, 10 and 11 as granted, which are identical with Claims 2, 6, 7, 11, 12 and 13 as originally filed, with their numbers and, where appropriate, appendancies adjusted.

4. The patent in suit concerns a thermoplastic resin composition having excellent impact resistance and heat resistance. Such a composition is disclosed in document (1), which the Board, like the Opposition Division, regards as the closest state of the art. More specifically, this citation describes the preparation of graft polymers by a three-step process (Claim 1) and the use thereof in combination with a so-called hard polymer component (Claim 3). This process comprises (a) the preparation by conventional emulsion polymerisation of a butadiene rubber latex having a particle diameter of 0.06 to 0.10 μm (column 3, line 21 to column 4, line 8), (b) the partial agglomeration of the first step rubber latex with an acrylate polymer, i.e. a copolymer of a monomer giving rise to water-soluble homopolymers and an acrylic ester derived from an alcohol having 1 to 4 carbon atoms,

whereby the particle diameters are increased to a range from 0.16 to 0.45 μm (column 4, lines 9 to 47), and (c) graft polymerisation of 20 to 90 parts by weight of styrene and/or acrylonitrile and/or methylmethacrylate onto 80 to 10 parts by weight of the second step agglomerated rubber latex (column 4, line 48 to column 5, line 12). As to the hard polymer components, they are defined as being polymers of styrene, α-methylstyrene, methylmethacrylate, acrylonitrile, methacrylonitrile, vinyl chloride as well as copolymers of two or more of these monomers (column 5, lines 13 to 17). The mixtures of graft polymer and hard polymer, which contain preferably 5 to 25% by weight of rubber, are said to exhibit an optimal combination of properties (column 3, lines 8 to 11 and column 5, lines 22 to 29). However, in spite of this promising teaching, the level actually achieved in terms of heat resistance and impact strength resistance is still regarded as unsatisfactory.

In the light of this shortcoming, the technical problem underlying the patent in suit can thus be seen to be the provision of a thermoplastic resin composition having improved heat resistance and impact strength resistance, without impairing its balance of properties.

According to the main claim of the patent in suit this problem is solved by (1) selecting as agglomerating agent a copolymer of n-butylacrylate and methacrylic acid, the latter being present in an amount of 3 to 30 % by weight, (2) using as graft monomers α-methylstyrene with acrylonitrile or with acrylonitrile and styrene, (3) choosing as a hard polymer component a copolymer of α-methylstyrene, and (4) compounding the polymers so as to give a content of α-methylstyrene units in the whole composition of 10 to 75% by weight.

In view of the experimental results in the patent in suit as well as the additional data in the comparative test reports submitted during the opposition and appeal procedures, the Board is satisfied that the combination of features (1) to (4) provides an effective solution to the above-defined technical problem.

5. After examination of the documents relied upon by the Appellant, the Board has come to the conclusion that this technical teaching is not disclosed in any of them and that the subject-matter of the patent in suit as defined in Claim 1 is, therefore, novel. Since the issue of novelty is no longer raised by the Appellant, it is not necessary to consider this matter in detail.

6. It still remains to be decided whether that subject-matter involves an inventive step having regard to the teaching of the documents relied upon by the Appellant.

6.1 In support of the inventiveness of feature (1) the Respondent has relied on the comparative data of the test report submitted on 11 March 1991 (Table 3).

The first experiment referred to as "Additional Test-3" is based on Example 1 of the patent as granted, wherein however α-methylstyrene has been used instead of styrene as graft monomer, all the other features being identical, in particular the agglomerating agent which is a n-butylacrylate/methacrylic acid copolymer (85/15). The fourth experiment referred to as "Additional Test-2" differs from the first one only by the use of an ethylacrylate/methacrylic acid copolymer (85/15) as agglomerating agent. In both experiments, thus, the same amount by weight of agglomerating agent has been introduced. As rightly objected by the Appellant, however, such comparison is not conclusive, for the intrinsic

activity of an agglomerating agent, i.e. its ability to agglomerate rubber latex particles, is determined by the number of active groups. In the present case, wherein the groups having a compatibilizing action are the ester groups, i.e. respectively the n-butylacrylate and ethylacrylate groups, it is self-evident that the addition of the same amount by weight of the two agglomerating agents will not result in the same number of active groups being present, but in a higher number of ethylacrylate groups. For this reason, the comparison on a weight basis is not appropriate to demonstrate the superiority of n-butylacrylate/methacrylic acid copolymer as an agglomerating agent according to feature (1).

The argument that the selection of n-butylacrylate/methacrylic acid copolymer as an agglomerating agent overcomes the prejudice resulting from the teaching of document (1) cannot be accepted either. It may be true that the acrylate esters envisaged in that citation are described as deriving preferably from alcohols having 1 to 4 carbon atoms and that ethylacrylate is said to be particularly preferred (column 4, lines 11 to 20), and actually used in the examples. However, as pointed out by the Appellant, this preference should be interpreted in connection with the technological context of the date of publication of that document (1975), when butanol, unlike ethanol, was not available on a large industrial scale; the choice of ethylacrylate as main monomer to prepare agglomerating agents was thus merely guided by practical considerations, not by technical superiority. This argument has not been disputed by the Respondent.

For these reasons, the choice of n-butylacrylate/methacrylic acid copolymer must be regarded as an arbitrary selection within the broader teaching of document (1), for which no technical advantage or

beneficial effect has been demonstrated. It follows that feature (1) cannot contribute to the inventiveness of the solution to the above-defined technical problem.

6.2 Document (2) describes the preparation of graft polymers by polymerisation of 95 to 40 parts by weight of α-methylstyrene, optionally together with styrene, and a copolymerisable nitrile monomer, preferably acrylonitrile, onto 5 to 60 parts by weight of a spine of a diene rubber latex, preferably polybutadiene and copolymers of butadiene with up to 40% of one or more copolymerisable monomers. The spines employed contain 30 to 100% of so-called small particles having a number average particle size of from 0.04 to 0.06 μm, and optionally up to 70% of so-called large particles having a number average particle size of from 0.3 to 0.4 μm. The graft polymerisation reaction is carried out in an aqueous medium and in the presence of a water-soluble, free-radical polymerisation initiator and an anionic emulsifier (Claim 1; page 1, lines 4 to 36). The resultant graft polymers exhibit good heat distortion temperature and a good balance between tensile strength, impact strength and flow properties (Abstract).

In Example III five graft polymers having different α-methylstyrene:styrene ratios, the other compositional features being identical, have been tested. The experimental results in Table III clearly show that impact strength markedly increases with the amount of α-methylstyrene and that the highest impact strength is obtained for graft monomer compositions containing no styrene. In that case, which corresponds thus to a graft monomer composition containing 56.6 parts by weight of α-methylstyrene and 25.4 parts by weight of acrylonitrile, the impact strength resistance is 14 times as high as for the corresponding graft polymer obtained with styrene.

Further, the comparison of heat distortion temperature for the nine graft polymers tested in Example I, Table I reveals, first, that this parameter is significantly lower for rubbers grafted with styrene and acrylonitrile (196°F in Runs 7 and 8) than for rubbers grafted with α-methylstyrene and acrylonitrile (210 to 218°F in Runs 1 to 6 and 9), and, secondly, that this parameter is hardly affected by variations of the other compositional features.

In the Board's view, this dual teaching represents a strong incentive for the skilled man faced with the problem of increasing the impact strength resistance and the heat resistance to operate along the same line, i.e. to substitute α-methylstyrene for part or all of styrene in the graft monomer mixture. It follows that feature (2) of the solution claimed by the Respondent must be regarded as obvious.

6.3 Document (4) relates to polymer blends exhibiting a desirable combination of physical properties, in particular of high impact value and high heat distortion resistance (column 1, lines 11 to 18; column 2, lines 22 to 24). These blends consist of 50 to 70% by weight of (A) a graft copolymer of 20 to 60% by weight of a synthetic aliphatic conjugated diolefine hydrocarbon polymer, 30 to 70% by weight of a compound selected from the group consisting of styrene, α-methylstyrene, vinyltoluenes, α-methylvinyltoluenes and mixtures thereof, and 10 to 30% by weight of a copolymerisable nitrile monomer, and, correspondingly, from 50 to 30% by weight of (B) a copolymer of 20 to 30% by weight of acrylonitrile, 80 to 60% by weight of α-methylstyrene and 0 to 10% by weight of styrene (Claim 1; column 9, lines 10 to 15).

Examples 3 to 6 illustrate the influence on heat distortion of the amount of resin B in the blends, resin B being a terpolymer obtained from 62.5 parts by weight of α-methylstyrene, 7.5 parts by weight of styrene and 30 parts by weight of acrylonitrile (see preparation thereof in Example 2). The other component in the blends is either graft copolymer Y, which is obtained by the graft copolymerisation of 45 parts by weight of styrene and 25 parts by weight of acrylonitrile onto 30 parts by weight of a polybutadiene latex, or graft copolymer X, which is obtained by the graft copolymerisation of 51 parts by weight of styrene and 29 parts by weight of acrylonitrile onto 20 parts by weight of a polybutadiene latex (see preparations thereof in Example 1). The experimental data in Examples 3 to 5 show that the heat distortion point of resin Y-resin B blends is quite constant and comparatively low (103-104°C) when the polymer blends contain from 10 to 20% by weight of resin B (Example 3), then increases to 108°C when the content of resin B in the blends is 30% by weight (Example 3), and eventually remains constant and high (109-110°C) when the resin B content is in the range 40 to 50% by weight (Examples 4 and 5). Similarly, according to Example 6 the heat distortion point of blends containing 80 parts by weight of graft copolymer X and 20 parts by weight of resin B is relatively low as well (106°C). All these results demonstrate the beneficial influence on the heat distortion point of the amount of resin B in the blend, i.e. the beneficial influence of α-methylstyrene itself on heat resistance, since that monomer is used as the major component in the preparation of resin B.

It is essential to appreciate that any increase of the amount of resin B in the blend in order to improve the heat distortion resistance will correspondingly lower the amount of resin Y and, thereby, the amount of rubber in

that blend. Since the amount of rubber is responsible to a large extent for the impact strength properties of the blend, a corresponding decrease of these properties is to be expected. Although this actually occurs, the Izod notch impact values reported in the examples demonstrate that the general level remains satisfactory (column 8, lines 48 to 52); this is the case for the resin Y-resin B blends containing 40 and 50% by weight of resin B (Examples 4 and 5), which exhibit on an average Izod notch impact values of 4.7 and 4.1 ft. lbs/inch respectively, as well as for the resin X-resin B blend in the proportion 80:20 (Example 6), which still exhibits a Izod notch impact value of 3.8 ft. lbs/inch in spite of a lower amount of rubber in the graft copolymer. This relatively slow decrease of impact strength as the polybutadiene content of the system decreases is a major advantage regarding the balance of properties of the blends; more specifically, the linear relationship between, on the one hand, the impact strength of these blends and, on the other hand, the hardness (Rockwell Hardness) and tensile strength thereof, makes it possible to obtain relatively high values for the latter parameters, while still preserving high impact strength (see experimental data in Examples 3 to 6).

Document (4) teaches thus that decisive advantages in terms of heat resistance can be obtained, without substantially impairing the impact strength properties, by blending copolymers of α-methylstyrene with rubber graft copolymers, which renders obvious feature (3) of the solution claimed by the Respondent.

6.4 The teaching of documents (2) and (4) being directed to polymer compositions exhibiting both high heat resistance and impact strength properties, the skilled man would self-evidently combine the technical features of these disclosures in order to optimise the properties of these

blends. In doing that, he would introduce α-methylstyrene both as the main graft monomer in the preparation of the graft copolymer (I) and as the main monomer in the preparation of the blend copolymer or thermoplastic resin (II). The determination of the most appropriate amount of α-methylstyrene in the blends would not involve more than routine manipulations of, first, the amounts of α-methylstyrene incorporated in each of the polymer components and, secondly, of the weight ratio of these two components carried out on the basis of trial and error in order to meet the specific criteria depending upon the proposed end use of the shaped objects fabricated therefrom. It follows that feature (4), i.e. the content of 10 to 75% by weight of α-methylstyrene units in the thermoplastic resin compositions according to Claim 1 of the patent in suit, cannot be regarded as an inventive feature.

6.5 Besides, the Board notes that the combination of features (1) to (4) does not result in any improvement of properties going beyond what the skilled man would normally expect; in particular, no unforeseen effect has been claimed, let alone demonstrated.

6.6 In conclusion, for the reasons given above, features (1) to (4), either in isolation or in combination, of the solution according to Claim 1 do not involve an inventive step, so that Claim 1 is not allowable.

7. In the absence of a separate request directed to the specific features mentioned in the dependent Claims 2 to 7, the latter must fall with the main claim, since a request can only be considered as a whole. Besides, no argument in favour of the inventiveness of any of these features has been provided by the Respondent.

Order

For these reasons, it is decided that:

1. The decision under appeal is set aside.

2. The patent is revoked.

The Registrar: The Chairman:

E. Görgmaier F. Antony

CG 22.10.92
IC 22/10/92
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