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D E C I S I O N
of 7 December 1994

Case Number: T 0642/91 - 3.3.3

Application Number: 83106955.4

Publication Number: 0100912

IPC: C09J 3/14

Language of the proceedings: EN

Title of invention:
Hot-melt adhesive composition

Patentee:
MITSUI PETROCHEMICAL INDUSTRIES, LTD.

Opponent:
Stamicarbon B.V.

Headword:
-

Relevant legal provisions:
EPC Art. 56

Keyword:
"Inventive step (affirmed)"
"Teaching of relevant state of the art - silence as to one essential parameter and absolute contradiction in respect of another"

Decisions cited:
-

Catchword:
-

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Case Number: T 0642/91 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 7 December 1994

Appellant: MITSUI PETROCHEMICAL INDUSTRIES, LTD.
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Respondent: Stamicarbon B.V.
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Representative: -

Decision under appeal: Decision of the Opposition Division of the
European Patent Office dated 9 July 1991 revoking
European patent No. 0 100 912 pursuant to
Article 102(1) EPC.

Composition of the Board:

Chairman: C. Gérardin
Members: R. Young
W. Schar

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Summary of Facts and Submissions

I. The mention of the grant of European patent No. 0 100 912, with 23 Claims, in respect of European patent application No. 83 106 955.4, filed on 15 July 1983 and claiming Japanese priorities of 16 July 1982 and 5 August 1982 (JP 123018/82 and JP 135718/82 respectively) was announced on 3 June 1987 (cf. Bulletin 87/23). Independent Claims 1, 16 and 17 read, respectively, as follows:

- "1. A hot-melt adhesive composition comprising a styrene-type hydrocarbon-modified polyolefin (c) composed of 100 parts by weight of a crystalline polyolefin (a) having a degree of crystallinity of at least 10% and 0.15 to 50 parts by weight of a styrene-type hydrocarbon (b) bound thereto by graft-copolymerization, said modified polyolefin (c) having an intrinsic viscosity $[\eta]$ of 0.5 to 5 dl/g, a degree of crystallinity of at least 10% and a weight average molecular weight \bar{M}_w /number average molecular weight \bar{M}_n ratio of from 1 to 15.
16. Use of the composition of claim 1 as an intermediate adhesive layer in laminating two polymers.
17. A laminated article composed of at least
 - (A) a first polymer layer,
 - (B) an intermediate adhesive layer composed of a hot-melt adhesive composition according to claim 1,
and
 - (C) a second polymer layer."

Dependent Claims 2 to 15 and 18 to 23 were directed to further elaborations of the composition of Claim 1 and the article of Claim 17 respectively.

II. Notice of Opposition was filed on 12 February 1988 on the grounds of lack of novelty and inventive step. The Opposition was supported inter alia by the documents:

D1: EP-A-0 090 418, and

D5: BE-A-742 272.

III. By a decision dated 9 July 1991, which was based on an amended set of Claims 1 to 8 filed on 2 November 1990 and corresponding in substance to Claims 16 to 23 as granted (cf. section I., above), the Opposition Division revoked the patent because the subject-matter of Claim 1 did not involve an inventive step.

According to the decision, neither of the closest state of the art documents D1 (which was relevant only under Article 54(3) EPC for the Contracting States DE, FR and GB), and D5 was detrimental to novelty. With regard to inventive step, it was concluded that the claimed parameters were not critical in practice, did not therefore represent a special selection, and had in any case not been asserted to present a surprising effect compared with D5.

IV. On 21 August 1991, a Notice of Appeal against the above decision was filed by the Patentee, together with payment of the prescribed fee.

With the Statement of Grounds of Appeal filed on 8 November 1991, the Appellant (Patentee) submitted a new, restricted Claim 1 and argued that the data in the

patent in suit showed that the relevant parameters were critical to obtaining an unexpected improvement in delamination strength vis-à-vis the state of the art.

V. The Respondent (Opponent) argued, in essence, as follows:

- (i) the new claim contravened Article 123 EPC, and
- (ii) the new parameters introduced no real limitation, since the ranges, in particular of the intrinsic viscosity and melt viscosity, were extremely broad, the latter reflecting a commercial polyethylene (cf. submission dated 24 March 1992).

VI. Following a communication by the Board, issued on 5 October 1994 together with a summons to oral proceedings, the Appellant filed, with a submission dated 10 November 1994, a further amended set of Claims 1 to 8, of which independent Claims 1 and 2 read, respectively, as follows:

- "1. Use of a hot-melt adhesive composition comprising a styrene-type hydrocarbon-modified polyolefin (c) composed of 100 parts by weight of a crystalline ethylene-type polymer (a) having a degree of crystallinity of 15 to 70%, an intrinsic viscosity $[\eta]$ of 0.75 to 3 dl/g, a weight average molecular weight \bar{M}_w /number average molecular weight \bar{M}_n ratio of 1 to 10 and a density of 0.84 to 0.96 and 0.2 to 5 parts by weight of a styrene-type hydrocarbon (b) bound thereto by graft-copolymerization, said styrene-type hydrocarbon-modified ethylene-type polymer (c) having a degree of crystallinity of 10 to 70%, an intrinsic viscosity $[\eta]$ of 0.7 to 3 dl/g, a weight average molecular weight \bar{M}_w /number average molecular weight

\bar{M}_n ratio of 1 to 10, a density of 0.84 to 0.96 and a melt viscosity at 230°C of from 10 to 0.5×10^5 Pa.s, as an intermediate adhesive layer in laminating two polymers.

2. A laminated article composed of at least
 - (A) a first layer of a polymer selected from the group consisting of polyolefins, aromatic polymers and chlorine-containing polymers,
 - (B) an intermediate adhesive layer composed of the hot-melt adhesive composition as defined in Claim 1,
and
 - (C) a second layer of a polymer selected from the group consisting of aromatic polymers and chlorine-containing polymers."

Dependent Claims 3 to 8 are directed to further elaborations of the article of Claim 2.

The claims were accompanied by an adapted description. Further amendments to the description were filed with submissions dated 30 November 1994 and 1 December 1994.

- VII. The Appellant requests that the decision under appeal be set aside and the patent be maintained on the basis of Claims 1 to 8 filed on 10 November 1994, and by implication the revised description filed therewith and incorporating the amendments filed on 30 November 1994 and 1 December 1994. In the alternative, oral proceedings are requested.

The Respondent, which had not commented in substance on the above communication and had informed the Board that it would not attend the oral proceedings, requests by implication that the appeal be dismissed.

VIII. The Board informed the parties by a communication issued on 1 December 1994 that the oral proceedings, scheduled for 6 December 1994, had been cancelled.

Reasons for the Decision

1. The appeal is admissible.
2. This decision is based on the following text of the patent in suit:

Claims:

Claims 1 to 8 filed on 10 November 1994, with the letter of the same date;

Description:

amended pages 2, 7, 10 and 12 of the printed specification as well as the three typed pages for insertion at page 2, lines 3 to 38 and the typed page for insertion at page 5, lines 14 to 23; all filed on 10 November 1994, with the letter of the same date;

amended pages 3, 4, 5, 8, 9 and 11 of the printed specification, filed on 30 November 1994, with the letter of the same date, and

amended page 6 of the printed specification, filed on 1 December 1994, with the letter of the same date.

3. *Amendments*

- 3.1 Claim 1 is based on Claims 1, 2, 7 and 16 of the application as filed, as well as its description, specifically as follows:

(i) for the polymer (a):

page 2, line 30 (degree of crystallinity);
page 3, line 2 (intrinsic viscosity);
page 3, line 10 (\bar{M}_w/\bar{M}_n ratio);
page 3, line 4 (density);

(ii) for the polymer (c):

page 9, lines 17, 18 (degree of crystallinity);
page 9, line 15 (intrinsic viscosity);
page 9, line 23 (\bar{M}_w/\bar{M}_n ratio);
page 9, line 26 (density);
page 9, line 31 (melt viscosity).

The specific combination of the parameters in Claim 1 is also adequately supported by the Examples, especially Examples 1, 2, 4, 9, 10, 12, 17, 18, 20, 25, 32, 39 and 47 as originally filed.

3.2 Claim 2 is based on Claim 17 together with page 11, lines 19 to 29 of the application as filed. Claims 3 to 8 are supported by Claims 18 to 23 respectively of the application as filed.

3.3 The amendments to Claims 1 and 2 are furthermore such as to render them considerably narrower in scope than the corresponding claims as granted.

3.4 The description has been adapted to the claims.

Thus, no objection arises under Articles 123(2) or 123(3) EPC in respect of the amendments.

4. *Closest state of the art*

The patent in suit is concerned with the use of a hot-melt adhesive composition as an intermediate layer in laminating polymers, the composition comprising a styrene-type hydrocarbon-modified-polyolefin (c) composed of an ethylene-type polymer (a) and a styrene-type hydrocarbon (b) bound thereto by graft-polymerization.

Compositions of this general type for use as adhesives are, however, known from D5, which is considered to represent the closest state of the art.

According to D5, graft polymers of poly α -olefines are prepared by the reaction of a low viscosity, unsaturated α -olefin polymer with at least one vinylic or acrylic comonomer, the starting polymer having a degree of non-saturation of at least 0.05%, and a viscosity not higher than 20 Poise (Claim 1).

The α -olefin polymer may be prepared by thermally degrading a high molecular weight poly α -olefin, especially polyethylene, to obtain a crystalline polymer of low molecular weight, having a melt viscosity of 0.1 to 15 Poise at 150°C, an inherent viscosity between 0.05 and 0.5 and a degree of non-saturation of at least 0.1% (page 3, lines 7 to 13 and page 3, line 33 to page 4, line 1). The comonomer may be e.g. acrylic acid or styrene (Claim 6 and page 5, line 29). The resulting graft polymers, which may have an inherent viscosity of between about 0.1 and about 0.5, can be used as thermosensitive adhesives (page 3, lines 2 to 5 and page 5, lines 23 to 34).

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According to Example 25, a mixture of acrylic acid and styrene is grafted on to a low viscosity polyethylene powder. The graft polymer obtained has a melt viscosity of 2.2 Poise at 190°C, and an index of saponification of 61. It can be emulsified and can serve for adhering paper to paper or to an aluminium foil (page 11, lines 13 to 18).

5. *The technical problem and its solution*

Compared with this state of the art, the objective technical problem is seen by the Board in the search for a further hot-melt adhesive composition suitable for use as an intermediate layer in laminating polymers.

The solution proposed according to Claim 1 of the patent in suit is to use, as the graft polymer for the intermediate adhesive layer, a composition comprising a styrene-modified polyolefin (c) composed of 100 parts by weight of a crystalline ethylene-type polymer (a) having inter alia an intrinsic viscosity $[\eta]$ of 0.75 to 3 dl/g and a weight average molecular weight \bar{M}_w /number average molecular weight \bar{M}_n ratio of 1 to 10, and 0.2 to 5 parts by weight of a styrene-type hydrocarbon (b) bound thereto by graft-copolymerization, the styrene-type hydrocarbon-modified ethylene-type polymer (c) having inter alia an intrinsic viscosity $[\eta]$ of 0.7 to 3 dl/g, a weight average molecular weight \bar{M}_w /number average molecular weight \bar{M}_n ratio of 1 to 10 and a melt viscosity at 230°C of from 10 to 0.5×10^5 Pa.s.

5.1 It can be seen from the illustrative Examples in the patent in suit that a laminate having a graft polymer adhesive layer with parameters falling within the specified ranges has a delamination strength (of the more easily peelable layer) in the region of at 500 to

600 g/cm (cf. Table 2, Examples 1, 2, 4, 9, 10, 12 and 17, 18 and 20) with a lowest value of 380 g/cm (cf. Table 4, Example 39) whereas the laminates of the Comparative Examples, to the extent they can be prepared at all (cf. Table 2, Comparative Examples 2, 8 and 14), have a delamination strength of not more than 200 g/cm and generally much lower (cf. Table 2, Comparative Examples 10, 11).

5.2 The fact that advantageously high delamination strengths are also recorded in relation to the "Reference Examples", in which the polymers used have one or more parameters falling outside the required ranges, is of no consequence for the effectiveness of the solution of the technical problem, since the "Reference Examples" belong neither to the solution as defined nor to the state of the art.

5.3 The closest state of the art document D5 on the other hand contains no data on adhesive strength, and indeed fails to disclose the use of adhesives in laminating polymers. No evidence as to the effectiveness of the latter has been filed, though the onus of proof lay with the Respondent.

Consequently, there is no ground for assuming a better adhesive strength for the graft polymers of D5 than that acknowledged to be obtained with the generally known modified polyolefins discussed in the patent in suit, which are stated to have "no excellent bonding property" (cf. patent as granted, page 2, lines 13 to 18).

5.4 Since, on the other hand, according to the uncontested evidence of the patent in suit, the adhesive compositions of the illustrative Examples have acceptable adhesive strengths which are higher than those of the Comparative Examples, the latter

representing variants lying, if anything, closer to the claimed subject-matter than the acknowledged prior art, it is reasonable to conclude that the adhesives illustratively exemplified are at least as effective as anything disclosed in D5.

Thus, it is credible to the Board that the claimed measures are effective to solve the stated problem.

6. *Novelty*

Novelty is given in relation to D1 and D5 for the reasons set out in the decision under appeal (cf. para. II.3). The remaining documents in the proceedings are less relevant.

Consequently, the claimed subject-matter is held to be novel.

7. *Inventive step*

It is necessary to answer the question of whether the skilled person would have had reason to expect an adhesive at least as effective as anything disclosed in D5, and especially suitable for the lamination of polymers to each other, to result from modifying a graft polymer as disclosed in D5 to have the particular parameters specified (cf. section 5, above), especially molecular weight ratio \bar{M}_w/\bar{M}_n , intrinsic viscosity and melt viscosity.

7.1 The disclosure of D5 is entirely silent as to the relevant parameter of molecular weight ratio (\bar{M}_w/\bar{M}_n).

7.1.1 In this connection, the method described in D5 of obtaining the starting polyolefins, which is by thermal degradation of a higher molecular weight polymer, is not

such as to result in a particularly uniform distribution of molecular weight.

- 7.1.2 In any case, it is clear from the information given in the patent in suit - itself in line with common general knowledge - that conventional ethylene-type polymers can indeed have molecular weight ratios falling well outside the restricted range concerned (cf., for instance, Comparative Examples 19 to 24).

Consequently, there is no reason for supposing the molecular weight ratios of the polymers of D5 would inevitably fall within the relevant restricted range.

- 7.1.3 Nor, in view of the silence in D5 regarding this parameter, and the absence of other relevant disclosure, was there any incentive for the skilled person to pay particular attention to this aspect, let alone to work within the relevant range.

- 7.2 Quite apart from this, the essential teaching of D5 is that the base polyolefin from which the graft polymer is prepared must be of "low viscosity", i.e., not greater than 20 Poise (2 Pa.s). In this connection, according to the only Example in which styrene is used as a comonomer (Example 25) the final graft polymer had a melt viscosity of only 2.2 Poise (0.22 Pa.s) at 190°C.

The melt viscosity of the graft polymer used according to the patent in suit, on the other hand, is required to be at least 10 Pa.s. Although it is true that temperature of the relevant viscosity measurement, at 230°C, is somewhat higher than that in D5, this is not sufficient to invalidate the general conclusion that the melt viscosity values of the polymers used for the solution of the technical problem are substantially higher than the maximum allowed according to D5.

7.3 According to the description of D5, moreover, the inherent viscosity of the starting polyolefin was in practice between about 0.05 and 0.5. None of the Examples discloses an inherent viscosity of the relevant species higher than 0.42 (cf. Example 2).

It is, on the other hand, a requirement of the starting polyolefin (a) according to the patent in suit that its intrinsic viscosity (which will in any case be lower than a corresponding inherent viscosity value because it is obtained by extrapolating the latter to zero concentration) is not less than 0.75 dl/g, that of the final graft polymer (c) being not less than 0.7 dl/g (cf. patent in suit, Claim 1).

Thus the minimum intrinsic viscosity permitted by the solution of the technical problem is greater than the maximum corresponding viscosity value disclosed in D5.

- 7.3.1 The argument of the Respondent concerning the breadth of the ranges of this parameter, even if it applies to commercial polyethylenes in general, is thus evidently not applicable to the graft polymers used in D5.
- 7.3.2 Nor is there anything in the other documents in the proceedings to suggest that the low viscosities of the polymers according to D5 could be significantly altered whilst leaving the associated properties, such as adhesiveness, unchanged.
- 7.3.3 In summary, D5 emphasizes a kind of polymer in which the \bar{M}_w/\bar{M}_n ratio is unimportant, and which necessarily has a markedly lower viscosity than the polymers used according to the patent in suit. The latter are not a selection from the former.

Consequently, the unexpected result or surprising effect critically associated with such a selection is not a necessary criterion (cf. section 5.2 above and decision under appeal, para. II. 4.3).

- 7.3.4 On the contrary, to arrive at a solution of the technical problem, it would have been necessary to fill the silence of the relevant state of the art as to one essential parameter and to overcome its absolute contradiction in respect of another.

8. Hence, the subject-matter of Claim 1 does not arise in an obvious way. It therefore involves an inventive step. The same considerations apply to the subject-matter of Claim 2, which requires the presence of the adhesive composition defined in Claim 1, and to that of Claims 3 to 8, which are dependent on Claim 2.

9. In view of the above, there is no barrier to maintenance of the patent in the form requested by the Appellant. Consequently, there was no need to hold oral proceedings.

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 specified in section 2 of the Reasons for the Decision, above.

The Registrar:


E. Gorgmaier

The Chairman:


C. Gérardin