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File Number: T 0139/90 - 3.3.3

Application No.: 83 104 455.7

Publication No.: 0 094 051

Title of invention: Ethylene-alpha-olefin-polyene random copolymer rubber

Classification: C08F 210/18

DECISION
of 25 March 1993

Applicant: Mitsui Petrochemical Industries, Ltd.

Opponent: Stamicarbon B.V.

Headword:

EPC Art. 54, 56, 114(2)

Keyword: "Novelty: yes (on balance of probability)"
"Inventive step: no"
"Late-filed evidence not admitted"

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Beschwerdekammern

European Patent Office
Boards of Appeal

Office européen des brevets
Chambres de recours

Case Number : T 0139/90 - 3.3.3

DECISION
of the Technical Board of Appeal 3.3.3
of 25 March 1993

Appellant : (Proprietor of the patent)	Mitsui Petrochemical Industries, Ltd. 2-5, Kasumigaseki 3-chome Chiyoda-ku Tokyo 100 (JP)
Representative :	Kador & Partner Corneliusstrasse 15 W-8000 München 5 (DE)
Respondent : (Opponent)	Stamicarbon B.V. P.O. Box 605 NL-6160 AP Geelen (NL)
Representative :	Hoogstraten, Willem Cornelis Roeland Octrooibureau DSM Postbus 9 NL-6160 MA Geleen (NL)
Decision under appeal :	**Decision of the Opposition Division of the European Patent Office of 28 November 1989, issued on 21 December 1989 revoking European patent No. 0 094 051 pursuant to Article 102(1) EPC.**

Composition of the Board :

Chairman : F. Antony
Members : P. Kitzmantel
J.A. Stephens-Ofner

Summary of Facts and Submissions

I. European patent application No. 83 104 455.7, which had been filed on 5 May 1983, claiming priority from a Japanese application filed 6 May 1982, was granted as European patent No. 94 051 on 7 January 1987, with six claims, independent Claims 1 and 4 reading as follows:

"1. Ethylene-alpha-olefin-polyene random copolymer rubber comprising ethylene, an alpha-olefin having 3 to 10 carbon atoms, and a polyene, characterized in that

(A) a molar ratio of ethylene/alpha-olefin is 55/45 to 95/5;

(B) the polyene is a mixture of 5-ethylidene-2-norbornene (ENB) and 5-vinyl-2-norbornene (VNB) in a molar ratio of ENB/VNB of 1/1 to 20/1;

(C) an iodine value of the copolymer rubber is 2 to 40;

(D) an intrinsic viscosity $[\eta]$ of the copolymer rubber is 1.0 to 6.0 dl/g measured in decalin at 135°C; and

(E) g_η^* defined by a ratio of the intrinsic viscosity $[\eta]$ to an intrinsic viscosity $[\eta]1$ of a linear ethylene-propylene random copolymer having an ethylene content of 70 mole% and the same weight-average molecular weight determined by a light-scattering method as that of the copolymer rubber is 0.2 to 0.8.

4. A process for preparing ethylene-alpha-olefin-polyene random copolymer rubber of Claim 1 comprising the step of copolymerizing ethylene, an alpha-olefin having 3 to 10 carbon atoms, 5-ethylidene-2-norbornene, and 5-vinyl-2-norbornene, wherein a molar ratio of 5-ethylidene-2-norbornene/5-vinyl-2-norbornene is 1/1 to

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20/1, in a hydrocarbon solvent in the presence of a catalyst formed by

(i) a soluble vanadium compound having the formula:

$$VO(OR)_nX_{3-n} \quad (I)$$

or

$$VX_4 \quad (II)$$

wherein R is a hydrocarbon radical having 1 to 10 carbon atoms; X is a halogen atom, and $0 \leq n \leq 3$; and

(ii) an organoaluminium compound

$$R'_mAlX'_{3-m} \quad (III)$$

wherein R' is a hydrocarbon radical having 1 to 10 carbon atoms, X' is a halogen atom, and $0<m<3$."

Claim 3 is directed to the vulcanisate of the copolymer rubber of Claim 1.

II. Notice of opposition was filed by Stamicarbon B.V. on 7 October 1987, requesting revocation of the patent in its entirety, on the grounds of lack of novelty and inventive step, in particular having regard to documents
(1) GB-A-1 311 946,
(2) GAK 4/1976, Jahrgang 29, pp. 184 to 189 and
(3) GB-A-2 047 254 (& corresponding NL-A-79.07300).

III. In a decision announced orally on 28 November 1989, and posted on 21 December 1989, the Opposition Division revoked the patent.

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That decision held that the subject-matter of Claims 1 to 3 was not novel in view of the disclosure in Example 1 of document (1). This Example explicitly described a copolymer having all the features of Claim 1, except for the g_η^* value. On the basis of certain extrapolations, it was concluded that it must be inferred from the comparative evidence submitted by the Patentee (especially Experiment I submitted during the grant procedure on 15 November 1985 and Experiment B as corrected on 26 October 1989), which evidence did not exactly repeat the conditions of said Example 1, that when properly adhering to said conditions, one would arrive at a copolymer having a g_η^* value falling within the claimed range. The subject-matter of the process Claims 4 to 6 was considered non-inventive.

IV. The Patentee (Appellant) filed an appeal on 20 February 1990 and paid the appeal fee on the same date. A statement of Grounds of Appeal was filed on 12 April 1990.

V. Oral proceedings were held on 25 March 1993.

The Appellant requested that the decision under appeal be set aside and that the patent be maintained as granted (main request), or on the basis of an amended Claim 1 (combining the features of granted Claims 1 and 4) submitted in the course of oral proceedings (auxiliary request).

The Respondent (Opponent) requested dismissal of the appeal.

VI. The written and oral arguments of the Appellant may be summarized as follows:

Novelty

As demonstrated by newly submitted Comparative Experiments 1 to 3, the conclusion in the appealed decision concerning the g_η* value of the products of Example 1 of (1) was wrong, since it did not take into account the presence of diethyl zinc as molecular weight modifier in Example 1 of (1). Moreover, since it was stated on page 2, lines 63 to 68 of (1) that the copolymers were linear and must therefore have a g_η* value close to 1, the burden was on the Opponent (Respondent) to prove the contrary.

While with telefax of 19 February 1993 the Respondent submitted the results of his reworking of said Example 1, this submission was too late considering the oral proceedings convened for 25 March 1993 and did not give the Appellant sufficient time to assess the validity of the Respondent's experiment. The latter should therefore not be admitted into the appeal proceedings. If it were admitted than the Appellant should be granted a time limit to carry out appropriate counter-experiments.

Inventive step

In spite of the statement in (3) page 4, lines 1 to 3 that preferably the polyene component was "at least one member" selected from dicyclopentadiene (hereinafter DCPD), 5-vinyl-2-norbornene (hereinafter VN) and 5-ethylidene-2-norbornene (hereinafter EN), the overall disclosure of (3) did not teach the conjoint use of more than one polyene. Nor could (1), in view of the variety of polyene combinations disclosed therein, suggest to the skilled man the benefits of the combined use of EN and VN. Concerning his auxiliary request the Appellant

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contended that (1) used catalysts different from the ones of the patent-in-suit and from those used in (3).

VII. The written and oral arguments of the Respondent may be summarized as follows:

Novelty

The relevance of Appellant's newly submitted Comparative Experiments was contested, since they still did not match all conditions of Example 1 of (1). The deviations were such as to necessarily reduce the molecular weight, thus enhancing the g_η^* value as demonstrated by Fig. 2 of (2) and by the synopsis of the results of the Respondent's experiments (Table 2 of the explanatory evidence filed in oral proceedings).

These conclusions would be in agreement with the Respondent's own repetition of Example 1 of (1) showing a g_η^* value of the copolymers obtained within the claimed range.

Besides, and quite irrespective of these results, the skilled person, taking into account his knowledge concerning the correlation between the g_η^* value and long chain branching or cross-linking, would conclude that the copolymers resulting from said Example 1 must inevitably have a g_η^* value within the claimed range of from 0.2 to 0.8.

Inventive step

Although (3) did not specifically disclose the combined use of EN and VN, it clearly pointed at the possibility of their combination (page 4, lines 1 to 3). Since the

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advantages of using each of the preferred polyenes DCPD, EN and VN were clearly set out in (3) page 3, lines 47 to 51, and since the influence on the g_η* value of using VN (or DCPD) was also indicated at page 2, lines 43 to 49, the skilled person would not have hesitated to combine EN and VN in a polyene mixture.

Concerning the auxiliary request, the sole important feature that it added was the use of certain catalysts in the preparation of the copolymers. These catalysts, however, were the same as those in (3). Furthermore, the catalyst system used in the patent-in-suit was a conventional Ziegler system which was also within the disclosure of (1) (page 1, line 69 to page 2, line 3).

VIII. At the end of the oral proceedings the Chairman announced the decision of the Board.

Reasons for the Decision

1. Admissibility

The appeal is admissible.

2. Procedural matters

The Board decides under Art. 114(2) EPC not to admit into the proceedings the evidence submitted by the Respondent on 19 February 1993 in support of the allegation that Example 1 of (1) leads to a product having a g_η* value of between 0.2 to 0.8, thereby destroying the novelty of the subject-matter of the patent-in-suit. In principle, the burden of proof resting on the Opponent (Respondent), such evidence should have been provided within the

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9-month opposition period. At the very latest it should have been provided in prompt response to the Appellant's data of 17 December 1991.

Main request

3. Compliance with Art. 123(2) and (3) EPC

The main request is based on the claims as granted. Their subject-matter finds support in Claims 1 to 7 and at page 5, line 17 of the originally filed application so that both Art. 123(2) and (3) EPC are complied with.

4. Novelty

4.1 The Board is satisfied that the copolymers prepared in Example 1 of (1) meet all the features defined in Claim 1 of the patent-in-suit except for the g_η^* value, which is not disclosed in that Example, as is indeed undisputed by the parties. The novelty of the subject-matter of the patent-in-suit thus hinges solely on whether the said Example inevitably yields a product having a g_η^* value lying within the claimed range of from 0.2 to 0.8.

4.2 Page 2, lines 63 to 68 of (1) sets out that "The copolymers of this invention generally have a linear structure, that is, they are free of long branchings, as is proved by their properties, in particular their viscous behaviour, which is practically the same as that of linear ethylene/propylene copolymers."

This statement implies that the g_η^* value of the copolymers of (1), including that of Example 1, should be close to the maximum of 1 (cf. definition of g_η^* at page 3, lines 44 to 52 in the patent-in-suit).

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4.3 During the grant, opposition and appeal proceedings, the Appellant produced several experiments purporting to repeat Example 1 of (1), all yielding copolymers having g_η* values of above 0.8. However, none of these experiments was fully in line with the operating conditions of this Example. The one that came closest was "Experiment 3" submitted with the Appellant's letter of 17 December 1991, and differing from Example 1 of (1) only in the use of four times the amount of diethyl zinc, which according to the Appellant was necessary in order to get a copolymer with a sufficiently low molecular weight to be soluble in decalin at 135°C so as to allow determination of the g_η* value.

While the Respondent was right in arguing that there was some correlation between the molecular weight and the g_η* value in the sense that lower molecular weights generally went along with higher g_η* values (see Fig. 2 of (2) and Table 2 of the synopsis presented by the Respondent in the oral proceedings), and while it was reasonable to expect that a higher amount of the molecular weight modifier diethyl zinc would reduce the molecular weight of the resulting copolymer, there was no convincing evidence for the Respondent's assertion that when reducing the amount of diethyl zinc in the above-referred "Experiment 3" to the amount used in Example 1 of (1) one would obtain a copolymer having a g_η* value within the range of from 0,2 to 0,8.

4.4 Upon weighing the credibility of the evidence submitted by the Appellant in the light of its imperfections (as pointed out by the Respondent), and the general disclosure of (1) referred to in section 4.2 above (which in view of the statement "proved by the properties ..." is clearly more than a mere assertion), the Board finds

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that, on the balance of probabilities, the g_η* value of the product of Example 1 of (1) cannot be assumed to within the claimed range of from 0.2 to 0.8.

4.5 In this situation the onus is clearly on the Respondent to provide sufficient counter-evidence for his assertion, which obligation, however, he did not discharge in due time.

4.6 The subject-matter of Claim 1 is therefore held to be novel over the disclosure of (1).

5. Inventive step

5.1 The closest state of the art is represented by (3), which relates to ethylene/1-butene/polyene copolymers where the polyene component is preferably selected from DCPD, EN and VN (page 4, lines 1 to 3; Claim 2).

The subject-matter of the patent is distinguished from (3) by the conjoint use of EN and VN in a molar ratio of from 1/1 to 20/1.

5.2 It was asserted by the Appellant that by that difference copolymers could be obtained having an improved balance of the following properties: tensile strength at break (equivalent to tensile stress at break) of the resulting vulcanizates, curing rate and extrudability (measured in terms of surface appearance of the extrudate).

5.3 There is no evidence before the Board which could prove that the copolymers of the patent-in-suit are superior to the copolymers of (3), insofar as either or both of the former two properties are concerned. It is, however, credible from the data in Table 2 of the patent-in-suit

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that the copolymers of the contested patent, by virtue of the conjoint use of EN and VN in a certain molar ratio, do have a surface appearance which is superior to that of the copolymers of (3); see Examples 2 and 3 having an EN/VN molar ratio of 2.3 and 9.2 (being within the claimed range) as compared to the Comparative Examples 3 and 4 having an EN/VN molar ratio of 0.75 and 30 (and being thus below and above the claimed range).

There is, furthermore, no reason to assume that the effect caused by the incorporation of EN and VN as polyenes on the extrudability of ethylene/1-butene/-polyene copolymers should significantly differ from the same effect produced in analogous ethylene/propylene/-polyene copolymers (only the latter are referred to in Table 2 of the patent-in-suit whereas (3) is solely concerned with ethylene/1-butene/polyene copolymers).

It is therefore to be expected that analogous results would be obtained when substituting 1-butene for propylene as comonomer in the copolymers referred to in Table 2 of the patent-in-suit.

5.4 It can thus be concluded that the **objective problem** to be solved by the contested patent was the provision of ethylene/alpha-olefin/polyene copolymers having an improved surface appearance of the extrudate while maintaining satisfactory tensile strength/stress at break properties and a satisfactory curing rate.

As already discussed above, this object is achieved by the subject-matter of the contested patent.

5.5 A decision on inventive step depends on whether or not it would have been obvious for the skilled person to arrive

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at the copolymers of the patent-in-suit when starting from (3) and wishing to solve the above problem.

5.5.1 Document (3), on page 3, lines 47 to 51 sets out which properties can be improved when selecting as polyene component DCPD, VN or EN: Copolymers obtained by using DCPD or VP as the polyene component "have good processability (in regard to the surface texture of the extrudate or the rate of extrusion) and superior strength characteristics ...". It is stated furthermore that copolymers obtained by using EN as the polyene component "have a fast rate of vulcanization and superior vulcanizate strength ..."

From this the skilled person would have known that he could solve the main part of the existing problem, i.e. improving the surface appearance of the extrudate, by selecting VN (or DCPD); furthermore he would have known from these statements in (3) that by changing the amount of EN, he could influence the curing rate and that he could expect a good vulcanizate strength in the case of each of the said three polyene components DCPD, VN and EN.

5.5.2 The Board does not find the Appellant's argument that (3) would militate against the use of mixtures of more than one polyene because no such mixtures are exemplified, a convincing one for the following reasons. In the absence of any specific statement or conclusive evidence in (3) warning against such a combination of polyenes, there is nothing that could deter the skilled person from the use of polyene mixtures. Furthermore it was known from (1), a document relating to the same class of ethylene/alpha-olefin/polyene copolymers, to use mixtures of polyenes and, thus, the skilled man was aware of the

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feasibility of this alternative. While it is true, as emphasized by the Appellant, that in (1) the combination of EN and VN is only one of several possible polyene combinations, this cannot detract from the fact that according to (1) copolymers with valuable properties can be obtained when using more than one polyene component (page 1, lines 28 to 59). Similarly, the fact that in (1) polyene molar ratios may be used which go beyond the EN/VN molar ratios according to Claim 1 of the patent (page 2, lines 45 to 50) cannot be considered as a deterrence for the skilled person to investigate on the optimization of the molar ratio when seeking to achieve the specific properties desired in the present case.

5.5.3 The skilled man was also informed by (3) page 4, lines 50 to 58, about the structural background and its relationship to the parameter g_η^*, i.e. that a g_η^* value in the range of 0.2 to 0.9, preferably 0.3 to 0.8, is obtained when long-chain branching or cross-linking is introduced into the copolymer by incorporating DCPD or VN, and that a g_η^* value above 0.9 impairs the processability (with consequential deterioration of the surface appearance).

5.5.4 It must therefore be concluded that (3) did offer to the skilled man all the information that was needed to solve the existing problem, namely to improve the extrudability by introducing VN and to obtain a satisfactory cure rate by introducing EN; in particular, he could have expected that he would thereby get copolymers whose vulcanizates would have the satisfactory tensile strength/stress properties contributed by both, EN and VN.

5.5.5 The Board therefore finds that the conjoint use of EN and VN as polyene component in the copolymers of (3) was

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obvious in view of the problem to be solved. Furthermore, the definition of certain molar ranges EN/VN is considered to be within the ambit of routine experimentation for the skilled person wishing to optimize the combination of properties.

5.5.6 This finding cannot be influenced by the fact that when using too low an amount of EN (about 43 mole%) in Comparative Example 3 of the patent a low tensile strength is achieved. This is not surprising, since the copolymer prepared in said comparative Example 3 has a g_η^* value of only 0.18, which is below the limit recommended in (3). It is furthermore noted that the Q value in said Comparative Example 3 (expressing the ratio of weight average and number average molecular weight) is 7.5 and thus far above the value of 3 which in (3) page 4, lines 30 to 36 is considered as a limit above which the vulcanized rubber has poor strength.

6. Conclusion

Claim 1 of the main request failing for lack of inventive step, the said request must be rejected as a whole, all the more so as no patentable features can be detected in dependent Claim 2, or in Claim 3 which is based on the same concept as Claim 1. Nor can any invention be recognized in the features of Claims 4 to 6.

7. Auxiliary request

7.1 Claim 1 of the auxiliary request restricts Claim 1 of the main request to copolymers obtained by the process of granted Claim 4. No objection under Art. 123(2) or (3) arises therefore.

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7.2 The novelty of Claim 1 of the auxiliary request follows from that of Claim 1 of the main request.

7.3 The only specific process features of Claim 1 of the auxiliary request concern the catalysts to be used. However, the catalyst systems used in (3) overlap largely with the catalyst systems of the patent-in-suit (see (3) page 4, line 59 to page 5, line 44). Consequently, the use of said catalysts does not represent a distinguishing feature over 3). Hence the inventive step considerations made for the main request apply also to the auxiliary request whose subject-matter is therefore also lacks any inventive step.

Order

For these reasons, it is decided:

The appeal is dismissed.

The Registrar:

E. Gorgmaier

The Chairman:

F. Antony

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