BESCHWERDEKAMMERN DES EUROPÄISCHEN PATENTAMTS

BOARDS OF APPEAL OF THE EUROPEAN PATENT OFFICE

CHAMBRES DE RECOURS DE L'OFFICE EUROPEEN DES BREVETS

Internal distribution code:
(A) [ ] Publication in OJ
(B) [ ] To Chairmen and Members
(C) [X] To Chairmen
(D) [ ] No distribution

D E C I S I O N
of 17 April 2003

Case Number: T 0800/01 - 3.3.3

Application Number: 95939411.5

Publication Number: 0796897

IPC: C08L 27/16

Language of the proceedings: EN

Title of invention:
Low compression-set rubber composition

Applicant:
DAIKIN INDUSTRIES, LIMITED

Opponent:
-

Headword:
-

Relevant legal provisions:
EPC Art. 54, 56

Keyword:
"Novelty - (yes)"
"Inventive step - (yes) after amendment"

Decisions cited:
G 0010/93, T 0653/93

Catchword:
-

Europäisches Patentamt | European Patent Office | Office européen des brevets

Beschwerdekammern | Boards of Appeal | Chambres de recours

Case Number: T 0800/01 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 17 April 2003

Appellant: DAIKIN INDUSTRIES, LIMITED
Umeda Center Building,
4-12 Nakazaki-nishi 2-chome
Kita-ku
Osaka-shi,
Osaka-fu 530 (JP)

Representative: Barz, Peter, Dr
Patentanwalt
Kaiserplatz 2
D-80803 München (DE)

Decision under appeal: **Decision of the Examining Division of the European Patent Office posted 14 December 2000 refusing European patent application No. 95 939 411.5 pursuant to Article 97(1) EPC.**

Composition of the Board:

Chairman: R. Young
Members: W. Sieber
R. Moufang

Summary of Facts and Submissions

I. European patent application No. 95 939 411.5, based on International application PCT/JP95/02532, filed on 11 December 1995, claiming a JP priority of 9 December 1994 (6/331865) and published under number WO 96/17890, was refused by a decision of the examining division which was announced orally on 11 October 2000 and issued in writing on 14 December 2000.

II. The decision was based on a main request (Claims 1 to 16) and an auxiliary request (Claims 1 to 11).

(i) Claim 1 of the main request read as follows:

"A rubber composition of low compression set comprising 100 parts by weight of a rubber mixture and 0.1 to 15 parts by weight of a peroxide cross-linking agent admixed therewith, the rubber mixture comprising 5 to 55 wt.% of a peroxide-crosslinkable fluororubber containing vinylidene fluoride copolymerized therein in a proportion of 45 to 88 mole % and having a number average molecular weight of 20,000 to 200,000 and 95 to 45 wt.% of an acrylic rubber containing 0.1 to 1.5 wt.% of a bifunctional monomer having two double bonds of different reactivity one of them rendering the acrylic rubber peroxide-cocross-linkable with the fluororubber and the other effecting the copolymerization with an acrylic ester."

Claims 2 to 15 were dependent claims directed to elaborations of the rubber composition of Claim 1.

Claim 16 was directed to a moulded rubber product obtained by crosslinking a composition of any one of Claims 1 to 15 with a peroxide.

(ii) Claims 1 and 2 of the auxiliary request read as follows:

"1. A rubber composition of low compression set comprising 100 parts by weight of a rubber mixture and 0.1 to 15 parts by weight of a peroxide cross-linking agent admixed therewith, the rubber mixture comprising 10 to 30 wt.% of a peroxide-crosslinkable fluororubber containing vinylidene fluoride copolymerized therein in a proportion of 45 to 75 mole % and having a number average molecular weight of 20,000 to 200,000 and 90 to 70 wt.% of an acrylic rubber containing 0.1 to 1.5 wt.% of a bifunctional monomer having two double bonds of different reactivity one of them rendering the acrylic rubber peroxide-cocross-linkable with the fluororubber and the other effecting the copolymerization with an acrylic ester.

2. A rubber composition of low compression set comprising 100 parts by weight of a rubber mixture and 0.1 to 15 parts by weight of a peroxide cross-linking agent admixed therewith, the rubber mixture comprising 10 to 50 wt.% of a peroxide-crosslinkable fluororubber containing vinylidene fluoride copolymerized therein in a proportion of 80 to 88 mole % and having a number average molecular weight of 20,000 to 200,000 and 90 to 50 wt.% of an acrylic rubber containing 0.1 to 1.5 wt.% of a bifunctional monomer having two

double bonds of different reactivity one of them rendering the acrylic rubber peroxide-cocross-linkable with the fluororubber and the other effecting the copolymerization with an acrylic ester."

Claims 3 to 10 were dependent claims directed to elaborations of the rubber composition of Claims 1 and/or 2.

Claim 11 was directed to a moulded rubber product obtained by crosslinking a composition of any one of Claims 1 to 10 with a peroxide.

III. The decision refused the main request on the ground that the subject-matter of Claims 1 to 3 and 16 was anticipated by document:

D1: EP-A-0 557 840 and/or

D2: EP-A-0 598 132.

The auxiliary request was refused because Claims 1 and 2 of this set of claims did not meet the requirement of unity of invention (Article 82 EPC).

IV. On 12 February 2001, a notice of appeal against the above decision was filed by the applicant (hereinafter referred to as the appellant) with simultaneous payment of the prescribed fee.

In the statement of grounds of appeal, filed on 20 April 2001, the appellant submitted amended Claims 1 to 9 which were argued to be novel over both D1 and D2 since the combination of parameters required in Claim 1

was neither disclosed in nor derivable from D1 and D2. As regards inventive step, the rubber compositions comprising a specific acrylic rubber in proportion of 45 wt.% or more exhibited good mechanical strength and heat resistance although the acrylic rubber was the predominant volume component in the claimed composition.

V. In a communication dated 7 February 2003 accompanying a summons to oral proceedings, the board raised objection against some of the amended claims filed on 20 April 2001 under Articles 123(2) and 84 EPC, respectively. The issue of inventive step was introduced into the proceedings based on G 10/93 (OJ EPO 1995, 172), and the question was raised whether the subject-matter of Claim 1, if allowable, was inventive in view of D2, in particular Example 2 of D2.

VI. In a letter filed on 17 March 2003, the appellant submitted a further amended set of Claims 1 to 9 which allegedly overcame the various objections raised by the board, and provided arguments as to the inventive step of the claimed subject-matter.

VII. Oral proceedings were held on 17 April 2003, in the course of which the discussion focussed on the question whether the claims filed on 17 March 2003 met the requirements of Articles 123(2), 84 and 56 EPC. In view of this discussion, the appellant filed a set of Claims 1 to 9 (main request) and a set of Claims 1 to 9 (auxiliary request 1).

(i) The main request read as follows:

"1. A rubber composition of low compression set

comprising 100 parts by weight of a rubber mixture, 0.1 to 15 parts by weight of a peroxide crosslinking agent and 0.1 to 10 parts by weight of an auxiliary crosslinking agent admixed therewith, the rubber mixture comprising 5 to 55 wt.% of an iodine-containing peroxide-crosslinkable fluororubber containing vinylidene fluoride copolymerized therein in a proportion of 45 to 75 mole % and having a number average molecular weight of 20,000 to 200,000 and 95 to 45 wt.% of an acrylic rubber containing 0.1 to 1.5 wt.% of a bifunctional monomer selected from allyl acrylate and allyl methacrylate copolymerized therein and being prepared from a combination consisting of said bifunctional monomer and a (meth)acrylic ester monomer having the formula $CH_2=C(R^1)COOR^2$ wherein R^1 is hydrogen or methyl and R^2 is alkyl or alkoxy-substituted alkyl having 1 to 8 carbon atoms."

2. A rubber composition as defined in claim 1 wherein the fluororubber is a copolymer which is comprising 45 to 75 mole % of vinylidene fluoride units, 0 to 55 mole % of tetrafluoroethylene units and 10 to 40 mole % of hexafluoropropylene units.

3. A rubber composition as defined in claim 1 or 2 wherein the proportion of copolymerized vinylidene fluoride in the fluororubber is 55 to 65 mole %.

4. A rubber composition as defined in any one of claims 1 to 3 wherein the fluororubber has a number average molecular weight of 20,000 to 70,000.

5. A rubber composition as defined in claim 3 or 4 wherein the fluororubber is obtained by polymerizing 55 to 65 mole % of vinylidene fluoride units, 15 to 25 mole % of tetrafluoro-ethylene units and 15 to 25 mole % of hexafluoro-propylene units.

6. A rubber composition as defined in any one of claims 1 to 5 wherein the acrylic rubber is a copolymer comprising 99.9 to 98.5 wt.% of C_{2-4}-alkyl (meth)acrylate units and 0.1 to 1.5 wt.% of bifunctional monomer units.

7. A rubber composition as defined in any one of claims 1 to 6 wherein the acrylic rubber contains copolymerized ethyl acrylate in a proportion of at least 40 wt.%.

8. A rubber composition as defined in any one of claims 1 to 7 which comprises 10 to 30 wt.% of fluororubber and 90 to 70 wt.% of acrylic rubber.

9. A molded rubber product obtained by cross-linking a composition of any one of claims 1 to 8 with a peroxide."

(ii) The claims of auxiliary request 1 corresponded with those of the main request with the following optional feature introduced at the end of Claim 1: "*optionally, up to 40 wt.% of the (meth)acrylic ester monomer being substituted by an ethylenically unsaturated monomer selected from acrylonitrile, styrene, vinyl acetate, ethylene or vinyl chloride*".

VIII. The appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of the main request or, in the alternative, on the basis of auxiliary request 1, both filed at the oral proceedings.

Reasons for the Decision

1. The appeal complies with Articles 106 to 108 EPC and Rules 1(1) and 64 EPC and is therefore admissible.

2. *Amendments (main request)*

2.1 Claim 1 differs from Claim 1 as filed by

(a) the incorporation of **0.1 to 10 parts by weight of an auxiliary crosslinking agent**,

(b) the definition of the peroxide-crosslinkable fluororubber as being **iodine-containing,**

(c) the restriction of the upper limit of the vinylidene fluoride content in that fluororubber to **75** mole %,

(d) the restriction of the polyfunctional monomer contained in the acrylic rubber to a **bifunctional monomer selected from allyl acrylate and allyl methacrylate**, and

(e) the indication that the acrylic rubber is **prepared from a combination consisting of said bifunctional monomer and a (meth)acrylic ester monomer having**

the formula $CH_2=C(R^1)COOR^2$ wherein R^1 is hydrogen or methyl and R^2 is alkyl or alkoxy-substituted alkyl having 1 to 8 carbon atoms.

2.1.1 Amendment (a) is supported by the paragraph bridging pages 10 and 11 of the application as filed.

2.1.2 For amendments (b) and (c), support can be found in Claims 2 and 4 as filed, respectively.

2.1.3 Amendment (d) is supported by page 8, lines 25 to 26 of the application as filed where allyl acrylate and allyl methacrylate are mentioned as preferred bifunctional monomers.

2.1.4 For amendment (e), support can be found on page 7, lines 9 to 16 of the application as filed whereby the term "consisting of" is clearly supported by Examples 1 to 6, 9 and 10 of the application as filed where the acrylic rubbers of the composition are prepared from an allyl (meth)acrylate and a (meth)acrylic ester of the above mentioned formula only.

2.2. Dependent Claim 2 finds its support in the passage bridging pages 6 and 7 of the application as filed whereby the upper limit of the vinylidene fluoride content has been amended according to Claim 1.

2.3 Dependent Claims 3 to 8 are supported by Claims 5 to 8, 10 and 11, respectively, as filed whereby Claim 6 refers to **bifunctional** monomer units as now required in Claim 1. Independent Claim 9 is supported by Claim 19 as filed.

2.4 Summing up, Claims 1 to 9 meet the requirements of

Article 123(2) EPC.

3. Unity of invention (main request)

The Board holds that the set of amended claims meets the requirements of Article 82 EPC because all claims are linked by a single general inventive concept, ie the rubber composition as defined in Claim 1.

4. *Sufficiency and clarity (main request)*

No objections under Articles 83 and 84 EPC were raised by the examining division. The board is satisfied that also the amended claims meet the requirements of Articles 83 and 84 EPC.

5. *Novelty (main request)*

5.1 Document D1

5.1.1 According to D1, an improvement of the low temperature flexibility, the amount of filler which could be added to, and the processability of a fluororubber composition can be achieved by blending from 35 to 98 parts by weight of a peroxidically vulcanisable fluororubber and from 2 to 65 parts by weight of an acrylic rubber where the acrylic rubber is a partially crosslinked acrylic rubber having gel contents of between 20 and 29% by weight and particle diameters (d_{50} values) of from 60 to 800 nm. According to column 3, lines 32 to 35, suitable fluororubbers contain units of vinylidene fluoride (VDF) and of at least one other copolymerisable fluoroolefin. Apart from the two tangible fluororubbers prepared in Examples 1 and 8 (78.9 and 69 mole % VDF content,

respectively), the VDF content for the fluororubbers is not indicated. The reactive sites for peroxidic crosslinking in the fluororubber may be bromine and/or iodine substituents or double bonds (column 3, lines 43 to 47). Suitable acrylic rubbers are at least partially crosslinked rubber-type copolymers consisting of one or several not less than C_3-alkyl acrylates and a polyfunctional polyvinyl- or allyl compound capable of copolymerization. Preferred polyvinyl- or allyl compounds are - *inter alia* - allyl acrylate and methacrylate, with triallyl cyanurate and triallyl isocyanurate being particularly preferred (column 4, lines 9 to 23). In order to produce the elastomers, ie the vulcanized rubber, from the rubber mixtures, the latter are mixed in a conventional manner with radical initiators as well as with other auxiliaries, such as co-crosslinking agents (column 5, lines 12 to 16). Preferred radical initiators are peroxides (column 5, lines 23 to 26). Suitable co-crosslinking agents are in particular compounds with several double bonds such as triallyl cyanurate and triallyl isocyanurate (column 5, lines 34 to 40).

5.1.2 It follows from the analysis above that the subject-matter of amended Claim 1 lies within the more general disclosure of D1. The emphasis in D1 is, however, on a different spectrum of properties. Thus, in order to arrive at something falling within the scope of Claim 1 of the main request one would have to pick and choose, ie make a "multiple selection" (ie at least fivefold) from the generic disclosure and a specific example of D1. In particular one would have to select (i) an appropriate ratio of fluororubber/acrylic rubber, (ii) an iodine-containing fluororubber, (iii) an appropriate VDF content for the fluororubber, (iv) allyl acrylate

or allyl methacrylate as the bifunctional monomer for the acrylic rubber, and (v) a vulcanizing system comprising a co-crosslinking agent in the appropriate amount.

5.1.3 According to decision T 653/93 (21 October 1996; paragraph 3.2 of the reasons; not published in the OJ EPO), in case of "multiple selection", the question of novelty cannot be answered by contemplating the ranges of various parameters separately. Moreover, one would have to show that the "combined selection" emerges from the prior art.

5.1.4 In the present case, the skilled person in the art had, when applying the teaching of D1, no reason to concentrate on the combination of the above mentioned parameters (i) to (v). Such a combined selection is neither explicitly disclosed in nor derivable from D1. Thus, Example 8, the only disclosure in D1 of a fluororubber having the required VDF content, provides no indication as to the other required features: the fluororubber contains bromine cure sites, ie not iodine cure sites, the amount of the fluororubber in the mixture is, with 90 weight percent, a long way outside the range of 5 to 55 weight percent required in Claim 1, and the crosslinking monomer used in the acrylic rubber of Example 8 is triallyl cyanurate, ie not allyl acrylate or allyl methacrylate.

5.1.5 It follows from the above that the subject-matter of Claim 1 is not disclosed in D1.

5.2 Document D2

5.2.1 D2 discloses in Claim 1 a crosslinkable composition

which comprises an internally crosslinked acrylic elastomer which is crosslinkable with a peroxide, a fluoroelastomer and a crosslinking agent for at least one of the elastomers. The ratio of the acrylic elastomer/fluoroelastomer is 5 to 90/95 to 10 by weight and more preferably 20 to 50/80 to 50 (page 5, lines 12 to 14). The acrylic elastomer can be prepared by polymerizing (meth)acrylic monomer, crosslinkable monomer and multifunctional monomer (page 3, lines 51 to 52; Claim 2). Examples of useful crosslinkable monomers are vinylsilyl-containing compounds which are preferably used in an amount of 0.1 to 10 parts by weight per 100 parts by weight of the combined amount of (meth)acrylic monomer, crosslinkable monomer and multifunctional monomer (page 4, lines 1 to 5). Examples of useful multifunctional monomers are - *inter alia* - allyl (meth)acrylate, ethylene glycol di(meth)-acrylate and 1,4-butanediol di(meth)acrylate, used in an amount of 0.1 to 10 parts by weight per 100 parts by weight of the combined amount of (meth)acrylic monomer, crosslinkable monomer and multifunctional monomer (page 4, lines 53 to 58). As regards the fluororubber, D2 mentions on page 5, lines 6 to 11, VDF-containing fluoroelastomers with VDF/hexafluoropropylene and VDF/tetrafluoroethylene/ hexafluoropropylene elastomers being preferred. The peroxide is used in an amount of 0.1 to 10 parts by weight per 100 parts by weight of the combined amount of acrylic elastomer and fluoro-elastomer (page 5, lines 27 to 29). When required, an auxiliary crosslinking agent, eg triallyl isocyanurate or triallyl cyanurate, can be used conjointly to achieve an improved crosslinking efficiently and afford improved physical properties (page 5, lines 23 to 26).

5.2.2 Thus, D2 relates likewise to mixtures comprising a

fluororubber and an acrylic rubber, both defined in rather broad terms, so that the issue of novelty again hinges on the question whether D2 discloses the specific fluororubber in combination with the specific acrylic rubber as required in amended Claim 1.

5.2.3 D2 lists on page 5, lines 6 to 11 *inter alia* vinylidene fluoride-containing fluororubbers, however, without providing further details on these fluororubbers such as the VDF content, the number average molecular weight or the presence of iodine in the rubber. As regards the acrylic rubber, D2 refers only to an acrylic rubber which can be prepared from acrylic or methacrylic monomer, crosslinkable monomer and multifunctional monomer exemplified by copolymerizing the combination of these monomers by a common method of polymerization. There is no disclosure, either explicit or implicit, of an acrylic rubber being prepared from a combination consisting of allyl (meth)acrylate and a (meth)acrylic ester monomer. In other words, there is no disclosure of an acrylic rubber where the only monomer with more than one double bond is allyl (meth)acrylate.

5.2.4 As regards the specific examples of D2, Example 2 is the example which comes nearest to the claimed subject-matter. But although Example 2 has some of the features required in Claim 1 of the main request, such as the weight ratio of fluororubber/acrylic rubber, the peroxide crosslinking agent and the co-crosslinking agent, neither the fluororubber nor the acrylic rubber meet the requirements of Claim 1. The fluororubber used in Example 2 is Dai-el G 801, an iodine-containing two-component copolymer fluoroelastomer prepared from vinylidene fluoride and hexafluoropropylene (page 6, lines 17 and 18). But, according to the appellant which

is also the producer of this commercial product, Dai-el G 801 has a VDF content of 78 mole % which is just outside of the range required in Claim 1 of the main request. The acrylic rubber used in Example 2 is prepared from ethyl acrylate, a vinylsilyl-containing monomer (crosslinkable monomer) and 1,4-butanediol diacrylate (multifunctional monomer). Thus, the acrylic rubber is not prepared from allyl (meth)acrylate but from other bifunctional monomers which are excluded by the definition of the acrylic rubber in Claim 1 of the main request.

5.2.5 Summing up, D2 does not disclose the claimed subject-matter.

5.3 It follows, in view of the above, that Claim 1 and, by the same token, Claims 2 to 9 are novel over D1 or D2 and meet the requirements of Article 54 EPC.

6. *The application in suit, the technical problem (main request)*

6.1 The application in suit is concerned in general terms with rubber compositions and more particular with compositions which comprise 5 to 55 wt.% of a specific fluororubber and 95 to 45 wt.% of a specific acrylic rubber and to products moulded therefrom. These compositions are excellent in processability and capable of giving mouldings which are excellent in compression set (page 1, lines 4 to 8 of the application as filed) whereby the values for the compression set obtained in Examples 1 to 6, 9 and 10 range from 13.1 to 21.5% (72h at 175°C).

6.2 As mentioned on page 2, lines 17 to 28 of the

application as filed, it was known to blend a fluororubber and an acrylic rubber partially cross-linked with a polyfunctional monomer, but the prior art techniques were effective only for compositions comprising a predominant amount of fluororubber. None of the previously available compositions containing larger amounts of acrylic rubber was satisfactory in such properties as compression set, heat resistance and processability (page 3, lines 23 to 26). Thus the present application focuses on rubber compositions comprising acrylic rubbers as the predominant volume component. As can be seen from a comparison of the specific gravities of the fluororubbers (1.80 to 1.82) with those of the acrylic rubbers (1.09 to 1.10), a weight ratio of about 62 wt.% fluororubber and about 38 wt.% acrylic rubber (1.81/1.095 = 62/38) corresponds to equal volumes of both rubbers. In other words, a proportion of more than about 38 wt.% acrylic rubber occupies more than half of the volume of the total rubber composition.

6.3 Rubber compositions comprising a fluororubber and an acrylic rubber which are capable of giving mouldings which are excellent in physical properties such as mechanical strength and compression set, heat resistance and workability are known from D2 (Claim 1; page 2, lines 33 to 35). Although the ratio of the acrylic elastomer/fluoroelastomer is 5-90/95-10 by weight (Claim 9), most of the examples in D2 use only 30 wt.% of acrylic rubber in the composition, so that the acrylic rubber is not the predominant volume component. Only Examples 2 and 8 use 50 wt.% of acrylic rubber, ie an amount falling within the range required in Claim 1. Since only Example 2 uses an iodine-containing fluoroelastomer, this example is considered

by the board as the closest prior art and the appropriate starting point for the assessment of inventive step.

6.4 At first glance, a comparison of the compression set of Example 2 of D2 (22% after 70 h at 150°C; Table 2) and Example 4 of the application in suit (14.8 % after 72 h at 175°C; Table 5), employing also a 50/50 ratio of a fluoroelastomer and an acrylic rubber, might lead to the conclusion that the prior art provides equally good compression set. However, as pointed out by the appellant it is expected that the compression set of 22% after 70 h at 150°C obtained in Example 2 of D2 translates into a compression set of 30 to 35% under the conditions employed in the application in suit (72 h at 175°C) which is much higher than the value of 14.8% obtained for the composition of Example 4 in the application in suit. Thus, the objective technical problem may be seen in the provision of rubber compositions with a predominant acrylic rubber portion having a lower compression set than the rubber composition of the closest prior art.

6.5 The solution proposed according to Claim 1 of the main request is the combination of a specific fluororubber and a specific acrylic rubber. In view of the above mentioned comparison of the compression set reported for Example 2 of D2 and Example 4 of the application in suit, the board finds it plausible that the claimed measures provide an effective solution to the stated problem.

7. *Inventive step*

7.1 It remains to be decided if the proposed solution, ie

the use of a specific fluororubber in combination with a specific acrylic rubber, is obvious from the prior art.

7.2 In D2 itself, there is no suggestion as to how the compression set properties of rubber compositions comprising a relatively high proportion of acrylic rubber might be further improved, let alone a hint to the specific acrylic rubber now required in Claim 1.

7.3 Document D1 would not give any hint to the solution proposed by the application in suit, since, as indicated above, it is not directly concerned with the relevant technical problem, and in any case does not disclose the specific acrylic rubber.

7.4 The board is satisfied that the disclosure of the remaining document mentioned in the supplementary European search report is still more remote.

7.5 In summary, the solution (combination of a specific fluororubber and a specific acrylic rubber) of the stated problem does not arise in an obvious way from the cited prior art documents. Thus, the subject-matter of Claim 1, and, by the same token, the subject-matter of Claims 2 to 9 involves an inventive step within the meaning of Article 56 EPC.

8. It follows, in view of the above, that a patent can be granted on the basis of the Claims 1 to 9 of the main request. Consequently, there is no need to consider the introduction of auxiliary request 1 into the 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 grant a patent on the basis of the set of claims 1 to 9 filed as main request at the oral proceedings and after any necessary consequential amendment of the description.

The Registrar: The Chairman:

E. Görgmaier R. Young