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DECISION
of 19 February 1999

Case Number: T 0941/94 - 3.3.3

Application Number: 86110782.9

Publication Number: 0211431

IPC: C08L 27/16

Language of the proceedings: EN

Title of invention:
Covulcanizable compositions of fluoroelastomers having improved chemical stability

Patentee:
Ausimont S.p.A.

Opponent:
E.I. Du Pont de Nemours & Company, Inc.

Headword:
-

Relevant legal provisions:
EPC Art. 56

Keyword:
"Inventive step (yes) - ex post facto analysis"

Decisions cited:
-

Catchword:
-

Case Number: T 0941/94 - 3.3.3

DECISION
of the Technical Board of Appeal 3.3.3
of 19 February 1999

Appellant:
(Opponent)

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Decision under appeal: **Interlocutory decision of the Opposition Division of the European Patent Office posted 21 October 1994 concerning maintenance of European patent No. 0 211 431 in amended form.**

Composition of the Board:

Chairman: C. Gérardin
Members: A. Däweritz
A. Lindqvist

Summary of Facts and Submissions

I. The mention of the grant of European patent No. 0 211 431 in respect of European patent application No. 86 110 782.9 filed on 4 August 1986 and claiming priority of 5 August 1985 of an earlier application in Italy (2186785), was announced on 27 May 1992 (Bulletin 92/22) on the basis of ten claims.

Claim 1 as granted read as follows:

"Covulcanizable compositions of fluoroelastomers comprising:

A) 55-100% by weight of a fluoroelastomer consisting of 40-85 mol% of vinylidene fluoride, 15-35 mol% of perfluoropropene, 0-30 mol% of tetrafluoroethylene, and containing from 0.001 to 2% by weight of bromine;

B) 0-45% by weight of a copolymer consisting of 40-60 mol% of tetrafluoroethylene and 40-60 mol% of propylene;

C) 0.7-2.5 parts per 100 parts of (A) + (B) of an organic polyhydroxy compound;

D) 0.3-1.5 parts per 100 parts of (A) + (B) of a quaternary ammonium, phosphonium or aminophosphonium compound;

E) 0.4-5 parts per 100 parts of (A) + (B) of an organic peroxide;

F) 1-5 parts per 100 parts of (A) + (B) of a peroxide crosslinking coagent."

Claims 2 to 10 were directed to preferred embodiments of the compositions according to Claim 1.

II. On 26 February 1993, a Notice of Opposition was filed in which revocation of the patent in its entirety was requested on the grounds of lack of inventive step within the meaning of Article 56 EPC. In the course of oral proceedings held on 20 September 1994, the Opponent for the first time raised an objection of lack of novelty (Article 54(1) and (2) EPC) which was admitted into the proceedings by the Opposition Division.

These objections were supported essentially by the following documents:

D1: US-A-3 876 654,

D2: US-A-4 214 060,

D3: R.D. Stevens in "Rubber World", 190, No. 6, September 1984, pages 31 to 37, 52 "Compounding fluoroelastomers for resistance to extrusion at high temperatures, pressures" (including a better copy (D3a) of page 36 of D3).

III. In an interlocutory decision announced orally on 20 September 1994 and issued in writing on 21 October 1994, the Opposition Division held that the grounds for opposition did not prejudice the maintenance of the patent in amended form in accordance with an auxiliary request, the amendment consisting in the addition of the following disclaimer at the end of Claim 1:

"a composition consisting of

(a) 100 parts by weight of a copolymer consisting of 35% by weight of vinylidene fluoride, 34% by weight of hexafluoropropylene, 29% by weight of tetrafluoroethylene and 2% by weight of bromotetrafluorobutene;

(b) 1.7 parts by weight of a composition consisting of 33% organophosphonium salt plus 67% fluoroelastomer;

(c) 4.0 parts by weight of a composition consisting of 50% dihydroxy aromatic compound plus 50% fluoroelastomer;

(d) 6 parts by weight of ZnO;

(e) 2 parts by weight of $Ca(OH)_2$;

(f) 60 parts by weight of MT black;

(g) 25 parts by weight of silica;

(h) 1.3 parts by weight of process aids;

(i) 3 parts by weight of triallyl isocyanurate; and

(j) 3 parts by weight of 2,5-dimethyl-2,5-bis(t-butylperoxy)hexane

being excluded."

Claims 2 to 10 remained unamended.

In substance the Opposition Division argued as follows:

(i) The novelty objection was overcome by this disclaimer.

(ii) Starting from D2, which was regarded as representing the closest prior art, it was not considered obvious to modify the known covulcanizable composition in accordance with the requirements of the patent in suit in order to improve the processing safety, the chemical and thermal stability especially against aggressive oils as well as the mechanical properties of the known compositions. Consequently, an inventive step was acknowledged.

IV. On 8 December 1994, a Notice of Appeal was filed by the Opponent (Appellant) against this decision together with payment of the prescribed fee.

(i) In the Statement of Grounds of Appeal filed on 21 February 1995 as well as in a later submission filed 16 January 1999, the Appellant disputed the findings of the Opposition Division regarding the issue of inventive step, in particular the choice of D2 as representing the closest state of the art. A correct approach based on D3 showed that the combination of cross-linking agents would have been obvious to a person skilled in the art.
To support its arguments the Appellant relied on further additional documents:

D13: Brochure "Viton Fluoroelastomer" by Du Pont, "Two-Stage Cures in Viton VT-R-5362", dated 1 May 1980,

D14: Materials Engineering, September 1981, pages 5 and 24, D15: Brochure "Viton GF - An improved fluid-resistant fluoroelastomer" by J. D. Eddy and J. G. Bauerle, Du Pont, E-42089, June 1981, and

D16: Brochure "Viton B" by A. L. Moran, Elastomer Chemicals Department, Report No. 59-4, October 1959.

(ii) Together with the submission filed on 16 January 1999, an experimental report was submitted in order to support the argument that D3 represented the closest state of the art. The report was also intended to demonstrate the influence of fillers on the mechanical properties of Viton® GF fluoroelastomer compositions.

V. In its counterstatement, the Respondent (Proprietor) objected to the late citation of the additional documents, requested that these documents as well as the additional experimental data should not be admitted into the proceedings and argued that a proper problem-solution approach could only be based on D2 as representing the closest state of the art.

In substance, the Respondent argued along the same lines as the first instance in the interlocutory decision.

VI. During oral proceedings held on 19 February 1999, following a preliminary discussion of the relevance of D3, both parties presented their arguments on the basis of D2. Whilst the Appellant took the view that a simple combination of D2 with either D1 or D3 would lead to the claimed subject-matter, the Respondent argued that the fluoroelastomers of D1 did not contain any bromine-cure sites and that the disclosure of D3 was rather speculative so that any suggestion as to a combination of any of these documents with D2 was based on hindsight.

VII. 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 and that the patent be maintained.

Reasons for the Decision

1. The appeal is admissible.

2. *Procedural matters*

2.1 The experimental test report submitted by the Appellant on 16 January 1999, i.e. four years after the filing of the appeal and one month before the oral proceedings, has to be regarded as late filed as it does not give the Respondent adequate time to review these results and to prepare a substantiated reply or to perform its own counter-experiments. In fact, as will appear hereinafter, the issue of inventive step does not depend on particular properties of the composition, but mainly on the non-obviousness of the cross-linking system used.

2.2 The additional documents submitted with the Statement of Grounds of Appeal and with the later submission have been duly examined by the Board, but not found sufficiently relevant to be admitted into the proceedings (Article 114(2) EPC).

3. Claim 1 as amended differs from Claim 1 as granted only by the addition of a disclaimer at the end of the claim. The wording of this disclaimer corresponds exactly to composition E-6 in Table 5 of D3, which was necessary to overcome the objection of lack of novelty. The Board concurs with the finding of the Opposition Division that Article 123(2) and (3) EPC is complied with.

4. *Novelty*

Following the introduction of the disclaimer in Claim 1, the issue of novelty was no longer raised by the Appellant, and the Board has no reason to take a different view of this question.

5. *Problem and Solution*

5.1 The patent concerns covulcanizable compositions of fluoroelastomers having improved chemical stability.

5.2 Such fluoroelastomer compositions are disclosed in D2, which the Board, like the Opposition Division and the parties during the oral proceedings, regards as the closest state of the art.

This citation describes fluoropolymer compositions which comprise

"(A) a copolymer whose interpolymerised units consist essentially of (1) up to 3 mol% of units derived from a bromine-containing olefin with the proviso that enough of such units are present to provide at least 0.05 weight% bromine in the copolymer, and (2) enough units derived from component (2.1) or (2.2) as follows to make up the remainder of the copolymer:

(2.1) at least one fluorine-containing monomer which is copolymerizable with component (1), contains 2-7 carbon atoms, contains no bromine atom, and contains at least as many fluorine atoms as carbon atoms, and

(2.2) about 50-99 mol% of tetrafluoroethylene, about 1-50 mol% of a C_2-C_4 olefinic hydrocarbon and from 0 to about 30 mol% of vinylidene fluoride, and

(B) about 0.5-10% of an organic peroxide based on the weight of component (A)" (Claim 1).

These compositions can easily be made in vulcanizable grades suitable for compounding and curing by practical and economical methods to yield plastic and elastomeric articles which, in their cured state, exhibit good creep-resistance and good resistance to damage by heat, solvents and corrosive chemicals. This makes them useful for applications such as films, gaskets, O-rings, coated fabrics, wire insulations, hoses, and protective coatings (see column 5, line 59 to column 6, line 9).

5.3 In its arguments presented in the oral proceedings, the Respondent confirmed its position already set out in the description of the patent in suit (see page 2, lines 39 to 43), that the known fluoroelastomer compositions suffered from shortcomings in terms of processing safety as well as thermal and chemical stability against corrosive media (fluids resistance) under severe temperature conditions, and pointed to its experimental data submitted on 15 October 1993.

5.4 In line with this submission and in accordance with the introductory statement in the patent specification (page 2, lines 5 to 43), the technical problem underlying the patent in suit was thus to provide compositions exhibiting good mechanical properties and showing improved stability against deterioration of these properties when exposed to aggressive fluid media at elevated temperatures over extended time periods.

5.5 According to the patent in suit, this technical problem is solved by a combination of a peroxide cross-linking (cure) system (components E) and F) in Claim 1) and an ionic cross-linking (cure) system (components C) and D) in Claim 1).

5.6 The experimental evidence on file, i.e. the examples in the patent specification and in the evidence filed during the opposition proceedings, shows that thereby the desired level of properties is not only achieved, but also maintained upon exposure to the severe conditions mentioned above. These properties render the compositions suitable for their intended uses referred to above, such as oil seal rings (see page 2, lines 9 to 14, 30 and 31 of the patent specification). More particularly, reference is made to the tests carried out in the examples of the patent specification, wherein the compositions were exposed to an oil SF at 150°C for 21 days (see the tables at the top of pages 6 and 7), and to the tests in the report filed on 15 October 1993 including such an exposure for 7 and 14 days, respectively. No counter-experiments or convincing arguments disproving these results were submitted. Thus, it is concluded that the required combination of features provides an effective solution of the above defined technical problem.

.../...

6. *Obviousness*

It remains to be decided whether the solution specified in Claim 1 was obvious to a person skilled in the art having regard to the state of the art relied upon by the Appellant.

6.1 As indicated above, D2 differs from the subject-matter claimed in the patent in suit in that the ionic cure system (components C) and D)) is not used. Table III (see column 19 of D2) shows mechanical properties of peroxide-cured compositions based on elastomeric bromine-containing vinylidene fluoride copolymers. Although the table refers to individual values for the tensile strength of up to 1700 or 2000 psi (11.7 or 13.8 MPa), for the elongation at break of up to 200 or 255% and for the 100% modulus of up to 550 or 1600 psi (3.8 or 11.0 MPa), each of these compositions shows poorer properties in at least one of these aspects. This unsatisfactory balance of properties is *expressis verbis* addressed e.g. in column 18, line 66 to column 19, line 2. Moreover, the document is silent about the stability of the compositions when exposed to aggressive fluids at elevated temperatures for an extended time.

In fact the disclosure of this citation must be regarded as an attempt to overcome specific difficulties, namely excessive compression set values and tendency to degradation, which occurred with prior art cure systems, such as amine or ionic cure systems (see column 6, first complete paragraph). At that time such systems were thus regarded as unsatisfactory. Consequently, when considering D2 in isolation there was no incentive to revert to the use of these systems, let alone to envisage the use of combinations of peroxide and amine or ionic cure systems.

6.2 D1 concerns a bromine-free fluoroelastomer composition which is cross-linked by means of an ionic cure system comprising an organic polyhydroxy compound and a quaternary phosphonium accelerator (Claim 1 in conjunction with column 2, line 50 to column 3, line 11). These compositions are suitable for the manufacture of special articles, such as high temperature-resistant gaskets, seals, diaphragms and tubing (column 1, lines 14 to 19) as well as O-rings (as in Example 43 *et seq.*). The document refers to good flow properties due to the absence of premature gelation, satisfactory cure rates with good processing safety and good storage stability of the uncured formulations. The formulations yield vulcanised products of good stress / strain properties and low compression set (see column 7, lines 15 to 25). Compression set, hardness, 100% modulus, tensile strength and elongation at break values of press-cured samples, which were then post-cured at elevated temperatures, are given. The compression set values of O-rings are additionally provided which had been subjected to compression at high temperatures (see e.g. Table V in column 13).

The document does not, however, provide any data concerning the properties of the cured articles towards aggressive fluids and cannot, consequently, contribute to the solution of the above-defined technical problem.

6.3 D3 reports experiments on fluoroelastomer compositions and the influence of the components in these compositions on extrusion resistance at high temperatures and pressures, as occur e.g. in high pressure oil field service.

6.3.1 Fluoroelastomers in general are said to be well-known for their excellent balance of heat and fluids resistance. The chemical resistance of fluoroelastomers to oil field environments, a subject under study by many companies, is not significant unless the fluoroelastomer can first seal effectively (see the middle of the left column on page 31).

As a result of the survey, it is concluded on page 37 that not all tested fluoroelastomer compounds perform in a similar manner when extrusion resistance is measured; in particular, the following trends are noted: Dipolymers are considered the best, terpolymers are about equal to peroxide-cured polymers, the high molecular weight polymers perform better than low molecular weight polymers, bisphenol cure is regarded as the best system for dipolymers and terpolymers, a combination of cure systems is best for "FKM-GF". Additionally the influence of fillers is mentioned. The identity of "FKM-GF" referred to in D3 (Table 5, Composition E-6) with component A) as defined in Claim 1 of the patent in suit is no longer disputed.

6.3.2 The test series is divided in some screening studies to examine, first, the properties of polymers cured with bisphenol and peroxide (Tables 3 and 5), secondly, the effect of some fillers on certain polymers (Table 4) and, thirdly, the extrusion resistance of fluoroelastomer stocks in comparison to high quality nitrile compounds (Table 6). In this decision, the latter two series of the studies need not be considered.

6.3.3 Peroxide-cured fluoroelastomers are regarded as of interest because they have been known to have superior steam and acid resistance compared to bisphenol-cured compounds (page 34, left column, page 35, right column lines 1 to 5, and page 37, right column, lines 1 to 5).

According to the right column of page 34, line 15 *et seq.*, the peroxide-cured FKM-GF has been found similar in extrusion resistance to the straight bisphenol- or diamine-cured versions of the same polymer, but, "with the sacrifice in fluids resistance that the latter cure systems dictate, they seem of little interest." The fluids resistance is said to be "under investigation." Due to the use of the peroxide cure system, "it is speculated that the extra fluids resistance of FKM-GF" may not be impaired. In the Board's view, the reference to experimental studies not completed and to the relative uncertainty as to the actual level of fluids resistance to be expected casts serious doubts on the conclusiveness of that teaching.

6.3.4 A number of experiments are based on FKM-GF cured with a combined bisphenol / peroxide cure system. Reference can be made to compositions C-4 and E-6. To what extent this also applies to E-2 to E-5 is unclear. First, some remarks to the latter examples: The heading in Table 5 indicates that at least some of the compositions E-2 to E-6 were cured by means of a combination of peroxide and bisphenol cure systems. The first paragraph on page 36 commenting on the results refers to "some of the compounds" but it fails to identify these compositions, except for E-4. Secondly, the detailed recipes in Table 5 indicate that the peroxide cure system (TAIC and peroxide) was only added to E-6. The recipes of compositions E-1 to E-5 are silent as to any amounts of peroxide and TAIC (triallyl isocyanurate), and - as far as E-2 and E-3 are concerned - this is also true for the bisphenol cure system ("Cure #20" and "Cure #30", which have been identified as phosphonium and bisphenol compounds).

In any case, the results of the compositions for which there is evidence for the combined cure system (C-4, E-4 and E-6) demonstrate that the hardness was increased (see in particular the Shore D values), and the elongation at break was reduced to below 100% in each case, irrespective of the filler amounts. This limit, however, is regarded as essential for O-rings, V-packings, T-rings, other high pressure seals and packers (see the penultimate paragraph on page 37).

6.3.5 In conclusion, in view of the absence in D3 of any reference to long term stability towards aggressive chemical media at elevated temperatures, the mention of "FKM-GF" cured by a peroxide and a bisphenol cannot be an incentive for a skilled person to consider a solution along that line.

6.4 It is not disputed that by selecting appropriate features from the various documents it may be possible to formulate fluoroelastomer compositions within the terms of the patent in suit. However, since these features are disclosed in technical contexts which are not related to the technical problem, their combination in accordance with the Appellant's line of argument can only be made by hindsight.

6.5 For these reasons, the prior art citations relied upon by the Appellant, whether considered in isolation or in combination, cannot render obvious the claimed subject-matter, which therefore involves an inventive step.

7. Claim 1 being allowable the same applies to Claims 2 to 10, which relate to preferred embodiments of the composition of claim 1 and whose patentability is supported by that of the main composition claim.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar:

E. Görgmaier

The Chairman:

C. Gérardin

C. Gérardin