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**D E C I S I O N**  
of 7 March 1996

**Case Number:** T 0088/94 - 3.3.3

**Application Number:** 86103004.7

**Publication Number:** 0193963

**IPC:** C09D 127/18

**Language of the proceedings:** EN

**Title of invention:**

Aqueous dispersion comprising fluorine-containing copolymer and article coated therewith

**Patentee:**

DAIKIN INDUSTRIES, LIMITED

**Opponent:**

HOECHST AKTIENGESELLSCHAFT

**Headword:**

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**Relevant legal provisions:**

EPC Art. 56

**Keyword:**

"Inventive step (no) - missing evidence for critical character of selected features"

**Decisions cited:**

T 0002/83

**Catchword:**

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Boards of Appeal

Chambres de recours

Case Number: T 0022/94 - 3.3.3

**D E C I S I O N**  
of the Technical Board of Appeal 3.3.3  
of 7 March 1996

**Appellant:**  
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**Respondent:**  
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**Representative:**      -

**Decision under appeal:**      Decision of the Opposition Division of the  
European Patent Office posted 1 December 1993  
revoking European patent No. 0 193 963 pursuant to  
Article 102(1) EPC.

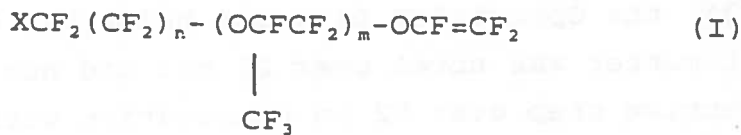
**Composition of the Board:**

**Chairman:**      C. Gérardin  
**Members:**      P. Kitzmantel  
                    J. A. Stephens-Ofner

Summary of Facts and Submissions

I. European patent application No. 86 103 004.7 in the name of Daikin Industries, Limited which had been filed on 6 March 1986, claiming priority from two JP applications filed on 6 March 1985 and 26 June 1985, respectively, resulted in the grant of European patent No. 0 193 963 on 23 October 1991, on the basis of 5 claims, independent Claims 1 and 5 reading as follows:

"1. An aqueous dispersion comprising colloidal particles of a copolymer of tetrafluoroethylene and a fluorovinyl ether of the formula:



wherein X is a hydrogen atom or a fluorine atom, n is an integer of 0 to 7 and m is an integer of 0 to 3 which contains 1 to 10 % by weight of the fluorovinyl ether units and has a specific melt viscosity of  $0.3 \times 10^4$  to  $10.0 \times 10^4$  poise ( $0.03 \times 10^4$  to  $1 \times 10^4$  Pas) and an average particle size of 0.3 to 1  $\mu\text{m}$  and an anionic or nonionic surfactant as a stabilizer."

"5. An article coated with a non-tacky coating prepared applying an aqueous dispersion of claim 1 on a surface of the article and drying and baking the dispersion to form a coating of the copolymer on the surface."

Claims 2 to 4 relate to preferred embodiments of the dispersion according to Claim 1.

II. Notice of Opposition was filed by Hoechst AG on 12 July 1992 requesting revocation of the patent in its entirety, on the grounds of Article 100(a) EPC. The Opponent contended in particular that the claimed subject-matter lacked novelty and/or inventive step i.a. over the documents

D1: EP-A-0 014 974,

D2: DE-A-2 145 960 and

D4: US-A-3 132 123 (submitted later in the opposition proceedings).

III. In its decision to revoke the opposed patent, announced orally on 15 November 1993 (written decision posted on 1 December 1993), the Opposition Division held that the claimed subject-matter was novel over D1 but did not involve an inventive step over D2 in combination with D4. In particular, D2 disclosed colloidal aqueous dispersions of homo- and copolymers of tetrafluoroethylene (TFE) which, owing to their large particle size (mean diameter of at least 0,25  $\mu\text{m}$ ), could be processed into crack-free coatings having increased thickness. With respect to possible comonomers, D2 referred to D4 which disclosed the use of perfluoropropyl perfluorovinyl ether (PPVE) as comonomer for TFE (which, according to the opposed patent, is the preferred fluoro vinyl ether comonomer) in an amount corresponding to that required by present Claim 1 to yield TFE-PPVE copolymers having a melt viscosity being as well within the scope of Claim 1. The transfer of these features of D4 into the dispersions according to D2 did not involve an inventive step.

IV. On 1 February 1994 the Patentee (Appellant) lodged an appeal against the decision of the Opposition Division and paid the appeal fee on the same day. A Statement of Grounds of Appeal was submitted on 7 April 1994.

In that statement as well as during oral proceedings held on 7 March 1996 the Appellant argued essentially that the features of Claim 1 concerning the choice of the comonomer and its amounts, as well as the ranges of the specific melt viscosity of the copolymer and of its average particle size were of critical importance for the solution of the problem to be solved, i.e. the provision of an aqueous copolymer dispersion capable of being processed into thick ( $\geq 25 \mu\text{m}$ ) and smooth (surface roughness  $< 0,5 \mu\text{m}$ ) coatings. This would be shown by the experimental evidence reported in the opposed patent, particularly in Tables 1 and 2.

Document D2, in the Appellant's opinion, did not contain any pointer towards the selection of those critical features; nor would the skilled person wishing to solve the above mentioned problem have had any reason to assume that the TFE-PPVE copolymer disclosed in Example II of D4, which copolymer had a PPVE content and a melt viscosity within the ranges required by Claim 1 of the opposed patent, would be helpful in this respect.

- V. The Respondent (Opponent) argued essentially that D2 already solved the problem of providing crack-free thick coatings from aqueous dispersions of TFE homo- and copolymers. The measures taken to this end in D2 as well as according to the opposed patent would be the same, namely the choice of an increased particle size of the dispersion. The second part of the problem allegedly underlying the patent in suit, i.e. the provision of coatings having a smooth surface, was closely related to the melt viscosity of the copolymer, which should be sufficiently low, and by that also to the kind and amount of comonomer units. The choice of PPVE or similar fluorovinyl ethers as comonomers for TFE was obvious not only on the face of the few alternatives disclosed in D2, but also because such copolymers (generally

designated "PFA"), together with copolymers from TFE and hexafluoropropylene (generally designated "FEP"), would be the only commercially feasible copolymers in the field of melt processable perfluorinated copolymers. Moreover, PFA, the product selected according to the patent in suit, was known for its outstanding temperature resistance.

VI. The Appellant requested that the decision under appeal be set aside and the patent be maintained as granted.

The Respondent requested that the appeal be dismissed.

#### Reasons for the Decision

1. The appeal is admissible.

2. *Novelty*

The novelty of the subject-matter of the patent in suit was acknowledged in the appealed decision and was not attacked in the appeal proceedings. The Board sees no reason to question it either.

3. *Closest prior art*

Document D2 relates to aqueous dispersions of TFE polymers having large particles which, as opposed to dispersions with smaller particles, are processable into crack-free coatings of increased thickness. In order to alleviate the increased sedimentation tendency of the larger particles, D2 recommends the addition of certain amounts of a non-ionic surface active agent (Claim 1; page 2, last paragraph to page 3, 3rd paragraph).

According to the statements at page 15, line 16 to page 16, line 9 the TFE polymers may be homopolymers or may be modified with comonomers. Dependent on the desired processing characteristics of the TFE polymer, melt processable or not, the amount of the comonomer may be up to 35 wt.% or it may be only "minor" (e.g. up to 2 wt.%). In both cases suitable comonomers are selected from perfluoro alkenes or perfluoro(alkyl vinyl ethers). With regard to further details concerning the latter comonomer species US-A-3 132 123, i.e. document D4, is explicitly referred to. For the melt processable copolymers perfluoro (2-methylene-4-methyl-1,3-dioxolan) and highly fluorinated monomers having a single H-atom are mentioned as further comonomer alternatives.

4. *Problem and solution*

4.1 According to the opposed patent the alleged invention relates to aqueous TFE copolymer dispersions which are useful for fabricating non-tacky coatings having a smooth surface and an increased thickness without "mud-cracking"; particularly, the coating should have a thickness of at least 25  $\mu\text{m}$  and a surface roughness of less than 0,5  $\mu\text{m}$  (page 2, lines 5 to 7, 24 to 27, 31 to 35 and 56 to 58).

4.2 As set out in Section 3 above, D2 discloses already that by an increase of the particle size of a TFE polymer in its aqueous dispersion the thickness of a crack-free coating made therefrom can be increased. D2 also discloses that this effect applies not only to TFE homopolymers but also to TFE copolymers, including those having perfluoro(alkyl vinyl ether) units, particularly those having 3 to 10 carbon atoms (page 15, lines 12 to

13 and 1 to 3 from the bottom). Such units are within the scope of the comonomer formula of Claim 1 of the patent in suit which comprises fluorovinyl ethers having 3 to 19 carbon atoms.

4.3 Starting from D2 the problem underlying the patent in suit can thus be seen to be the provision of further aqueous dispersions of TFE/perfluoro(alkyl vinyl ether) copolymers having a particle size so increased as to allow the preparation of thick crack-free coatings which furthermore should exhibit an excellent surface smoothness.

4.4 From the evidence in the patent in suit it can be concluded that the above problem has been solved by the alleged invention. In particular, the "inventive" Examples 1 to 6 satisfy the criteria for crack-free thickness and surface roughness required according to the patent in suit, i.e. a minimum "Threshold cracking thickness" of 25  $\mu\text{m}$  and a "Surface roughness" of less than 0,5  $\mu\text{m}$  (see page 2, lines 56 to 58; page 4, lines 33 to 39; page 8, Table 2).

5. *Inventive step*

It remains to be decided whether the solution of the above mentioned technical problem by the subject-matter of Claim 1 involves an inventive step.

5.1 The dispersion according to present Claim 1 is distinguished from the disclosure in D2 by the following four features:

- (i) formula of the fluorovinyl ether comonomer,
- (ii) a comonomer content of 1 to 10 % by weight of fluorovinyl ether units,



(iii) a specific melt viscosity of the copolymer of  $0.3 \times 10^4$  to  $10.0 \times 10^4$  poise ( $0.03 \times 10^4$  to  $1 \times 10^4$  Pas) and

(iv) an average particle size of 0.3 to 1  $\mu\text{m}$ .

5.2 From the experimental evidence reported in the patent in suit the following conclusions can be drawn concerning the relevance of the above features (i) to (iv) with respect to the problem to be solved as defined in Section 4.3 above.

5.2.1 This evidence comprises the "inventive" Examples No. 1 to 6 and the Comparative Examples No. 1 to 5. All inventive Examples were prepared by the seed latex technology: according to Example 1 in a first step (1) TFE and PPVE were polymerized to an aqueous dispersion of their copolymer in the presence of methanol as chain transfer agent and of ammonium persulfate (APS) as initiator (page 4, lines 45 to 58). In a second step (2) the seed latex resulting from step (1) was reacted with further PPVE, again in the presence of methanol and APS; the aqueous dispersion obtained by step (2) was mixed with a nonionic surfactant as stabilizer (page 5, lines 15 to 28 and page 7, Table 1).

5.2.2 According to the Comparative Examples No. 1 to 3 the "Threshold cracking thickness" and the "Surface roughness" of TFE/PPVE copolymer dispersions which have been prepared using different amounts of methanol, but otherwise in the same way as the seed latex according to Example 1 (and thus without a second polymerization step), were tested. The values of the "Threshold cracking thickness" and of the "Surface roughness"

measured for Comparative Examples No. 1 and 2 are beyond the limits of 25  $\mu\text{m}$  and 0,5  $\mu\text{m}$ , respectively, required by the alleged invention. Comparative Example No. 3 is a borderline case.

However, the Appellant's contention that this failure of Comparative Examples 1 to 3 could be attributed to the fact that all these dispersions have particle sizes below the minimum of 0,3  $\mu\text{m}$  set by present Claim 1 and in one case (Comparative Example No. 2) also too high a melt viscosity is untenable. The different one-step technology employed according to these comparative examples prevents any conclusions as to such alleged interdependencies because there are factors which are likely to have much bigger impact on the measured properties. These factors comprise i.a. (a) the different cross-sectional copolymer structure of the particles resulting from the presence or not of a "graft" copolymerization step and (b) the different particle size distribution caused by the possible formation of new particles during the second step copolymerization. These two factors should have an important impact on the melt behaviour of the respective particles, influencing thereby both the stress situation within the coating (and thus the "Threshold cracking thickness") and the melt flow which is responsible for the surface structure of the coating, i.e. being more or less smooth.

5.2.3 According to Comparative Example No. 4 the two step seed technology was used to prepare a dispersion by subjecting the seed latex according to Comparative Example No. 2 to further copolymerization. Since there is no "inventive" Example corresponding to Comparative Example No. 4, a reliable conclusion is again not possible with regard to the influence of the too high specific melt viscosity of the copolymer particles (this

being the only cited copolymer property outside the ranges claimed in the opposed patent) of said Comparative Example on the "Threshold cracking thickness" and "Surface roughness" of the coatings prepared from these dispersions.

5.2.4 The only genuinely corresponding pair of "inventive" and "comparative" examples is represented by Example No. 1 and Comparative Example No. 5. Both these examples use the same seed latex and, with respect to the process features of the second polymerization step, differ only in the amount of methanol and in the reaction time, both being much higher according to the comparative example. The PPVE content of the copolymer particles prepared according to this comparative example is 2,4 wt.%, and thus within the range of 1-10 wt.% specified in Claim 1 of the patent in suit; the same holds true for the average particle size of the copolymer particles of 0,34  $\mu\text{m}$  which is within the range of 0,3 - 1  $\mu\text{m}$  specified in Claim 1. The only property differing from the dispersions according to present Claim 1 is a specific melt viscosity of  $0,2 \times 10^4$  poise which is slightly below the lower limit of  $0,3 \times 10^4$  according to said Claim 1. Significantly, the "Threshold cracking thickness" and the "Surface roughness" of the coatings prepared from the dispersions according to Comparative Example No. 5 are both excellent with respect to the desired limit values ("Threshold cracking thickness" of 40-45  $\mu\text{m}$  as compared with an "inventive" lower limit of 25  $\mu\text{m}$ ; "Surface roughness" of 0,30  $\mu\text{m}$  as compared with an "inventive" upper limit of 0,50  $\mu\text{m}$ : see page 8, Table 2).

It is, however, set out in footnote 2), attached to the value of the "Surface roughness" according to Comparative Example No. 5, that "mud cracking appeared on the coating after several days". This statement, in

its essence, is contradictory to the very high value of 40-45  $\mu\text{m}$  of the respective "Threshold cracking thickness" because the latter property is presented in the patent in suit as the only method for the evaluation of "mud cracking". The conditions under which the coating was stored, leading to mud cracking "after several days", are not specified; neither is there any information in the patent in suit concerning the quantitative aspects of this phenomenon (how many cracks, how deep, etc.) which would allow to appreciate the practical consequences of this phenomenon (cf. page 3, lines 9 to 14 of the patent in suit).

In these circumstances this footnote does not represent evidence sufficiently weighty to outbalance the very positive results of the measurements of the "Threshold cracking thickness" and the "Surface roughness" of the coatings prepared according to Comparative Example No. 5.

Moreover, the fact that the coatings according to Comparative Example No. 5 have good "Threshold cracking thickness" and "Surface roughness" is corroborated by the respective statement on page 6, lines 31 to 32 of the patent specification. However, it also stated there that the copolymer "had too poor coating strength to be practically used". This vague assertion lacks experimental proof and is not, therefore, a reliable basis for the acknowledgement of the alleged deficiency as a decisive phenomenon imputable to the slightly lower melt viscosity of the copolymer.

5.3 In view of the preceding analysis of the evidence contained in the patent in suit, it must be concluded that none of the features (i) to (iv) has been shown to have any, let alone any unexpected impact on the "Threshold cracking thickness" and the "Surface roughness" of coatings prepared from the dispersions according to Claim 1 of the patent in suit.

In consequence, the specific comonomer formula as well as the limits of ranges set by features (ii) to (iv) for the comonomer content, the specific melt viscosity and the average particle size cannot be recognized to have, with respect to the problem to be solved (see Section 4.3 above), any critical importance.

There is thus no factual basis for the Appellant's contention that the features (i) to (iv), separately or in combination, would be the result of a critical selection from the more general teaching in D2.

5.4 Rather, as explained below, the skilled person starting from the disclosure in D2 would have no difficulty in solving the relevant problem by the measures taken in Claim 1 of the patent in suit.

5.4.1 Since the choice as comonomer of a fluorovinyl ether of the formula according to present Claim 1 (feature (i)) has not been shown to have, with respect to the problem to be solved, any particular advantage over the other comonomers disclosed in D2, it must be qualified as a purely arbitrary selection with respect to the solution of said problem.

The could/would approach, i.e. whether the skilled person would (or only "could") have chosen these comonomers in the expectation of some improvement or advantage (T 2/83, OJ EPO 1984, 265) is therefore not applicable here, since all comonomers disclosed in D2 are equivalent with respect to the problem to be solved.

Moreover, there have been strong reasons for selecting comonomers having this formula as comonomers for TFE which are not related to the present problem but concern the very favourable compromise such copolymers offer by combining the excellent mechanical, physical and chemical properties afforded by TFE with the melt processability and the good thermal resistance provided by fluorovinyl ether comonomers (see D4: column 1, lines 26 to 49; Encyclopedia of Polymer Science and Technology, Suppl. volume 1, 260 to 266, cited by the Respondent).

- 5.4.2 Concerning feature (ii), i.e. the comonomer content of the copolymer being from 1 to 10 % by weight, this is within the broader range of up to 35 wt.% disclosed in D2, page 15 lines 6 to 14 from the bottom. There it is emphasized that the amount of comonomer determines the degree of melt processability of the copolymer. Too low amounts of comonomer are not sufficiently effective in this respect, too high amounts impair too much the mechanical, chemical and physical properties of the TFE backbone polymer (see also Encyclopedia of Polymer Science and Technology, Suppl. volume 1, 260 to 266). A skilled person being aware of this well known reciprocity and wishing to guarantee that the thick coatings disclosed in D2 have a smooth surface will have no difficulty to sort out by a few orientating experiments those amounts of ether comonomer, which will provide sufficient melt flow under the chosen baking conditions of the coating.

- 5.4.3 Feature (iii), i.e. the specific melt viscosity of the copolymer of  $0.3 \times 10^4$  to  $10.0 \times 10^4$  poise ( $0.03 \times 10^4$  to  $1 \times 10^4$  Pas), is related on the one hand to the amount of comonomer, i.e. to feature (ii), and on the other hand to the conditions of the copolymerization: reaction temperature as well as kind and amount of chain transfer agent and polymerization initiator (see patent in suit, page 3, lines 21 to 24).

Again a skilled person looking for a solution of the existing problem will have no difficulty to find out by routine experimentation which melt viscosities will be appropriate for achieving the desired melt viscosities that will provide a smooth surface of the coating and prevent a surface roughness caused by too little flow. Document D4, Example II (column 2, lines 55-72) demonstrates that TFE/PPVE copolymers meeting features (ii) and (iii) have been known to exist for many years, i.e. since 1964.

- 5.4.4 With regard to feature (iv), i.e. an average particle size of 0.3 to 1  $\mu\text{m}$ , this is fully in line with the disclosure of D2 (page 1, first paragraph; page 2, second paragraph; Claims 1 and 2) according to which a particle size having a mean diameter of at least 0,25  $\mu\text{m}$ , preferably from 0,32 - 0,45  $\mu\text{m}$ , is required to obtain thick, crack-free coatings. This teaching is clearly an incentive to the skilled person to investigate the influence of the particle size on the thickness of the coating to be prepared and to optimize the copolymer dispersion in this respect. Such experiments do not go beyond ordinary workshop activities.

5.5 The above considerations show that the claimed subject-matter merely represents a combination of obvious features whose concrete determination does not require more than routine experiments. This subject-matter does not, therefore, involve an inventive step in accordance with Article 56 EPC.

The same conclusion applies to the subject-matter of Claim 5 which is directed to an article coated with the dispersion according to Claim 1.

The dependent Claims 2 to 4 must share the fate of independent Claim 1.

6. In consequence the appeal must be dismissed.

**Order**

**For these reasons it is decided that:**

The appeal is dismissed.

The Registrar:

  
E. Görgmaier

The Chairman:

  
C. Gérardin