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 E C I S I O N
of 18 April 2000

Case Number: T 0785/96 - 3.3.3

Application Number: 86308129.5

Publication Number: 0220910

IPC: C08F 214/26

Language of the proceedings: EN

Title of invention:
Stable tetrafluorethylene copolymers

Patentee:
E.I. DU PONT DE NEMOURS AND COMPANY

Opponent:
Dyneon GmbH

Headword:
-

Relevant legal provisions:
EPC Art. 56

Keyword:
"Inventive step (yes) - problem solution - non-obvious combination of known features"

Decisions cited:
T 0606/89, T 0795/93, T 0097/94

Catchword:
-

Europäisches Patentamt | European Patent Office | Office européen des brevets

Beschwerdekammern | Boards of Appeal | Chambres de recours

Case Number: T 0785/96 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 18 April 2000

Appellant: (Opponent)
Dyneon GmbH
Werk Gendorf
Patentabteilung
D-84504 Burgkirchen (DE)

Representative:
Rauh, Peter A., Dr.
VOSSIUS & PARTNER
Postfach 86 07 67
D-81634 München (DE)

Respondent: (Proprietor of the patent)
E.I Du Pont De Nemours and Company
1007 Market Street
Wilmington
Delaware 19898 (US)

Representative:
Cresswell, Thomas, Anthony
J.A. KEMP & CO.
14 South Square
Gray's Inn
London WC1R 5LX (GB)

Decision under appeal: **Decision of the Opposition Division of the European Patent Office posted 2 July 1996 rejecting the opposition filed against European patent No. 0 220 910 pursuant to Article 102(2) EPC.**

Composition of the Board:

Chairman: C. Gérardin
Members: B. ter Laan
B. Schachenmann

Summary of Facts and Submissions

I. Mention of the grant of European patent No. 0 220 910 in respect of European patent application No. 86 308 129.5, filed on 20 October 1986, claiming priority from an earlier application in the US (789 893 of 21 October 1985), was published on 9 September 1992 (Bulletin 92/37) on the basis of five claims, Claim 1 reading:

"A melt-fabricable, nonelastomeric, tetrafluoroethylene copolymer substantially free of end groups that can react or decompose to emit HF, comprising recurring units of tetrafluoroethylene and at least one perfluoro(alkyl vinyl)ether where the alkyl group contains 1-8 carbon atoms and wherein the vinyl ether comprises between 1 and 10 weight percent of the copolymer; said copolymer characterized by having

(a) less than 6 endgroups of $-CF_2CH_2OH$, $-CONH_2$ and -COF per 10^6 carbon atoms and

(b) an extractable fluoride level of 3 ppm or less by weight."

Claims 2 to 4 referred to preferred embodiments of the copolymer according to Claim 1.

Independent Claim 5 was directed to:

"A process for producing a copolymer as claimed in any one of claims 1 to 4 which comprises treating a said copolymer containing more than 6 per 10^6 carbon atoms of end groups of $-CF_2CH_2OH$, $-CONH_2$ and -COF with a fluorine-

containing gas at a temperature, time and pressure sufficient to reduce the end groups $-CF_2CH_2OH$, $-CONH_2$ and -COF to less than 6 per 10^6 carbon atoms, and sparging the fluorinated copolymer with an inert gas until the sparge gas tests negative in the starch/iodide test."

II. On 8 June 1993 a Notice of Opposition against the granted patent was filed, in which the revocation of the patent in its entirety was requested on the grounds set out in Article 100(a) EPC.

The opposition was supported by the following documents:

D1: EP-A-0 150 953

D2: GB-A-1 210 794

D3: US-A-3 085 083

D4: US-A-3 642 742

III. By a decision issued in writing on 2 July 1996, the Opposition Division rejected the opposition. It was held that

(a) Novelty had not been contested and was acknowledged.

(b) Regarding inventive step, D4 was considered to be the closest document since it referred to the stabilisation of the same polymers as in the patent in suit, whereas D2 gave a more general disclosure of reducing the number of unstable end groups of fluorocarbon polymers. The problem solved by the patent in suit was to reduce the

corrosivity of tetrafluoroethylene/perfluoro(alkyl vinyl)ether copolymers towards materials in direct contact with it or in close proximity to it. None of the cited documents referred to the same problem as the patent in suit, nor was the low number of end groups as now required anywhere reported, so that those documents could not provide a solution to the above-defined problem. Hence the claimed subject-matter was inventive.

IV. On 29 August 1996 the Appellant (Opponent) lodged an appeal against the above decision. The Statement of Grounds of Appeal was filed and the prescribed fee was paid simultaneously. In a letter filed on 17 March 2000, further arguments were filed as well as an experimental report.

The Appellant, in writing and during the oral proceedings held on 18 April 2000, argued essentially as follows:

(a) Regarding the late filed experiments, their necessity had become clear only after a change of representative. They showed that there was a correlation between the specific value of fluoride content (feature (b) in Claim 1) of the patent in suit and the lack of colour in the iodine/starch test. Although D1 mentioned the test, no fluoride values were indicated. Since the experiments confirmed the findings of the patent in suit, they contained nothing new so that they should be admitted to the proceedings.

(b) As regards the problem allegedly solved by the patent in suit, the corrosivity problem, redefined

by the Opposition Division as being a post-fabrication problem, was not based upon the information contained in the patent specification. There was no disclosure of any post-fabrication problem as opposed to pre-fabrication problems.

(c) According to the patent specification, it had been found that the unstable end groups evolved HF; this was, however, common knowledge, as illustrated by D3 as well as D2, in which documents the removal of unstable end groups was described. Even if it had been found that the small amounts of unstable end groups remaining after the known treatments still caused problems, this did not constitute an invention. First, since in that case it was a problem-invention, to which, according to standard jurisprudence of the Boards, very strict standards should be applied. Secondly, because it was obvious to reduce the number of unstable end groups in order to arrive at a lower level of corrosion. Regarding the fluoride level, every commercial product must be purged since else the noxious gases made it impossible to handle it. The upper limit now required was known from D1. Therefore, if the product of D2 contained noxious gases, D1 taught how to remove them, so that the present combination of requirements was known from a combination of D1 and D2. In view of that, the problem as presented in the patent specification had already been solved in the prior art.

(d) The solution as defined in the claims was obvious. The closest document was not D4, but either of D1 or D2. Both documents taught to fluorinate tetrafluoroethylene copolymers in order to reduce

the number of unstable end groups. D2 specifically mentioned the copolymer treated in the patent in suit, but not the purging step after fluorination, whereas D1 disclosed the fluorination and purging of a similar copolymer. The now claimed copolymer was in fact the result of the direct application of the teaching of D1 to D2 so that it was obvious. The limits to the end groups and fluoride content as now required were arbitrary and could not serve to render the claimed subject-matter inventive.

V. The Respondent (Proprietor), in its written and oral submissions, argued essentially as follows.

(a) The late filed experiments, as well as the late arguments regarding the fluoride level, which was not discussed in the impugned decision, should not be admitted into the proceedings.

(b) The problem of the degradation of products after their formation or, in other words, the post-fabrication stability, was derivable from the original application. The problems addressed in the prior art concerned the fabrication process, as illustrated by the fact that in D2 also other groups than the present ones were considered to be unstable. Hence none of the cited documents addressed any post-fabrication problem.

(c) D4 was the closest document as it was filed more recently than D2 and it concerned the same type of polymer as the patent in suit. Although the polymers according to D4 as well as those described in D2 were considered adequate, they did

not meet the present requirements. There was no hint to reduce the unstable end groups any further in either of the documents. Also the polymers according to D2 had a much higher unstable end group level than now required, since at that time the detection level was higher. Therefore, the term "quantitative conversion" did not necessarily imply complete conversion. In view of the unpleasantness of fluorine radicals there was no reason to intensify the fluorination procedure of D2, which, on the contrary, emphasized the mild conditions of the fluorination. Therefore, whether D2 or D4 were considered to be the closest state of the art, no hint at the extremely reduced unstable end group content was given.

(d) As regards the fluoride content, the difference between purging, a quick process, and sparging, a slower process, was not mentioned in the prior art, so that the extremely low level of remaining fluoride as now required was not suggested by the prior art, in particular D1. Therefore, the claimed subject-matter was inventive.

VI. The Appellant requested that the decision under appeal be set aside and the patent revoked.

The Respondent requested that the appeal be dismissed.

Reasons for the Decision

1. The appeal is admissible.

Procedural matters

2. With a letter sent just one month before the oral proceedings before the Board, the Appellant filed additional experiments.

2.1 The Appellant stated that the reason for this late filing was a change of representative, after which the necessity of the experiments had appeared. Furthermore, the experiments merely confirmed the disclosure in the patent specification that there is a correlation between a specific value of residual fluoride content and the results (no colouration) of the I_2-test. Therefore, they should be admitted to the proceedings.

2.2 It is established case law by the boards of appeal that the change of representative does not form an acceptable ground for late filing. In the present case it was not shown that the change of representative at such a late stage of the proceedings was due to *force majeure*. On the contrary, the introductory statement in the Appellant's letter of 17 March 2000, one month before the oral proceedings, clearly demonstrates that this change was simply the wish of the client. The new representative was therefore obliged to continue the proceedings from the point they had reached when he took over from his predecessor (cf. T 97/94, OJ EPO 1998, 467, point 3.5.3).

As to the additional experiments, they only demonstrate the correctness of the statements or implications made in the patent in suit; as such they merely confirm information already available in the file and do not add anything of substance to the Appellant's case.

For both reasons the late filed experiments cannot be admitted into the proceedings.

The documents

3. The Opposition Division and the Respondent considered D4 as the closest prior art document. However, the Appellant gave arguments for using D2 as the starting point for assessing the inventive step. Therefore, a preliminary discussion of the documents on file is regarded as appropriate in order to decide upon the closest state of the art.

3.1 D1 concerns the removal of sources of volatiles from copolymers of tetrafluoroethylene with perfluorinated alpha olefins so as to avoid the formation of bubbles or voids in the final products (page 1, lines 8 to 15). The sources of volatiles are described to include unstable end groups and unstable backbone linkages, the former mostly being carboxylic acid end groups. Several other end groups after extrusion are identified (page 1, lines 16 to 28). The unstable backbone linkages appear to be an inverse function of molecular weight (and melt viscosity) (page 2, lines 5 to 14). D1 concerns the removal of the latter by a process which comprises subjecting the copolymer to a high shear rate for a time sufficient to reduce the backbone volatiles index of the copolymer (Claim 1).

Any unstable end groups can be removed by fluorination and D1 therefore refers to the conditions as described in D2. The reaction is preferably carried out with a fluorine/inert gas mixture (page 4, lines 20 to 28). Regarding -COF end groups, it is advised to prevent their formation by removing atmospheric oxygen or by injecting water into the extruder so as to hydrolyse them (page 5, lines 10 to 25). Reference is also made

to other methods for the removal of unstable end groups (page 6, lines 1 to 3). In the examples the polymers are fluorinated after the extrusion treatment by exposure to a fluorine/nitrogen mixture at elevated temperature, after which the fluorine is purged with nitrogen. The end group analyses show almost complete removal of all unstable end groups (page 11, line 7 to page 13, line 12), sufficient to permit bubble-free fabrication (page 13, lines 16 to 20). The purging is carried out until the nitrogen purge was free of fluorine, which was checked by means of potassium iodide (page 13, lines 4 to 9).

3.2 D2 describes a relatively mild and short process for chemically stabilising a solid high molecular weight fluorocarbon polymer which contains chemically unstable end groups which comprises contacting the solid polymer in the absence of oxygen, with a source of fluorine radicals under conditions at which said source generates fluorine radicals, whereby at least 40% of the chemically unstable end groups are converted to chemically stable end groups (Claim 1 in conjunction with page 1, lines 44 to 49). By fluorocarbon polymer a polymer is meant which is either perfluorinated or highly fluorinated (page 1, lines 62 to 67), such as polymers derived from tetrafluoroethylene and its copolymers (page 2, lines 72 to 109). The chemically unstable end groups are described to include carboxylate and vinyl end groups as well as other end groups which are convertible to a more stable form, like e.g. $-CF_2H$ and amide groups (page 1, lines 76 to 82). Preferably, the conversion is quantitative, but a lesser degree of stabilization, e.g. at least 40% or at least 75%, may be acceptable (page 4, lines 33 to 41). In the examples tetrafluoroethylene copolymers are fluorinated by charging the reactor with fluorine gas and heating under autogenous pressure. After cooling, the gas is vented. The amount of unstable end groups is greatly reduced, partly even below the detection limit, which is described as 5 end groups of carboxylate, monomer or dimer, and 10 vinyl end groups. The thus treated copolymers have improved stability (page 5, lines 8 to 27; Table I).

The problem solved by the stabilisation of the unstable end groups is the prevention of long term HF formation in fuel cells caused by degradation of the membrane by hydroxyl radicals (page 3, lines 38 to 51), or in any

usual application where stability is a problem (page 4, lines 87 to 91).

3.3 According to D3, gas bubbles may form in the extruded product due to volatile products formed during polymerization of the polymer, much of which can be removed through a finishing operation. However, some of the volatile gas results from continuous decomposition of the polymer end groups, which cannot be removed by finishing (column 1, lines 51 to 59). The main cause of that instability is the presence of carboxylate end groups in the polymer chain (column 2, lines 24 to 59). D3 proposes to improve the thermal stability of a solid, cold-drawable copolymer of tetrafluoroethylene and a fluoroolefin having the general formula CF_2=CFY where Y is a perfluoroalkyl radical having from 1 to 8 carbon atoms, said copolymer having a molecular weight in excess of 10,000 and having at least half of the molecular end-groups in the form of carboxylate end groups, by a process which comprises contacting said copolymer in a finely divided, loose unsintered form with water, the concentration of said water being at least 2% by weight of the copolymer environment, at a temperature of 200 to 400°C, for a period sufficient to remove substantially all of the carboxylate end groups and recovering a fluorocarbon copolymer in which at least half of the molecular end groups, as measured by infrared analysis, have the structure -CF_2H (Claim 6). The reaction with water is enhanced by the addition of bases, neutral or basic salts to either the aqueous phase or the polymer (column 2, line 61 to column 3, line 8; Claims 7 to 12 and 14). In the examples various copolymers are treated with aqueous basic solutions, which results in greatly reducing the amount of carboxylate end groups and the occurrence of -CF_2H end-

groups.

3.4 D4 describes a process for forming a polymer of tetrafluoroethylene monomer and at least one fluorovinylether monomer copolymerizable therewith which comprises polymerizing tetrafluoroethylene with a specified comonomer in the presence of a halogenated solvent and a hydrogen-containing chain transfer agent selected from the groups consisting of methanol, isopropanol and ethanol thereby to provide a copolymer having stable end groups (Claim 1). It is said that an important cause of instability in fluorocarbon polymers is the presence of acid fluoride end groups which are readily converted to carboxylic acid end groups and which are formed by rearrangement of the fluorovinyl ether on the end of the growing polymer chain. During storage, these end groups are hydrolysed and decompose during extrusion forming gases which show up as bubbles in extruded products (column 1, lines 33 to 49). With increasing molecular weight, the number of unstable end groups decreases, but also the ease of fabricability of the polymer decreases to a large extent (column 2, line 68 to column 3, line 3). D4 proposes to add a hydrogen containing chain transfer agent to the polymerization recipe, in order to reduce the number of chain terminations made by rearrangement; that way, stable hydride end groups ($-CF_2H$) are formed (column 3, lines 3 to 15). In the examples the number of unstable end groups varies from 29 (Example VI) to 109 (Example I) per 10^6 carbon atoms.

Novelty

4. Novelty has been recognised by the Opposition Division and the parties did not contest that part of the

decision. In the light of the disclosure of the documents on file (see point 3 above), the Board also comes to the conclusion that the claimed subject-matter is novel.

Problem and solution

5. The patent in suit concerns stable tetrafluoroethylene copolymers. Such copolymers are described in all of D1 to D4. The Opposition Division and the Respondent considered D4 to be the closest prior art document since it referred to the same type of copolymer as the patent in suit and it was the most recent disclosure, whereas the Appellant used D2 as the starting point for the definition of the problem to be solved.

5.1 According to the established jurisprudence of the boards of appeal, generally, the claimed invention should be compared with the art concerned with a similar use which requires the minimum of structural and functional modifications. This involves not only comparing the claimed compositions with those of the prior art, but also giving consideration to the particular properties which render the compositions suitable for the desired use. Therefore, a document serving as the starting point for evaluating the inventive merits of an invention should relate to the same or a similar technical problem or, at least, to the same or a closely related technical field as the application in suit (see decisions T 606/89 of 18 September 1990 and T 795/93 of 29 October 1996; both unpublished in OJ EPO).

According to the patent specification, the end groups - CF_2CH_2OH, $-CONH_2$ and -COF generate HF, which is corrosive

to metals, leading to the formation of metal fluoride corrosion products (page 2, lines 37 to 40 and page 3, lines 24 to 27). Therefore, the problem to be solved as arising from the patent specification is to provide a copolymer having low corrosivity toward metals (page 4, line 21).

5.2 From the analysis of the documents on file, it is clear that none of them mentions that problem. D1, D3 and D4 concern the problem of bubble formation, whereas D2 refers to the long term formation of HF in fuel cells, indicative of the degradation of the membrane under the influence of hydroxyl radicals. However, being the only document that specifically mentions the generation of HF, which is mentioned as the cause of the corrosion problems to which the patent in suit refers, in the Board's view it qualifies as a proper starting point for the evaluation of the inventive merits of the claimed subject-matter.

5.3 Although, as elucidated above (point 3.2) the compositions of D2 may have a low content of undesirable end groups and are said to be suitable for applications where stability is a problem, their corrosivity toward metal was still capable of improvement. In other words, the number of undesirable end groups could not be regarded as optimally adapted to applications requiring contact with metals.

5.4 Therefore, the technical problem to be solved by the patent in suit can be defined as to improve the metal corrosivity of tetrafluoroethylene copolymers; more specifically, to further reduce the number of undesirable end groups at an extremely low level of extractable fluoride.

5.5 The examples in the application demonstrate that that problem is effectively solved. In particular, from Examples 1 to 3 it appears that the present compositions have an extremely low content of both undesirable end-groups and extractable fluoride.

Obviousness

6. It remains to be decided whether the claimed subject-matter is obvious having regard to the documents on file.

6.1 In D2 the problem of the HF generation is solved by contacting the solid polymer in the absence of oxygen, with a source of fluorine radicals under conditions at which said source generates fluorine radicals, whereby at least 40% of the chemically unstable end groups are converted to chemically stable end groups. There is no suggestion that the amount of unstable end groups and the level of extractable fluoride should be as low as now required by present Claim 1. Although quantitative and 100% conversion are mentioned, in view of the relatively high detection limit, this cannot be interpreted as disclosing the range of unstable end groups now required. In fact, the relatively mild and short fluorination conditions (varying from 50°C to 190°C during 2 hours in the examples of D2 as compared to 200°C during 8 hours, 200°C during 16 hours and 210°C during 6 hours in the present examples) would not be sufficient to produce polymers coming within that range. Also, even if venting of the fluorine gas after fluorination is mentioned, the very low amount of residual extractable fluoride is not mentioned. Therefore, D2 by itself cannot render the present combination of features obvious.

6.2 The same is valid for the other documents on file: none of them refers to the extremely low end group content of the polymer. D3, disclosing end group conversion by means of a reaction with water, does not suggest any fluorination method and even less to intensify the method of D2. In D4 the level of unstable end groups is reduced during polymerization, so that it does not teach any method of reducing their number after polymerization. Only D1 mentions fluorination of the polymer, referring to D2. Although in the examples complete removal of unstable end groups is reported, like in D2, in view of the detection limits this cannot be interpreted in a way that the number of unstable end groups would fall within the range now required. Like D3 and D4, D1 does not contain any suggestion to intensify the fluorination conditions of D2. Therefore, even if the residual fluorine test is negative in D1 and this would mean that the extractable fluoride level falls within the range now required, and if the purging step described in D1 would be applied to the fluorination process of D2, that combination would not lead to the claimed subject-matter. In this light, a possible difference between the terms "sparging" and "purging", as argued by the Respondent, plays no role.

6.3 For the above reasons, the Board comes to the conclusion that the subject-matter of Claim 1 involves an inventive step.

7. As Claim 1 of the main request is allowable, the same is valid for dependent Claims 2 to 4, the patentability of which is supported by that of Claim 1. The above considerations also apply to independent Claim 5 since its subject-matter is based on the same combination of features as in Claim 1.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar: The Chairman:

E. Görgmaier C. Gérardin