BESCHWERDEKAMMERN DES EUROPÄISCHEN PATENTAMTS

BOARDS OF APPEAL OF THE EUROPEAN PATENT OFFICE

CHAMBRES DE RECOURS DE L'OFFICE EUROPEEN DES BREVETS

A		B		C	X

File Number: T 494/92 - 3.3.3

Application No.: 87 100 189.7

Publication No.: 0 232 716

Title of invention: Polyphenylene ethers of improved stability

Classification: C08G 65/48

D E C I S I O N
of 13 June 1993

Applicant: General Electric Company

Headword:

EPC Article 56

Keyword: "Inventive step (yes) - adequate technical evidence provided"

Europäisches Patentamt | European Patent Office | Office européen des brevets

Beschwerdekammern | Boards of Appeal | Chambres de recours

Case Number : T 494/92 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 13 June 1993

Appellant : General Electric Company
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Decision under appeal : **Decision of the Examining Division of the European Patent Office dated 20 January 1992 refusing European patent application No. 87 100 189.7 pursuant to Article 97(1) EPC.**

Composition of the Board :

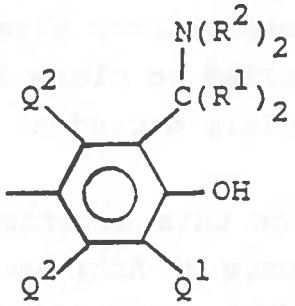
Chairman : F. Antony
Members : C. Gérardin
S. Perryman

Summary of Facts and Submissions

I. European patent application No. 87 100 189.7 filed on 9 January 1987, claiming the priority of 14 January 1986 from an earlier application in the United States and published on 19 August 1987 under the publication number 232 716, was refused by a decision of the Examining Division dated 20 January 1992.

This decision was based on a set of 12 claims filed on 12 June 1991, of which Claim 1 read as follows:

"A method for converting a polyphenylene ether having amino-alkyl-substituted end groups of the formula



wherein,
Q^1 is halogen, primary or secondary alkyl containing up to 7 carbon atoms, phenyl, haloalkyl, aminoalkyl, hydrocarbonoxy or halohydrocarbonoxy wherein at least two carbon atoms separate the halogen and oxygen atoms, each Q^2 is independently hydrogen or as defined for Q^1, each R^1 is independently hydrogen or alkyl, with the proviso that the total number of carbon atoms in both R^1 radicals is 6 or less, and each R^2 is independently hydrogen or a C_{1-6} primary alkyl radical, to derivatives of increased stability which comprises reacting said polyphenylene ether, at a temperature within the range of about 200-300°C, with at least one nitrogen-containing compound wherein at least one NH moiety forms

part of an amide, imide, amidine, 2- aminocarboxylic acid or sulfonamide group, in the absence of materials which catalyze polymerization of said nitrogen-containing compound."

Minor clerical amendments have been made by the Board, in particular change of Q' and R' into Q^1 and R^1 respectively for consistency reasons, addition of a comma before "to derivatives" and a hyphen in the first expression " nitrogen-containing".

Claims 2 to 7 were dependent process claims directed to preferred embodiments of the method according to Claim 1. Claim 8 was an independent product claim concerning a composition comprising polyphenylene ethers obtainable by the process according to Claim 1. Claims 9 to 11 were dependent product claims related to particular compositions according to Claim 8; the same obviously applied to Claim 12, inappropriately drafted as a method claim dependent on Claim 11.

II. The ground for this decision was non-compliance with the requirements of Article 56 EPC with regard to the teaching of US-A-4 092 294 (document (2)). More specifically, it was stated in this decision that novelty was not under dispute, for document (2) described only polyphenylene ethers containing aminoalkyl end groups, not the subsequent modification thereof with a nitrogen-containing compound; however, in the absence of appropriate experimental evidence demonstrating advantageous properties, this reaction could not be related to the solution of a technical problem. In particular, the comparative examples filed by the Applicant which showed that the stability of 2-aminomethyl-substituted phenol was improved by reaction with caprolactam, were inadequate since they concerned a low molecular weight compound; no arguments had been

submitted that conclusions drawn on the basis of such compounds would hold also for polymers.

III. The Appellant (Applicant) thereafter lodged a Notice of Appeal against this decision on 13 March 1992 and paid the prescribed fee at the same time. Together with the Statement of Grounds of Appeal filed on 15 May 1992 the Appellant submitted a new set of 12 claims and a test report carried out with polymers according to the application in suit.

(i) The new set of claims was practically identical with the claims filed on 12 June 1991. The only amendments concerned Claim 1, which had been modified as mentioned above in point I and wherein additionally the word "about" before "200-300°C" had been deleted, and Claim 12, which was now appropriately drafted as a composition claim.

(ii) In the test report various modified polyphenylene ethers had been analysed by carbon-13 nuclear magnetic resonance. These spectra showed in particular that a composition within the meaning of the application in suit was thermally stable, whereas an aminoalkyl terminated polyphenylene ether after heating at temperatures over 270°C in the melt by itself contained an undesirable complex mixture of end groups and coupling sites.

(iii) Moreover, when a caprolactam reaction product was heated in the melt with an equal amount of polyamide 66, a graft copolymer was obtained which had been found essential in the preparation of compatible polyphenylene ether-polyamide blends. By contrast, no grafting

occurred when a regular aminoalkyl terminated polyphenylene ether was used.

(iv) Following an objection raised by the Board regarding the incomplete definition of R^5 in Claims 4 and 10 as well as in the description, the Appellant amended these claims and page 4 accordingly on 8 June 1993.

IV. The Appellant requested that the decision under appeal be set aside, a patent be granted on the basis of the claims filed on 15 May 1992 as amended on 8 June 1993, and oral proceedings be arranged.

Reasons for the Decision

1. The appeal complies with Articles 106 to 108 and Rule 64 EPC and is admissible.

2. At the end of the Statement of Grounds of Appeal (point 3.(e)) the Appellant made an apparently unconditional request for oral proceedings. In view of the positive conclusion reached by the Board regarding the question of inventive step, as will appear hereinafter, and in the absence of any other substantive and/or procedural issues, such oral proceedings would serve no purpose. The Board thus treats this request as merely conditional, and not intended to prevent an immediate decision in favour of the Appellant.

3. The wording of the claims does not give rise to any objections under Article 123(2) EPC.

Claim 1 differs from Claim 1 as originally filed by the incorporation of the formula of the aminoalkyl end

groups and by the requirement that the reaction is carried out in the absence of materials which catalyse the polymerisation of the nitrogen-containing compound. This formula is disclosed in original Claim 12 and on page 3, lines 8 to 24 in conjunction with page 2, lines 18 to 22 of the original application as published. The condition that the reaction mixture should be free from materials which may have a catalytic action on the inactivating agent is supported by the original description on page 5, lines 6 to 9, as published.

The dependent method Claims 2 to 7 are identical with the original version of these claims. The independent composition Claim 8 corresponds to original Claim 12. Similarly, the dependent composition Claims 9 and 10 correspond to Claims 13 and 14 as originally filed with their numbers and appendancies adjusted. As to Claims 11 and 12, they recite in the framework of further dependent composition claims all the features which were mentioned in original Claims 15 and 16 inappropriately drafted as method claims.

As far as the last amendments in Claims 4 and 10 are concerned, namely the deletion of the radical NR^5 in the definition of Z^1 and the possibility for Z^1 and R^4 taken together in formula (VI) for form a cyclic amidine, they are not objectionable, since the former results in a restricted definition of Z^1 and the latter is supported by the original description on page 7, lines 22 to 28, as published.

4. The application in suit concerns polyphenylene ethers of improved stability and a method for preparing them. Modified polyphenylene ethers are disclosed in document (2) which the Board, like the Examining Division, regards as a suitable starting point for the definition of the technical problem underlying the application in

suit. This citation describes a process for the preparation of a polyphenylene ether resin by oxidatively coupling a 2,6- disubstituted phenolic compound in the presence of a catalyst system which comprises (i) a copper compound, (ii) an aliphatic or cycloaliphatic secondary diamine, (iii) a tertiary amine, (iv) a bromine compound selected among specific organic and inorganic compounds, and (v) a secondary monoamine (Claim 1). According to a preferred embodiment di-n-butylamine is used as component (v) of the catalyst system (column 2, line 66 to column 3, line 3; column 6, lines 26 to 36), which corresponds to the method of preparation of the polyphenylene ethers containing aminoalkyl end groups mentioned in the application in suit (compare page 3, lines 25 to 30, as published). Although these functional groups are beneficial in terms of impact strength and compatibilization of polyphenylene ethers with other blend components, at the same time they increase the reactivity of these polymers, whereby their stability at high temperatures is impaired.

In the light of this prior art shortcoming the technical problem underlying the application in suit may thus be seen as the provision of further modified polyphenylene ethers of improved stability, particularly at high temperatures.

According to Claim 1 this problem is solved by reacting the polyphenylene ethers containing aminoalkyl end groups with at least one nitrogen-containing compound, wherein at least one NH moiety forms part of an amide, imide, amidine, 2- aminocarboxylic acid or sulfonamide group.

The experimental results submitted together with the Statement of Grounds of Appeal demonstrate that such

modification provides an effective solution to the above-defined technical problem. More specifically, the comparison of the carbon-13 nuclear magnetic resonance spectra of aminoalkyl terminated polyphenylene ethers and of the same polymers modified with caprolactam shows that only the latter maintains its chemical structure when heated over 270°C (cf. Exhibit B, spectra 1 and 2).

5. After examination of document (2) the Board has come to the conclusion that this technical teaching is not disclosed therein and that the subject-matter of the application in suit as defined in present Claim 1 is, therefore, novel. Since the issue of novelty has not been raised in the decision under appeal, it is not necessary to consider this matter in further detail.

6. It still remains to be decided whether this subject-matter involves an inventive step having regard to the teaching of document (2).

6.1 The catalyst system used in this citation for the preparation of polyphenylene ether by oxidative coupling of a 2,6- disubstituted phenolic compound can be regarded as a conventional system in which a minor amount of a secondary monoamine (v) has been incorporated. This inclusion serves a dual purpose.

In the first place, it overcomes the shortcomings in terms of impact strength and thermal oxidative stability of blends of polyphenylene ether and styrene resins, such as rubber modified polystyrene. In the second place, it aids in the phase separation of the polymer containing phase after addition of an aqueous solution containing the chelating agents as compared to a reaction mixture from a polymerisation carried out in the absence of a secondary monoamine (column 1, line 36 to column 2, line 41). The reactivity of the resulting

polymer, which corresponds to the aminoalkyl terminated polyphenylene ether to be stabilised in the application in suit, is not discussed in the citation, and a subsequent chemical modification of the polymer is not envisaged.

There is thus no incentive for the skilled person to consider a reaction of this polymer with a specific nitrogen-containing compound for inactivating purposes; it follows that the claimed subject-matter cannot be regarded as obvious to a person skilled in the art, and, so involves an inventive step.

6.2 Additionally, the resulting aminoalkyl terminated polyphenylene ether-caprolactam reaction product shows a distinct advantage over the non-modified polymer when used for reaction with polyamides.

When a reaction product within the terms of the application in suit is heated in the melt with an equal weight amount of polyamide 66 and the blend is subsequently extracted with chloroform to remove the polyphenylene ether homopolymer, 19 percent by weight of the polyphenylene ether remains bound to the polyamide as a graft copolymer; by contrast, when a regular aminoalkyl terminated polyphenylene ether is used, no polyphenylene ether is found to be bound to the polyamide. This means that by modifying an aminoalkyl terminated polyphenylene ether according to the method of the application in suit, blends of polyphenylene ether and polyamide can be made compatible (Statement of Grounds of Appeal, point (c)). In the Board's view, this unexpected property of caprolactam modified polymers is a further evidence of inventive step.

6.3 As noted above, the test report submitted by the Appellant demonstrates that the problem of improved

stability of aminoalkyl terminated polyphenylene ether at high temperatures is effectively solved by subjecting the polymer to a reaction with one of the nitrogen-containing compounds specified in Claim 1. This technical evidence clearly overcomes the lack of appropriate experimental data objected to by the Examining Division in its communication of 19 July 1991 (cf. point 2) as well as in the decision of refusal (cf. point 7), i.e. the use of a monomer for the reaction with caprolactam instead of a polymer. The Examining Division therefore could and, in the Board's view, should have rectified its decision in accordance with Article 109(1) EPC.

6.4 Quite apart from this, it is difficult to see how the decision under appeal could come to the conclusion that there was no evidence that the distinguishing feature over document (2) which it acknowledged did not lead to the solution of a technical problem. If indeed the comparative examples showing an increased stability of monomeric phenols modified with caprolactam were to be considered "irrelevant" (a finding the correctness of which it is not necessary for the Board to consider), there would still have been an identifiable technical problem solved by the claimed proposal, *viz.* the provision of another (as opposed to a more stable) polymer product and its method of preparation, the inventiveness of which would have had to be examined. The mere absence of an unexpected advantageous effect *per se* is insufficient for a finding of obviousness, "technical advance" being no separate requirement of the EPC.

6.5 Since, the Examining Division took the view that the other documents cited in the search report did not provide an incentive for the skilled person along the lines defined in the main method claim (communication of

14 March 1991, point 2, last paragraph), an opinion with which the Board sees no reason to disagree, all the requirements to grant a patent on the basis of the claims on file would appear to be met.

7. Although the application is patentable in principle, a patent cannot yet be granted in the absence of a description adapted to the allowable claims.

The case must thus be remitted to the Examining Division.

Order

For these reasons, it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the Examining Division with the order to grant a patent on the basis of Claims 1 to 12 filed on 15 May 1992, as amended on 8 June 1993, and a description yet to be adapted.

The Registrar: The Chairman:

E. Görgmaier F. Antony