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D E C I S I O N
of 27 May 1997

Case Number: T 0246/93 - 3.3.3

Application Number: 88103175.1

Publication Number: 0284803

IPC: C08G 77/42

Language of the proceedings: EN

Title of invention:

Novel soluble polyimidesiloxanes and methods for their
preparation and use

Applicant:

Occidental Chemical Corporation

Opponent:

-

Headword:

-

Relevant legal provisions:

EPC Art. 84, 123(2)

Keyword:

-

Decisions cited:

-

Catchword:

"Amendments - added subject-matter (no)"
"Claims - clarity (yes, after amendment)"



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Boards of Appeal

Chambres de recours

Case Number: T 0246/93 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 27 May 1997

Appellant:

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Decision under appeal:

Decision of the Examining Division of the
European Patent Office posted 5 November 1992
refusing European patent application
No. 88 103 175.1 pursuant to Article 97(1) EPC.

Composition of the Board:

Chairman: C. Gérardin
Members: B. ter Laan
A. C. G. Linqvist

Summary of Facts and Submissions

- I. European patent application No. 88 103 175.1, filed on 2 March 1988, claiming priority from an earlier application in the USA (32272 of 31 March 1987), and published on 5 October 1988 under publication No. 0 284 803, was refused by a decision of the Examining Division of the European Patent Office dated 5 November 1992.
- II. That decision was based upon a set of 47 claims filed on 24 August 1992, Claim 1 reading as follows:

"A fully imidized polyimidesiloxane soluble in diglyme for use in the microelectronics industry, which comprises the reaction product of an organic dianhydride, an organic diamine and a difunctional siloxane monomer, wherein at least 80 weight percent of said organic dianhydride is an oxydiphthalic anhydride and wherein at least 75 weight percent of said organic diamine is an asymmetrical diamine that provides an asymmetrical structure in the polyimidesiloxane polymer chain."

Dependent Claims 2 to 25 referred to preferred embodiments of the polyimidesiloxane polymer according to Claim 1.

Independent Claim 26 was directed to:

"A process for producing a polyimidesiloxane which comprises reacting an oxydiphthalic anhydride, a difunctional siloxane monomer, and an organic diamine, wherein at least 80 weight percent of said organic

dianhydride is an oxydiphthalic anhydride and wherein at least 75 weight percent of said organic diamine is an asymmetrical diamine that provides an asymmetrical structure in the polyimidesiloxane polymer chain."

Dependent Claims 27 to 41 referred to preferred embodiments of the process according to Claim 26.

Claims 42 and 43 referred to solutions comprising the polyimidesiloxane polymer of Claim 1, Claims 44 to 46 were directed to substrates coated with or articles prepared from the polymer of Claim 7, and Claim 47 referred to cured compositions of Claims 5, 17 and 24.

The Examining Division held that the claimed subject-matter was neither clear nor novel. In particular, it was found that

- (a) the expression "asymmetrical diamine" (which was not directly and unambiguously disclosed in the original application), in the light of the examples given in the description, did not indicate which kind of asymmetrical structure was to be provided in the polyimidesiloxane and
- (b) the novelty objection was based upon the opinion that the unclear expression could not serve to establish novelty.

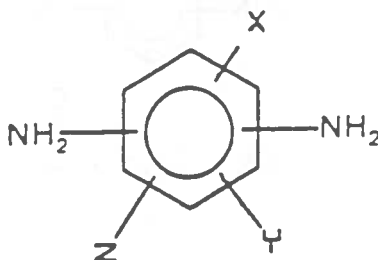
Hence the requirements of Articles 84 and 54 EPC were not fulfilled.

III. On 4 January 1993 a Notice of Appeal was lodged against that decision, together with payment of the prescribed fee. With the Statement of Grounds of Appeal filed on 3 March 1993 the Appellant (Applicant) filed three sets of claims as main, first and second auxiliary requests respectively.

After a communication from the Board, in which several objections were raised against the three sets of claims then on file, on 25 April 1997 three new sets of claims were filed replacing the claims that were filed with the Statement of Grounds of Appeal. Those new claims were withdrawn during the oral proceedings before the Board, to be replaced by another set of 30 Claims as the sole request, of which Claim 1 reads as follows:

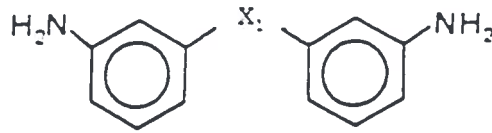
"1. A fully imidized polyimidesiloxane soluble in diglyme for use in the microelectronics industry, which comprises the reaction product of an organic dianhydride, an organic diamine and a difunctional siloxane monomer, wherein

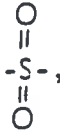
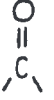
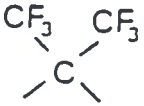
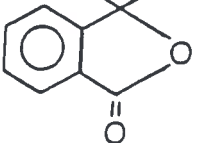
- (a) said organic dianhydride is an oxydiphthalic anhydride containing up to 20 weight percent of other organic dianhydrides,
- (b) said organic diamine is a diamine providing an asymmetrical structure in the polyimidesiloxane chain selected from
 - (b1) a diamine having the following formula



where X, Y and Z are independently selected from hydrogen, halogen, alkyl or halogenated alkyl of 1 to 12 carbon atoms or aryl or halogenated aryl of 6 to 12 carbon atoms,

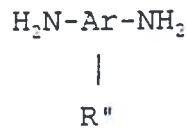
(b2) a diamine having the following formula



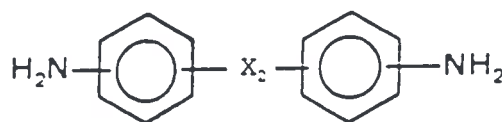
wherein X₁ is -CH₂-, , O, ,  or 


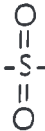
(b3) diaminoanthraquinone and

(b4) a diamine having the following formula:



wherein Ar is aromatic and R'' is a hydroxyl, hydrothiol or carboxylic acid radical, and may contain up to 25 weight percent of an organic diamine having the following formula that is not asymmetrical in the polymer chain



wherein X₂ is -CH₂-, , -O-, -S- or 

and

(c) said difunctional siloxane monomer is an α , ω -siloxane diamine."

Dependent Claims 2 to 15 refer to preferred embodiments of the polyimidesiloxane polymer according to Claim 1.

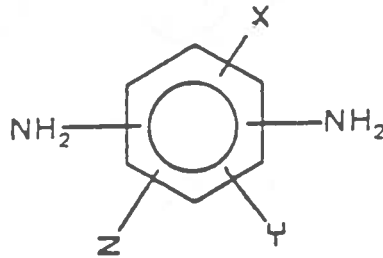
Independent Claim 16 is directed to:

"16. A process for producing a fully imidized polyimidesiloxane soluble in diglyme for use in the microelectronics industry, which comprises reacting an organic dianhydride, an organic diamine and a difunctional siloxane monomer, wherein

(a) said organic dianhydride is an oxydiphthalic anhydride containing up to 20 weight percent of other organic dianhydrides,

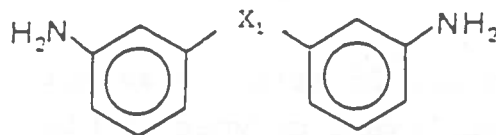
(b) said organic diamine is a diamine providing an asymmetric structure in the polyimidesiloxane chain selected from

(b1) a diamine having the following formula



where X, Y and Z are independently selected from hydrogen, halogen, alkyl or halogenated alkyl of 1 to 12 carbon atoms or aryl or halogenated aryl of 6 to 12 carbon atoms,

(b2) a diamine having the following formula

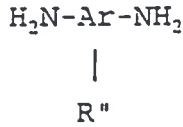


wherein X₁ is -CH₂-, , O, , , or

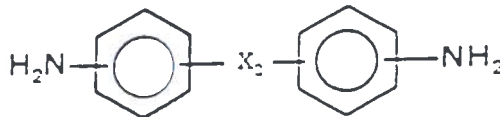
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(b3) diaminoanthraquinone and

(b4) a diamine having the following formula:



wherein Ar is aromatic and R'' is a hydroxyl, hydrothiol or carboxylic acid radical, and may contain up to 25 weight percent of an organic diamine having the following formula that is not asymmetrical in the polymer chain



wherein X₂ is $-\text{CH}_2-$, $-\overset{\text{O}}{\parallel}{\text{C}}-$, $-\text{O}-$, $-\text{S}-$ or $-\overset{\text{O}}{\parallel}{\text{S}}-\overset{\text{O}}{\parallel}-$

and

(c) said difunctional siloxane monomer is an α , ω -siloxane diamine."

Dependent Claims 17 to 24 refer to preferred embodiments of the process according to Claim 16.

Claims 25 and 26 refer to solutions comprising the polyimidesiloxane polymer of Claim 1, Claims 27 to 29 are directed to products containing the polymer according to Claim 1 and Claim 30 refer to cured compositions of Claims 5 or 15.

The Appellant argued that all amendments were based upon the description as originally filed, that the expression "asymmetrical diamine" objected to by the Examining Division had been deleted and that the expression "providing an asymmetrical structure in the polymer chain" clearly meant asymmetrical with respect to a line drawn between the nitrogen atoms, as otherwise it would not provide the polymer chain with the bended structure caused by that asymmetry, which was responsible for an improvement of the solubility without lowering the glass transition temperature. Therefore, the requirements of Articles 123(2) and 84 EPC were fulfilled.

- IV. The Appellant requested that the decision under appeal be set aside and a patent be granted on the basis of Claims 1 to 30 filed on 27 May 1997.

Reasons for the Decision

1. The appeal is admissible.

Article 123(2) EPC

2. The wording of the claims does not give rise to any objections under Article 123(2) EPC for the following reasons.
- 2.1 Product Claim 1 is based upon Claims 1, 9 and 25 as originally filed. The amount of organic anhydrides other than oxydiphthalic anhydride can be found on page 7, second full paragraph, and the amount of organic diamine that is not asymmetrical in the polymer chain on page 11, lines 1 to 2 of the original description. The definitions of the diamines (b1) to (b4) providing the polymer chain with an asymmetrical

structure are based upon the original description, page 9, last paragraph to page 10, last paragraph but one, page 20, third paragraph and original Claim 22. The use of α,ω -siloxane diamines is disclosed on original page 12, full paragraph 2.

Claim 2 is based upon original Claims 2 to 4. The basis for Claim 3 can be found in a combination of original Claim 5 and the implication of page 20, lines 9 to 11 of the original description. Claim 4 is supported by original Claim 6. Claim 5 is based upon original claim 7 and the implication of original page 20, lines 9 to 11. The basis for Claim 6 can be found in original Claim 8. Claim 7 is supported by original Claims 10 to 13 and Claim 8 by original Claims 14 to 21 with a correction of an obvious error in original Claim 17 (2,4,6-trimethyl-1,3-aminobenzene instead of 2,4,6-trimethyl-2,3-aminobenzene). Claim 9 is based upon original Claim 26, Claim 10 upon original Claim 27, and Claims 11 to 15 are supported by original Claims 29 and 33.

2.2 Process Claim 16 refers to the process as disclosed in original Claim 43 and is amended in conformity with present Claim 1. Claims 17 and 18 are based upon original Claims 44 and 45. Claim 19 finds its basis in original Claims 46 and 26. Claim 20 is supported by original Claims 47 and 27, Claim 21 by original Claims 54 and 29, Claim 22 by original Claims 55 and 30, Claim 23 by original Claims 56 and 31, and Claim 24 by original Claims 60 and 32.

2.3 The basis for Claims 25 and 26 can be found in original Claims 63 and 64, respectively. Claims 27 and 28 are based upon original Claims 65 and 66, respectively. Claim 29 is supported by original Claims 67 to 70 and Claim 30 by original Claims 71, 72 and 33.

Clarity

3. The Board is satisfied that the claimed subject-matter complies with the requirements of Article 84 EPC for the following reasons.
- 3.1 The Examining Division objected to the use of the word "asymmetrical" before "diamine" in the independent claims, which was not only not disclosed in the application as originally filed but also caused unclarity regarding its meaning. That word has been deleted, so that the asymmetry required according to the present claims now clearly refers to "an asymmetrical structure in the polyimidesiloxane chain". In the Board's view, this can only mean that the diamine must cause asymmetry in the polymer chain, implying that the diamine must itself have an asymmetrical structure with regard to the groups which caused its incorporation into the polymer chain, or, in other words, the amine groups. Such diamines may, however, be symmetrical with reference to another axis. This is in conformity with the definition of the required diamines on page 9, first full paragraph to page 10, last paragraph but one, of the original description.
- 3.2 According to Claim 1, the oxydiphthalic dianhydride can contain up to 20 weight percent of other organic dianhydrides. The 100 weight percent basis for that amount is indicated on page 7, second full paragraph, of the original description as being the oxydiphthalic anhydride itself, and not the mixture of oxydiphthalic dianhydride and additional dianhydride. Likewise, the diamine providing the polymer chain with an asymmetrical structure can contain up to 25 weight percent of an organic diamine that is not asymmetrical in the polymer chain. The 100 percent basis for that amount is, however, not clearly indicated. In the

Board's view, the amount of additional diamine should, by analogy with the amount of additional dianhydride, be interpreted as referring to the diamine causing asymmetry in the polymer chain and not to the mixture of both kinds of diamine.

Novelty and inventive step

4. The Examining Division in its decision to refuse the application also indicated its willingness to recognize the novelty and inventiveness of the claimed subject-matter provided the definition of the organic diamine would be replaced by the list of diamines mentioned on page 10 of the original description (point 5 of the decision). However, no grounds for that opinion have been given and in particular it is not clear whether the problem-solution approach for assessing inventiveness, as commonly practised in the proceedings before the EPO, has been applied. Since the scope of the claimed subject-matter has been profoundly amended and the amendments do not exactly correspond to the Examining Division's proposal, the considerations and arguments upon which the Examining Division based its opinion regarding novelty and inventive step do not necessarily apply anymore. In these circumstances, in order not to deprive the Appellant from its right to appeal to a second instance, the Board deems it appropriate to make use of the power conferred upon it by Article 111(1) EPC to remit the case to the first instance for further prosecution.

Order

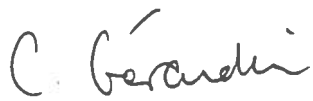
For these reasons it is decided that:

1. The decision under appeal is set aside.
2. The case is remitted to the first instance for further prosecution.

The Registrar:


E. Börner

The Chairman:


C. Gérardin

