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Aktenzeichen / Case Number / N° du recours : T 589/88

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Bezeichnung der Erfindung:
Title of invention:
Titre de l'invention : Copolymers utilising isoimides and method of preparing same

Klassifikation / Classification / Classement : C08G 73/12

ENTSCHEIDUNG / DECISION
vom / of / du 31 May 1990

Anmelder / Applicant / Demandeur : HUGHES AIRCRAFT COMPANY

Patentinhaber / Proprietor of the patent /
Titulaire du brevet :

Einsprechender / Opponent / Opposant :

Stichwort / Headword / Référence :

EPÜ / EPC / CBE Article 56

Schlagwort / Keyword / Mot clé : "Inventive step (yes) - acknowledged for the alternative embodiment providing a solution to the technical problem"

Leitsatz / Headnote / Sommaire

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Case Number : T 589/88 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 31 May 1990

Appellant : Hughes Aircraft Company
200 North Sepulveda Boulevard
El Segundo, CA 90245
USA

Representative : Kuhnen, Wacker & Partner
Schneggstrasse 3-5
Postfach 1729
D-8050 Freising

Decision under appeal : Decision of Examining Division 012 of the European Patent Office dated 25 April 1988 refusing European patent application No. 84 901 709.0 pursuant to Article 97(1) EPC

Composition of the Board :

Chairman : F. Antony
Members : C. Gérardin
J. Stephens-Ofner

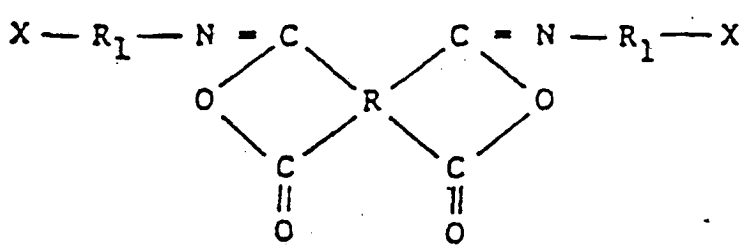
Summary of Facts and Submissions

I. European patent application No. 84 901 709.0, having an international (WIPO) filing date of 10 April 1984, claiming priority of 27 December 1983 from an earlier application in the United States and published on 4 July 1985 under the publication number WO 85/02853, was rejected by a decision of the Examining Division dated 25 April 1988, posted on 27 June 1988.

The rejection was based on a set of 14 claims of which Claim 1 filed on 25 April 1988 reads as follows:

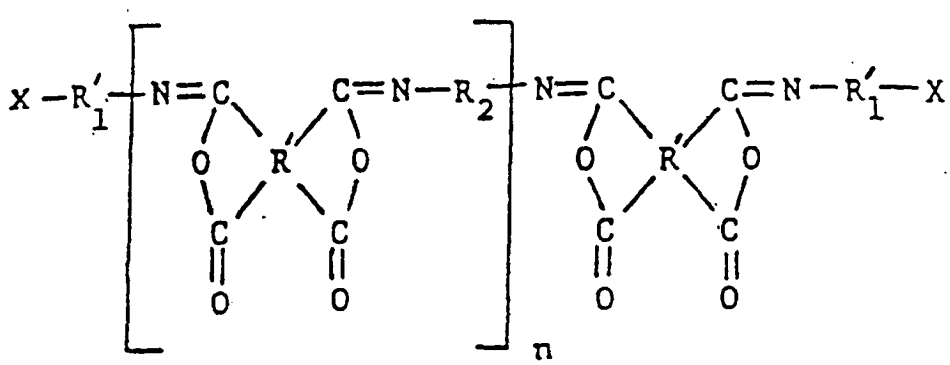
"The process for producing a copolymer of (A) an isoimide compound or oligomer having a structure:

(a)



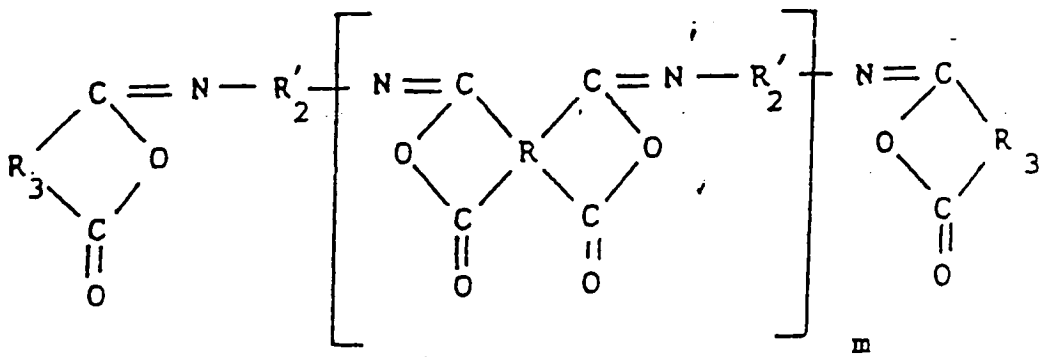
where R is a tetravalent organic group containing 2 to 27 carbon atoms, R_1 is a divalent organic group containing 1 to 20 carbon atoms, and X is selected from the group consisting of $-C\equiv CH$, $-CH=CH_2$, and -CN;

(b)



where R' is a tetravalent aryl group containing 6 to 18 carbon atoms, R'_1 is a C_6 to a C_{20} arylene group, R_2 is a divalent organic group containing 2 to 30 carbon atoms, X is as defined above, and n denotes the degree of polymerization and is 1 to about 30; or

(c)



where R is a tetravalent aryl group containing 2 to 27 carbon atoms, R'_2 is a divalent aryl group containing 6 to 30 carbon atoms, R_3 is a radical containing an alkenylene group, or a trivalent aryl group or heterocyclic group having substituted thereon X as defined above, and m denotes the degree of polymerization and is 1 to about 30 with (B)4,4'-bis-(3-ethynylphenoxy)-diphenyl sulphone (ATS) comprising:

(a) dissolving said isoimide (A) in (B) to produce a solution in liquid form; and

(b) heating said solution to cause said isoimide (A) and (B) to react to form said copolymer."

II. The ground for that decision was non-compliance with the requirements of Article 56 EPC with regard to the teaching of following documents:

(I) US-A-4 100 138
(II) EP-A1-71 372.

The decision under appeal stated that the alleged invention boiled down to the selection of an adequate compound to be copolymerised with the ethylenically unsaturated oligomers with isoimide groups described in document (II). A class of compounds suitable for this purpose was disclosed in document (I) which dealt with copolymers from ethynyl-substituted polyimide oligomers and di-or polyethynyl-substituted diluents. The choice of the most appropriate comonomer was in fact suggested in the introductory section of the application in suit itself, where the good rheological properties of acetylene-terminated phenylene resins in general, and of 4,4'-bis(3-ethynylphenoxy)-diphenyl sulphone in particular, were underlined. In the absence of any unexpected effect the selection made by Applicant (Appellant) could thus not be regarded as inventive.

III. A notice of appeal was lodged against the above-referred decision on 7 September 1988 with payment of the prescribed fee. The arguments presented in the Statement of Grounds of Appeal filed on 19 October 1988 can be summarised as follows:

The key feature of the process according to the application in suit was that the isoimide oligomer could be dissolved in ATS with or without further addition of solvent, and that the liquid blend was non-reactive until it was heated to the copolymerisation temperature. As appeared from Table II in Example XII, such liquid blends exhibited a remarkable increase in fluidity, which allowed them to be easily poured and made the penetration of the resin into the structure to be encapsulated, and thereby the encapsulation or moulding process, much easier. Moreover, the cured products possessed the high strength and temperature characteristics as well as the good electrical properties of the polyisoimide materials described in document (II).

IV. Together with the Statement of Grounds of Appeal, the Appellant filed two new sets of claims to be considered respectively as main and auxiliary requests.

Claim 1 according to the main request differs from Claim 1 as quoted in the Decision under appeal in only the following respects:

(i) step a) at the end has been replaced by the phrase "forming a solution of said isoimide (A) in said (ATS)(B) to produce a solution in liquid form";

(ii) the words "said ATS" before "(B)" in the last line;

(iii) two obvious errors have been introduced, viz. "≡CH" instead of the correct "-C≡CH" in the definition of X following formula (a) and "(b)" instead of the correct "(B)" in the penultimate line preceding step a); additionally "... sulphone" has been spelt "... sulfone" in the same line.

Claim 1 according to the auxiliary request differs from Claim 1 as quoted in the Decision under appeal in only the following respects:

(i) step a) at the end has been replaced by the phrase "dissolving said isoimide (A) in said ATS (B) to produce a solution in liquid form";

(ii) the words "said ATS" before "(B)" in the last line;

(iii) "20 to 40% by weight of" has been inserted before "(A)" in the second line of the claim and "80 to 60% by weight of" before "(B)" in the penultimate line preceding step (a);

(iv) the first above mentioned error and the same difference in spelling also occur.

V. The Appellant requests that the impugned decision be set aside and that a patent be granted on the basis of the set of 14 claims filed as main request or, alternatively, on the basis of the set of 11 claims filed as auxiliary request on 19 October 1988.

Reasons for the Decision

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

Main request

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

Claim 1 results basically from the incorporation into original process Claim 20 of the three structures (a), (b) and (c) of the isoimide compounds or oligomers as defined originally in Claims 10, 13 and 16 in combination with the preferred meanings of the radical X disclosed in original Claim 7 and on page 17, lines 1 to 4 of the original description, as well as the selection of ATS as compound (B) mentioned in original Claim 18.

The features of Claims 2 to 9, 10, 13 and 14 are to be found respectively in original Claims 6, 9, 11, 14, 17, 19, 7, 21, 22, 25 and 26. Claim 11 corresponds to the combination of original Claims 47 and 64. As to the features of Claim 12, they are disclosed on page 33, lines 5 to 10, of the original description.

3. The application in suit concerns a process for producing copolymers of unsaturated isoimide compounds or oligomers. Such compounds and oligomers are known from document (II) which mentions various embodiments to prepare such isoimide derivatives. The simplest structure results from the reaction of an organic carboxylic polyanhydride and a monoamine containing a functional group capable of undergoing addition polymerisation reactions (page 4, paragraph 2; page 7, paragraph 2); according to a second alternative, an aromatic polyamine is reacted with a dianhydride prior to reaction with the monoamine (page 14, paragraph 2; page 17, reaction scheme); according to a further method, the reaction product of a dianhydride and a diamine is end-capped with a monoanhydride containing a functional group capable of undergoing additional polymerisation reactions (page 20, paragraph 3 to page 22, reaction scheme). Besides terminal unsaturations, the resulting condensation products contain isoimide functional groups interspersed randomly throughout the main chain, which reduces the melting point of these products and improves the solubility thereof in common lacquer solvents with regard to the corresponding conventional polyimides (page 3, paragraph 1; page 6, paragraph 2 to page 7, paragraph 1; page 30, Table 1). These features and properties are used in polymerisable compositions based on homopolymers or copolymers, to produce void-free composite structures (page 3, paragraphs 2 to 4); however, in spite of the relatively low melting point of isoimide oligomers, adequate fluidity can only be achieved in the presence of lacquer solvents which give rise to objectionable volatile by-products upon heating.

In the light of this prior art teaching the technical problem underlying the application in suit may thus be seen as the provision of a curable composition which exhibits a high intrinsic fluidity and thereby ensures the production of void-free composite structures, even in the absence of lacquer solvents.

According to Claim 1 of the application in suit, this problem is solved by forming a solution of an isoimide oligomer according to one of the three above formulae (a) to (c) in ATS to produce a homogeneous solution, and then heating said solution to initiate the copolymerisation reaction.

4. After examination of the cited documents, the Board has come to the conclusion that this technical teaching is not disclosed in either of them and that the subject-matter of the application in suit according to the main request is, therefore, novel. Since the issue of novelty has not been raised by the Examining Division, it is not necessary to consider this matter in further detail.

5. It still remains to be examined whether the subject-matter of the application in suit according to the main request involves an inventive step with regard to the teaching of the cited documents.

5.1 The use of ethynyl-substituted compounds, such as diethynylbenzene, as copolymerisable diluents with polyimide oligomers is known from document (I). According to Claim 1 in combination with column 3, paragraph 5 thereof, di- or polyethynyl-substituted diluents are reacted with ethynyl-substituted polyimide oligomers to

provide laminating resins and composite materials. The incorporation of these reactive diluents lowers the viscosity of the resin; owing to the resulting good melt flow characteristics, laminates and composites can be obtained having zero void content together with high thermal stability and good mechanical properties (column 2, lines 19 to 63; column 3, lines 9 to 19). This advantage is attributed to the dual function of diethynyl-substituted compounds in the blend (column 4, lines 4 to 17): at first, they effectively thin out the oligomer when the latter is heated and moulded, allowing the molten oligomer to flow readily into the pores and crevices in fillers and fabric reinforcements; then, upon cure, they co-react with the oligomer, yielding a complex structure by cyclisation having a high cross-link density.

5.2 Although this prior art teaching is directed to polyimide oligomers and the application in suit concerns polyisoimide oligomers, this cannot be regarded as a significant difference.

As mentioned above when discussing document (II), the presence of isoimide groups interspersed randomly throughout the structure of the oligomer increases the solubility in lacquer solvents and lowers the melting point of polyisoimides with regard to the corresponding polyimides (page 30, paragraph 1 and Table 1). This means that the compatibility between polyisoimides and diethynyl-substituted diluents should be at least as good as that between the corresponding polyimides and these reactive diluents known from document (I). As to the curing mechanism, this cannot be affected by the replacement of imide groups by isoimide groups, since it only involves the same addition reactions.

In reality, the structural difference between isoimide groups and imide groups is at most a distinguishing feature before the reaction, since the properties and physical characteristics of cured resins made from polyisoimide oligomers are virtually the same as those of the resins derived from polyimide oligomers; as specified in document (II) (page 24, paragraph 2) and in the application in suit (page 33, lines 24 to 28), this similarity is attributed to the conversion of isoimide groups to the corresponding imide groups or isomerisation when the oligomer is subjected to elevated temperature during cure (page 24, paragraph 2).

5.3 As stated above, the reactive diluents according to document (I) are defined as di- or polyethynyl-substituted compounds, i.e. as compounds having ethynyl substituents as common functional feature. Although the description of this document is more specifically directed to diethynylbenzene used in both examples and to diethynyldiphenyl ether to a lesser extent (column 2, line 66 to column 3, line 3), the advantages put forward there, namely thinning effect on the polyimide oligomer and zero void content of the composite structure, must be regarded as general features obtainable with all these reactive diluents; in this respect, the selection of ATS within the class of diethynyl-substituted compounds does not provide particularly beneficial effects.

The argument that the lower cure temperature of the liquid blends at present claimed results from the selection of ATS, cannot be accepted (Statement of Grounds of Appeal, page 5, paragraph 3). It is not disputed that the liquid blends of polyisoimide oligomers and ATS at present claimed have a lower cure temperature than the polyisoimide oligomers alone as disclosed in document (II); however, this comparison is not conclusive, since it does not take

into account the beneficial effects of diethynyl-substituted compounds on polyimide oligomers already known from document (I). According to the only data available in this document, curing of a polyimide containing 10% by weight of diethynylbenzene occurs at 251°C; this temperature has to be viewed in the light of the fact that imide oligomers have a significantly higher gel temperature than the corresponding isoimide oligomers, about 60°C higher according to the figures in Table 1 of document (II). According to Example X of the application in suit, curing of a blend of 20% by weight of isoimide oligomer and 80% by weight of ATS starts at 150°C and post-cure is carried out stepwise up to 300°C. In view of the differences in ingredients and and their relative amounts, a strict comparison of the two curing temperatures would be meaningless; however, since the curing reaction involves the same cyclisation mechanism in both cases, the two temperatures can be regarded as being of the same order of magnitude. This means that the selection of ATS does not provide unexpected advantages in terms of curing temperature with regard to the ethynyl-substituent diluents considered in document (I) and that, therefore, there can be no question of an inventive selection.

5.4 Although the selection of ATS as such cannot be regarded as inventive for the reasons given above, it is essential to consider the question of the auxiliary solvent in the curable blend in the application in suit, which requires the interpretation of the features of the mixing operation of isoimide oligomer and ATS in the light of the description (Article 69 EPC).

The description specifies that in preparing mixtures of the isoimide oligomer and the sulfone compound, the mixture may be heated to a relatively low temperature to melt the solid compound and form a liquid blend of the two compounds.

Alternatively, it may be necessary to dissolve the oligomer and the sulfone compound in a common solvent, if the two reactants would not otherwise be soluble; the solvent is then stripped below the polymerisation temperature of the two reactants and the resulting solids containing the oligomer and the sulfone compound are heated to a relatively low temperature to melt the solid and form a liquid blend (page 32, line 6 to page 33, line 20).

In practice, both embodiments are illustrated in the application in suit. According to Examples XIII to XV, a blend having enough fluidity to be poured upon a substrate can be obtained by heating the mixture of the two ingredients moderately. By contrast, according to page 43, lines 2 to 6 in combination with Table II, films from blends of 20 to 100% by weight of isoimide oligomer and 80 to 0% by weight of ATS are cast on glass plates, employing a 10% solvent mixture of the blend in a solvent of 95% tetrahydrofuran and 5% N-methylpyrrolidone; the films are cured after a drying step. This embodiment, incidentally, corresponds to the method described in Examples 1 and 2 of document (I), wherein a conventional lacquer solvent is present additionally.

Thus, an interpretation of the scope of Claim 1 in the light of the description shows that the isoimide oligomer and ATS can be dissolved mutually with or without any additional solvent. That both embodiments are actually envisaged is in fact explicitly acknowledged in the Statement of Grounds of Appeal (point 2, paragraph 1) and the use of a solvent is even claimed in Claims 12 and 13. This means that the presence of a conventional solvent in the prior art blends cannot be regarded as a distinguishing feature; this also means that the subject-matter of Claim 1

encompasses one alternative process which involves the use of a lacquer solvent and therefore does not provide a general solution to the above defined technical problem.

5.5 In conclusion, for the various reasons given above, the solution claimed by the Appellant does not involve an inventive step.

6. Claim 1 not being allowable, the same applies to the dependent process Claims 2 to 14, which merely represent preferred embodiments of the subject-matter of Claim 1 and thus fall with it.

Auxiliary request

7. With regard to Claim 1 of the main request, Claim 1 of the auxiliary request additionally specifies that the copolymer derives from 20 to 40% by weight of the isoimide oligomer and 80 to 60% by weight of ATS; these ratios are disclosed in original Claim 64 and on page 30, lines 15 to 17 of the original description. As to the dissolution of the isoimide in ATS to produce a solution in liquid form, it corresponds to the embodiment which does not require the use of an additional lacquer solvent to prepare homogeneous liquid blends. There is thus adequate support for these amendments in the application as originally filed (Article 123(2) EPC).

As to the dependent process Claims 2 to 11, they correspond to Claims 2 to 10 and 14 of the main request and are thus formally allowable for the reasons mentioned above in point 2.

8. As far as the scope of Claim 1 is concerned, the Board follows the Appellant's interpretation of the claim in the Statement of Grounds of Appeal (page 9, point 4) according

to which the dissolution of the isoimide (A) in ATS (B) to produce a solution in liquid form occurs without the use of any solvent.

As apparent from Table II on page 42 of the application in suit showing the temperature of start of melting of blends of isoimide oligomers and ATS, the use of 60 to 80% by weight of ATS in the blend lowers the start of melting down to a range from 95 to 55°C depending on the degree of polymerisation of the isoimide oligomer. According to the description (page 42, lines 22 to 34), in this range of relative amounts of their ingredients, the blends exhibit sufficient fluidity at 135°C that they can be poured into a test tube or beaker, degassed and cured at 135 to 150°C and further post-cured up to 350°C. This alternative embodiment provides thus a solution to the above defined technical problem.

Moreover, the possibility as such to use as much as 60 to 80% by weight of ATS in the blend is regarded as surprising in view of the teaching of document (I) which specifies that diethynylbenzene should not exceed 20% by weight in the oligomer for safety reasons. Additionally, as noted in the application in suit (page 31, lines 1 to 7), the use of low amounts of the relatively expensive isoimide oligomer in the blends represents a financially substantial advantage, which is a further index of inventive step.

For these various reasons, Claim 1 of the auxiliary request is thus allowable.

9. These arguments apply equally to the dependent process Claims 2 to 11 which represent preferred embodiments of the subject-matter of Claim 1 and whose patentability is supported by that of the main claim.

10. As mentioned above, the present decision is based on the specific interpretation of Claim 1 of the auxiliary request according to which no lacquer solvent is involved in the mixing operation of the isoimide (A) and ATS (B). This should be explicitly acknowledged in the amended description to be filed, and any reference to other embodiments in step a) should be deleted accordingly.

Order

For these reasons, it is decided that:

1. The decision under appeal is set aside.

2. The main request is rejected.

3. The case is remitted to the Examining Division for further prosecution on the basis of Claims 1 to 11 filed on 19 October 1988 as auxiliary request and a description to be adapted according to point 10.

The Registrar: The Chairman:

M. Beer F. Antony