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D E C I S I O N
of 21 January 1999

Case Number: T 0916/95 - 3.3.3

Application Number: 89300619.7

Publication Number: 0326311

IPC: C08G 59/24

Language of the proceedings: EN

Title of invention:
Fiber reinforced composites with improved glass transition temperatures

Applicant:
Minnesota Mining and Manufacturing Company

Opponent:
-

Headword:
-

Relevant legal provisions:
EPC Art. 123(2), 84, 54, 56

Keyword:
"Amendments - added subject-matter (no)"
"Claims - clarity (yes)"
"Novelty - composition and method (yes)"
"Inventive step - unobvious combination of known features - no incentive"

Decisions cited:
-

Catchword:

-

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Beschwerdekammern | Boards of Appeal | Chambres de recours

Case Number: T 0916/95 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 21 January 1999

Appellant: Minnesota Mining and Manufacturing Company
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Decision under appeal: **Decision of the Examining Division of the European Patent Office posted 3 July 1995 refusing European patent application No. 89 300 619.7 pursuant to Article 97(1) EPC.**

Composition of the Board:

Chairman: C. Gérardin
Members: B. ter Laan
J. Stephens-Ofner

Summary of Facts and Submissions

I. European patent application No. 89 300 619.7, filed on 24 January 1989, claiming priority of 27 January 1988 from an earlier application in the USA (US 149101) and published on 2 August 1989 under No. 0 326 311 was refused by a decision of the Examining Division of the European Patent Office dated 3 July 1995. That decision was based on a set of 13 claims filed on 16 May 1995, Claim 1 reading:

"A fiber reinforced composite structure having a glass transition temperature greater than 150°C comprised of:

a) reinforcing fibers which optionally are metal coated, and
b) a cured epoxy resin composition, the uncured precursor of which comprising
   (i) one or more epoxides having more than one epoxide group per molecule, of which epoxides at least 10 percent by weight is a 9,9-bis[4-(2,3-epoxypropoxy)phenyl]fluorene which optionally is substituted by at least one halogen atom, all of the fluorene epoxy present being free of ortho substitution;

(ii) an effective amount of one or more epoxy curing agents in said curable composition, said curing agent being selected from the group consisting of polybasic carboxylic acids and their anhydrides; aliphatic, cycloaliphatic, and aromatic amines; carboxylic acid amides; said composition being free of fluorene-containing amine curing agents, the curing agent being chosen as to effect a sufficiently low dissolution temperature of the curing agent on the epoxy resin composition to allow hot melt prepreg filaments, fibres or fabrics."

Dependent Claims 2 to 9 referred to preferred embodiments of the composite structures according to Claim 1, Claim 10 was directed to a printed wiring board comprising the cured composite structure of Claim 9.

Independent Claim 11 read:

"A composition capable of curing to a fiber reinforced composite structure having a glass transition temperature greater than 150°C, said composition comprising:

a) reinforcing fibers which optionally are metal coated, and

b) a curable epoxy resin composition comprising

(i) one or more epoxides having more than one epoxide group per molecule, of which epoxides at least 10 percent by weight is a

9,9-bis[4-(2,3-epoxypropoxy)phenyl]fluorene which optionally is substituted by at least one halogen atom, all of the fluorene epoxy present being free of ortho substitution;

(ii) an effective amount of one or more epoxy curing agents dissolved in said curable composition, said curing agent being selected from the group consisting of poly basic carboxylic acids and their anhydrides; aliphatic, cycloaliphatic, and aromatic amines; carboxylic acid amides; said composition being free of fluorene-containing amine curing agents, the curing agent being chosen as to effect a sufficiently low dissolution temperature of the curing agent on the epoxy resin composition to allow hot melt prepreg filaments, fibres or fabrics."

Claim 12 and Claim 13, although they did not refer directly to Claim 11, concerned a method of preparing a composition as described in that claim and a method of preparing a fibre reinforced composite structure from the composition of Claim 11, respectively.

II. The Examining Division held that the claimed subject-matter was novel, but did not involve an inventive step. In particular, it found that, starting from D1 (US-A-4 684 678), the subjective problem was seen as to provide fibre reinforced epoxy compositions having an improved melt-processability and which, when cured, still exhibited a high glass transition temperature. However, the wording of the claims also included compositions less suitable than those described in D1. Therefore, the objective problem was to provide alternative fibre reinforced fluorene epoxy/curing agent compositions having a high glass transition temperature and which were suitable for hot melt prepregging. The patent in suit proposed to solve that problem by using epoxy curing agents free of fluorene-group containing amine curing agents and chosen such as to provide a sufficiently low dissolution temperature of the curing agent to allow for hot melt prepregging of filaments, fibres or fabrics. Since the skilled person would recognize that the curing agents used in D1 might have a too high dissolution temperature for hot melt prepreg applications, he would replace them by conventional curing agents, expecting, in the light of the disclosure of D3 (US-A-4 707 534), no deterioration of the glass transition temperature. In view of the available evidence it could not be accepted that a prejudice against the use of conventional curing agents had been overcome, so that the claimed subject-matter was obvious.

III. On 18 August 1995 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

9 November 1995, the Appellant (Applicant) submitted two sets of claims as the main and an auxiliary request, the latter corresponding to the request upon which the decision of the Examining Division was based. The Appellant also commented upon the issues of Article 123(2) EPC and inventive step.

IV. After a communication from the Board, in which several objections under Articles 123(2), 84, 54 and 56 EPC were raised, on 18 December 1998 two new sets of claims were filed replacing the claims then on file. At the oral proceedings before the Board, held on 21 January 1999, after further objections by the Board, those claims were again replaced by two new sets of claims as the main and one auxiliary request. Claim 1 of the main request reads a follows:

"A fiber reinforced composite structure having a glass transition temperature greater than 150°C consisting of:

a) reinforcing fibers which optionally are metal coated, and

b) a cured epoxy resin composition, the uncured precursor of which is suitable for use in hot melt prepregging filaments, fibers or fabrics and comprises a homogeneous mixture consisting of:

(i) one or more epoxides having more than one epoxide group per molecule, of which epoxides at least 10 percent by weight is a 9,9-bis[4-(2,3-epoxypropoxy)phenyl]fluorene which optionally is substituted by at least one halogen atom, all of the fluorene epoxy present being free of ortho substitution;

(ii) an effective amount of one or more epoxy curing agents, said curing agent being selected from the group consisting of polybasic carboxylic acids and their anhydrides; aliphatic, cycloaliphatic, and aromatic amines; carboxylic acid amides; said composition being free of fluorene-containing amine curing agents."

Dependent Claims 2 to 9 refer to preferred embodiments of the composite structures according to Claim 1, Claim 10 is directed to a printed wiring board comprising the fiber reinforced composite structure of any of the previous claims.

Independent Claim 11 reads as follows:

"A composition capable of curing to a fiber reinforced composite structure having a glass transition temperature greater than 150°C, said composition consisting of:

a) reinforcing fibers which optionally are metal coated, and

b) a curable epoxy resin composition which is suitable for use in hot melt prepregging filaments, fibers, or fabrics and comprising a homogeneous mixture consisting of:

(i) one or more epoxides having more than one epoxide group per molecule, of which epoxides at least 10 percent by weight is a 9,9-bis[4-(2,3-epoxy-propoxy)phenyl]fluorene which optionally is substituted by at least one halogen atom, all of the fluorene epoxy present being free of ortho substitution;

(ii) an effective amount of one or more epoxy curing agents dissolved in said curable composition, said curing agent being selected from the group consisting of poly basic carboxylic acids and their anhydrides; aliphatic, cycloaliphatic, and aromatic amines; carboxylic acid amides; said composition being free of fluorene-containing amine curing agents."

Independent Claim 12 refers to:

"A method of preparing a fiber reinforced composite structure comprising:

(i) mixing one or more epoxides having more than one epoxide group per molecule, of which epoxides at least 10 percent by weight is a 9,9-bis[4-(2,3-epoxy-propoxy)phenyl]fluorene which optionally is substituted by at least one halogen atom, all of the fluorene epoxy present being free of ortho substitution;

(ii) dissolving an effective amount of one or more epoxy curing agents in said curable composition, said curing agent being selected from the group consisting of poly basic carboxylic acids and their anhydrides; aliphatic, cycloaliphatic, and

aromatic amines; carboxylic acid amides; said composition being free of fluorene-containing amine curing agents;

(iii) optionally, heating the resulting mixture of (i) and (ii) to effect the dissolution of the curing agent and/or degassing the mixture;

(iv) hot melt impregnating the resulting mixture of (i), (ii) and (iii) into filaments, fibers or fabrics; and

(v) curing the resulting laminating structure of (iv) in the temperature range of 50 to 250°C."

The auxiliary request contains as sole claim the subject-matter of Claim 12 of the main request.

The Appellant's arguments submitted in writing and during oral proceedings, can be summarised as follows:

(i) Regarding Article 123(2) EPC, the homogeneity of the curable epoxy resin composition was implied by the disclosure of the requirement that the curing agent should be dissolved in the epoxide composition and that this mixture was used for hot melt prepregging.

(ii) As to clarity, the use of the term "consisting of" for the structure as a whole as well as the suitability of the uncured precursor composition for hot melt prepregging excluded the presence of a solvent, since solvent containing mixtures could not be used for that purpose. Thus, it was clear that the absence of a solvent was a

requirement of the claimed subject-matter.

(iii) Regarding novelty, the compositions disclosed in D1, as illustrated in Example 10, did not form a homogeneous mixture without a solvent and hence were not suitable for hot melt prepregging, due to the use of the curing agents described there. D3 referred only in a general way to composites; it did not describe the composite structures now being claimed. Therefore, the claimed subject-matter was novel.

(iv) As regards inventive step, D1 was the closest document as it specifically taught fibre reinforced compositions. The object of the invention was to provide a solventless prepreg system resulting in fibre reinforced structures having a high glass transition temperature. As could be seen from the examples in the application in suit, the fibres influenced the glass transition temperature to such an extent that no conclusion regarding that property could be drawn on the basis of non-reinforced cured epoxy compositions. In view of this, the information contained in D3, referring to such systems, was not relevant for the problem to be solved. Therefore, D3 did not provide an incentive to the skilled person to change the composition of D1 in the sense of the present claims. Hence, the claimed subject-matter involved an inventive step.

V. The Appellant requested that the decision of the first

instance be set aside and that a patent be granted on the basis of Claims 1 to 12 of the main request or, alternatively, Claim 1 of the auxiliary request.

Reasons for the Decision

1. The appeal is admissible.

Main request

Article 123(2) EPC

2. The amendments to the claims are in conformity with the requirements of Article 123(2) EPC.

2.1 Claim 1 of the main request differs from the one as originally filed in that the unclear expression "comprised of" has been replaced by "consisting of". This is based upon the original description, page 11, second full paragraph, as well as Examples 5, 7, 9, 11 and 13, which refer to hot melt prepregging and in which no other material is present apart from the ones mentioned in Claim 1. On page 1, lines 19 to 29 hot melt prepregging is described in detail, as opposed to "other processing techniques" described on page 1, lines 30 to 34, of which solvent prepregging is one. Those passages also form the basis for the added requirement that the uncured precursor of the cured epoxy resin composition b) should be suitable for hot melt prepregging. The amendment of "curable" to "cured" for the epoxy resin composition b) finds its basis in original Claim 10.

2.2 The deletion in Claim 2 of the minimum amount of adjuvant is supported by the paragraph bridging original pages 11 and 12.

2.3 The amendment in Claim 3 of "lower alkyl" to C_1-C_4 alkyl finds its basis in original page 6, line 2.

2.4 Claim 11 has no direct counterpart in the original claims. A mixture of a curable epoxy resin composition and optionally metal coated reinforcing fibres was however referred to in original Claim 1. The additional amendments are the same as those in present Claim 1.

2.5 Claim 12 refers to a method for preparing a fibre reinforced structure. Originally, such subject-matter was not claimed. It was, however, mentioned on page 11, second full paragraph. The absence of ortho substitution in the fluorene compounds is implied by the passage on page 10, third full paragraph: "One method of forming the fiber matrix composition **of the invention**..." (emphasis added) in conjunction with page 11, second full paragraph: "An alternative method...".

Clarity

3. From the wording of Claims 1, 11 and 12 it is clear that the composite structure may contain no other compounds than the reinforcing fibres and the curable epoxy resin precursor. In particular, the presence of a solvent either in the curable precursor composition (by the requirement "suitable for use in hot melt prepregging") or in the composition as a whole (by the expression "consisting of") is excluded, while the presence of other compounds, e.g. adjuvants in the curable precursor composition (by the expression

"comprises") is still allowed. Therefore, the Board is satisfied that the claims do in fact only refer to hot melt prepregging systems, as argued by the Appellant.

Novelty

4. In view of the objection of lack of novelty raised by the Board in the course of the appeal proceedings, it seems appropriate to deal with this issue with respect to both D1 and D3.

4.1 D1 describes an epoxy resin composition comprising (a) at least one aromatic polyepoxide, and (b) at least one 9,9-bis(aminophenyl]fluorene curing agent present in an amount sufficient to provide in the range of 0.1 to 1.1 amino groups, -NHR, per epoxy group in said aromatic polyepoxide, wherein each R is independently a hydrogen or a linear or branched alkyl group of 1 to 6 carbon atoms (Claim 1). In Example 10, in conjunction with Example 3, a mixture of 2,2-bis[4-(2,3-epoxypropoxy)phenyl]propane, 9,9-bis[4-(2,3-epoxypropoxy)phenyl]fluorene, 9,9-bis(4-aminophenyl)-fluorene, a curing agent (79.5 wt.% of 9,9-bis(4-methylaminophenyl)fluorene, 20.1 wt.% 9-(4-methylaminophenyl)-9-(4-aminophenyl)fluorene and 0.4 wt.% 9,9-bis(aminophenyl)fluorene) and methyl ethyl ketone is sonicated to form a uniform dispersion, with which a prepreg was prepared of pieces of graphite fabric.

The fibre reinforced composite structure thus obtained as well as the uncured precursor mixture differ from the present claims at least in that the curing agent is

a fluorene-containing amine compound, so that D1 does not anticipate the subject-matter of Claim 1.

4.2 D3 describes glycidyl ethers of ortho-substituted-4-hydroxyphenylfluorenes (Claim 1) and curable epoxy resin compositions comprising (a) at least one curable epoxy resin of which at least 25 percent by weight is a glycidyl ether of an ortho-substituted-4-hydroxyphenylfluorene; and (b) epoxy resin curatives (Claim 8), as well as the cured products of that curable resin composition (Claim 13) and substrates impregnated or bearing a layer of that curable composition (Claim 14). The curing agents that can be used are well-known in the art and include fluorene-containing amine compounds (column 8, lines 32 to 66). Reinforcing material like woven and nonwoven webs of fibres can also be added to the compositions (column 9, lines 27 to 40) and woven and unwoven webs can be impregnated with the composition to produce composite articles (column 9, line 64 to column 10, line 3). In Table I various cured compositions containing glycidyl ethers of ortho-substituted-4-hydroxyphenylfluorenes are compared with a cured composition containing fluorenes free of ortho-substitution (Comparative Example C). That table does however not refer to fibre reinforced composite structures; in fact, D3 only mentions the possibility of preparing composites out of mixtures containing ortho-substituted fluorenes in a general way without giving any specific example. Therefore, although Comparative Example C of D3 discloses the uncured precursor mixture of epoxy resin and curing agent, in which the latter is dissolved in the former, and also the cured product of that mixture,

it does not mention the impregnation of reinforcing fibres with that particular mixture free of ortho-substituted fluorenes, either with or without the use of a solvent. Therefore, D3 does not prejudice the novelty of the subject-matter of Claim 1.

Inventive step

5. The application in suit concerns fibre reinforced composites with improved glass transition temperatures.

According to the description of the application in suit the object of the invention is to provide fibre reinforced composites with improved glass transition temperatures (page 3, lines 26 to 33, page 4, lines 17 to 23 and page 12, second full paragraph), in particular above 150°C (page 3, lines 2/3 and 33/34).

5.1 Such fibre reinforced composite structures having high glass transition temperatures were already known from e.g. D1, Example 10, in which a composite with a glass transition temperature of 214°C is prepared by using the solvent prepregging technique. Since D3 does not disclose any specific detail regarding fibre reinforcement, the Board, like the Examining Division and the Appellant, considers D1 as the closest state of the art.

5.2 In view of the disclosure in D1 of fibre reinforced composites having high glass transition temperatures, the technical problem underlying the application in suit has to be reformulated. In the light of the

desirability for environmental reasons of solvent-free systems, hot melt prepregging would offer a solution; hence the problem to be solved is seen as to provide fibre reinforced composite structures prepared by hot melt prepregging without any deterioration of the high glass transition temperature as obtained with solvent prepregging.

5.3 According to the application in suit that problem is to be solved by the fibre reinforced composite structures as defined in Claim 1.

5.4 The examples in the application show that the above-defined problem is effectively solved. In particular, it has been shown that the composites according to Claim 1 have high glass transition temperatures.

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

6.1 According to the general teaching of D1, epoxy resin compositions which on curing yield resins having a high glass transition temperature, high ductility and low moisture pick-up are obtained by using 9,9-bis(amino-phenyl)-fluorenes as curing agents (column 2, lines 58 to 67). According to D1, the compositions it describes are "useful in protective coatings for various articles such as appliances, for impregnating and embedding materials for electrical components, for composite articles of woven or nonwoven webs impregnated with the composition" (column 11, lines 45 to 50). No details about prepregging are given apart from Example 10, which involves solvent prepregging. D1 provides however

no information that a solvent-free system could also be used provided one did not use ortho-substituted fluorene epoxy compounds, nor fluorene-containing amine curing agents. Therefore, D1 by itself cannot render obvious the subject-matter of Claim 1.

6.2 D3 describes the use of diglycidyl ethers of ortho-substituted-4-hydroxyphenylfluorenes for the preparation of curable compositions and cured resins thereof having a high glass transition temperature and improved modulus of elasticity (column 2, lines 18 to 22). Any kind of curing agent can be used; both conventional ones as well as fluorene-containing amine compounds (column 8, lines 32 to 66). Even if di(aminophenyl)sulfone is the only curing agent used in the examples, nothing in D3 would induce the skilled person to turn away from the other curing agents.

Liquefaction by heating of a mixture of the polyepoxides and the curing agents is also described, but it is said that heat is used to liquefy the mixture only if the mixture is not already liquid or if solvents have not been used and that, in general, liquefaction of the mixture is accomplished by dissolving it in any suitable organic solvent (column 9, lines 18 to 26). The latter statement points away from solvent-free systems. As neither the description nor the examples specifically refer to impregnation systems, D3 provides no teaching in that respect.

Therefore, the general teaching of D3 is to cure ortho-substitued fluorenes with any kind of curing agent in

order to obtain resins with a high glass transition temperature and modulus of elasticity. Accordingly, nothing in D3 would provide the skilled person with the incentive to combine D3 with D1 in order to arrive at the desired result, nor would any combination of those documents lead to modifying the composition described in D1 into the specific composite structure defined in present Claim 1.

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

7. The above considerations also apply to independent Claims 11 and 12 since their subject-matter is based on the same combination of features as Claim 1.

8. As Claim 1 of the main request is allowable, the same goes for dependent Claims 2 to 10, the patentability of which is supported by that of Claim 1.

9. Since the Appellant's main request is granted, there is no need to consider the auxiliary request.

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 the main request and after any consequential and necessary amendment of the description.

The Registrar: The Chairman:

E. Görgmaier C. Gérardin