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File Number: T 0124/91 - 3.3.3

Application No.: 84 200 107.5

Publication No.: 0 127 198

Title of invention: Preimpregnated reinforcements and high strength composites  
therefrom

Classification: C08G 59/50

DECISION  
of 31 March 1993

Applicant: Amoco Corporation

Opponent: BASF Aktiengesellschaft  
Ciba Geigy AG

Headword:

EPC Article 56

Keyword: "Inventive step (yes, after amendment of the claims)"



Case Number : T 0124/91 - 3.3.3

**D E C I S I O N**  
of the Technical Board of Appeal 3.3.3  
of 31 March 1993

**Appellant :**  
(Proprietor of the patent)

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(Opponent 01)

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**Decision under appeal :**

Decision of the Opposition Division of the  
European Patent Office dated 9 November 1990,  
issued in writing on 29 November 1990 revoking  
European patent No. 0 127 198 pursuant to  
Article 102(1) EPC.

**Composition of the Board :**

**Chairman :** C. Gérardin  
**Members :** H. Fessel  
S. Perryman

## Summary of Facts and Submissions

- I. European patent No. 0 127 193 based on European patent application 84 200 107.5, filed on 27 January 1984 and claiming the priority of 20 May 1983 from an earlier application in the United States (US-496 504), was granted with 14 claims on 23 December 1987 (Bulletin 87/52) to Amoco Corporation (Appellant).

This set of claims comprised, first, a main claim and ten dependent claims, i.e. Claims 2 to 10 and 13, directed to a composition, then two claims concerning a prepreg based on the same composition, i.e. Claims 11 and 12, and last Claim 14 related to a cured article prepared from the product of any of the preceding claims.

- II. On 24 March and 21 September 1988 BASF (Respondent 01) and Ciba Geigy (Respondent 02) filed a notice of opposition based on Article 100(a) EPC. To support the oppositions the following documents still relevant in the appeal proceedings were cited:

- (1) JP-A-5464599 (English translation)
- (2) SU-A-476 296 (German translation and Derwent abstract)
- (6) Encyclopedia of Chemical Technology, Kirk Othmer, Vol. 9, pages 277 and 289 (Wiley 1980).

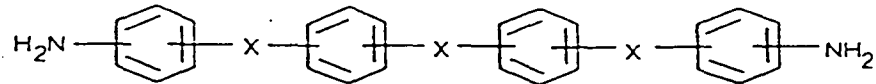
- III. By a decision which was given at the end of oral proceedings held on 9 November 1990 and issued in writing on 29 November 1990, the Opposition Division held that the subject-matter as defined in the three requests, i.e. the main and two subsidiary requests, met the requirements of Article 123(2) and 123(3) EPC and was novel, but did not involve an inventive step.

The main and the first subsidiary requests were directed to fibre filled compositions, whereas the subject-matter of the claims of the second subsidiary request was the use of a fibre filled composition for producing structural components in aircraft.

Claim 1 of this request read as follows:

"1. The use of a composition comprising:

- (a) 5 to 60 weight percent of a diamine hardener represented by the following general formula:



wherein the X's are independently selected from a direct bond, O, S,  $SO_2$ , CO, COO,  $C(CF_3)_2$ ,  $C(R_1, R_2)$  wherein  $R_1$  and  $R_2$  are independently hydrogen or alkyl of 1 to 4 carbon atoms;

- (b) 10 to 60 weight percent of an epoxy resin containing two or more 1,2 epoxide groups per molecule, and
- (c) 3 to 85 weight percent of a structural fiber selected from carbon fiber, aromatic fiber and silicon carbide fiber and having a tensile strength of greater than  $6.9 \times 10^2$  MPa, a tensile modulus of greater than  $13.8 \times 10^3$  MPa, and a decomposition temperature of greater than  $200^\circ C$  for producing structural components in aircraft."

As to the subject-matter of this claim the Opposition Division held document (6), page 269, to represent the closest prior art and the claimed subject-matter to be novel over said prior art, since the tetra aromatic

diamine as specified under item (a) of Claim 1 was not disclosed therein. Such diamine curing agents were, however, known in the art (cf. documents (1) and (2)). Moreover, (1) taught that such a composite cured with a diaromatic hardener yielded a product having a higher stiffness, as measured by the torsion modulus, than that cured by a curing agent falling within the scope of the claims, i.e. a tetraaromatic hardener. Since stiffness appeared to the Opposition Division to be a crucial feature of structural parts for aircraft and said properties were inferior in comparison with those discussed in (6), inventive step could not be based thereupon. There was no evidence what specific problem was solved by the use of the specified generally known curing agents. The claimed subject-matter thus did not involve any inventive step.

- IV. On 24 January 1991 a notice of appeal against the above decision was filed, together with payment of the prescribed fee. With the statement of grounds of appeal filed on 8 April 1991, the Appellant (Patentee) filed a new set of 10 claims, wherein both the epoxy resin component and the application had been defined more specifically.

Together with the statement of grounds of appeal the Appellant also filed comparative tests intended to demonstrate that a sample of the composition of the disputed patent showed a remarkable improvement of compressive strength under dry conditions as well as hot/wet conditions. Moreover the water absorption of the material according to the prior art was by 62.5% higher than in the patent in suit (1.3 versus 0.8). Since said improvement could not have been predicted by the man skilled in the art, it amounted to evidence for inventive step.

Together with the statement filed on 11 February 1993 the Appellant filed a further set of claims to be considered as an auxiliary request.

V. Respondent 02 informed the Board that he would not comment on the arguments presented in the statement of grounds of appeal, whereas Respondent 01 contested that the said properties were surprising, since with regard to what was known from (1) and (2) it was obvious to try the tetraaromatic hardeners for producing parts of aircraft generally known from (6).

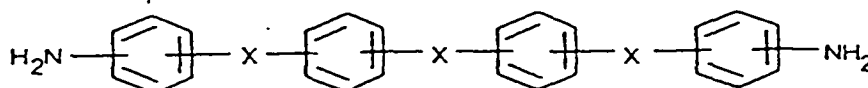
Both Respondents informed the Board that they would not attend the oral proceedings.

VI. Oral proceedings were held on 31 March 1993. During said oral proceedings the Appellant emphasised his previous submissions, underlining that the problem to be solved right from the beginning was to produce composites with high compressive strength. To demonstrate the importance of compressive strength especially under hot/wet conditions for the intended use, he provided some late published documents.

Moreover he withdrew his former requests and submitted a new set of 8 claims together with a description adapted thereto. The only independent claim of said set of claims reads as follows:

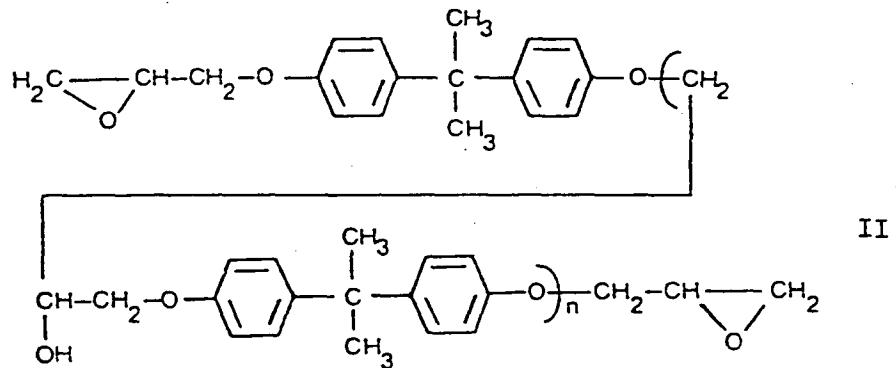
"The use of a composition comprising:

- (a) 5 to 60 weight percent of a diamine hardener represented by the following general formula:

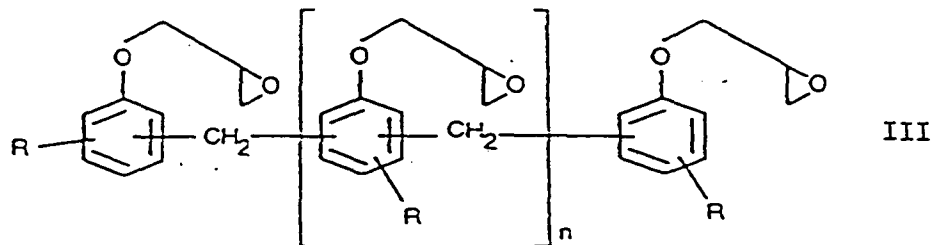


wherein the X's are independently selected from a direct bond, O, S, SO<sub>2</sub>, CO, COO, C(CF<sub>3</sub>)<sub>2</sub>, C(R<sub>1</sub>,R<sub>2</sub>) wherein R<sub>1</sub> and R<sub>2</sub> are independently hydrogen or alkyl of 1 to 4 carbon atoms;

- (b) 10 to 60 weight percent of an epoxy resin selected from bis(2,3-epoxycyclopentyl) ether copolymers of bis(2,3-epoxycyclopentyl) ether with ethylene glycol, mixtures of bis(2,3-epoxycyclopentyl) ether with a bisphenol A epoxy resin of the formula:



where n has a value from about 0 to about 15; or with an epoxidized novolak resin of the formula:



where n is 0.1 to 8 and R is H or CH<sub>3</sub>,

or with N,N-diglycidyl aniline or N,N,N',N'-tetraglycidyl-4,4'-diaminodiphenyl methane;

- (c) 3 to 85 weight percent of a structural fiber having a tensile strength of greater than 6.9 x 10<sup>2</sup> MPa, a tensile modulus of greater than 13.8 x 10<sup>3</sup> MPa, and a decomposition temperature of greater than 200°C as such or in the form of prepregs or composites, for

producing aircraft parts, automotive parts, pressure vessels, tanks, pipes, protective armor on military vehicles and sporting goods."

VII. The Appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of the claims and description submitted during the oral proceedings.

Respondent 02 requested with his letter dated 19 June 1991 that the appeal be dismissed.

Respondent 01 did not file any request.

#### **Reasons for the Decision**

1. The appeal is admissible.
2. There are no formal objections under Article 123(2) EPC since the subject-matter of Claims 1 to 8 is supported by the original disclosure.

Claim 1 is supported:

as to the amounts of the components (a), (b) and (c) by page 11, lines 5 to 9 (Claim 1 of the patent specification);

as to the selected epoxy resin component (b) by page 8, lines 15 to 17 in conjunction with page 10, lines 3 to 10 and page 13, lines 1 to 4 of the original files (page 4, line 65 to page 5, line 8 in conjunction with lines 49 to 52 and page 6, lines 30 to 32 of the patent specification);

as to the structural fibre component (c) by page 10, lines 26 to 30 as originally filed (Claim 1 of the patent specification) and as to the intended use by page 13, line 22 to page 14, line 2 in conjunction with Claims 31 to 36 and 38 as originally filed (page 6, lines 41 to 46 in conjunction with Claims 11 to 14 of the patent specification).

The remaining Claims 2 to 8 find their basis in Claims 2, 7, 24, 25 and page 11, lines 5 to 9 as originally filed (Claims 2, 3 and 6 to 10 in the patent specification). The description in the patent in suit has been suitably adapted to the revised claims.

The Board is also satisfied that the provisions of Article 123(3) EPC are met by the claims since the subject-matter - use of a composition for producing aircraft parts, automotive parts, ..., and sporting goods, instead of a composition as such - has not been amended in a way as to extend the protection conferred beyond the scope of Claim 14 of the patent as granted (identical to Claim 14 as originally filed) which was directed to the cured article prepared from the product of any preceding claims. Even if the use claims are considered as being in reality process claims to produce the specified (cured) product, such a product was anyway within the scope of original Claim 14.

3. The Board is satisfied that the claimed subject-matter is novel over the cited prior art, which has not been disputed in appeal proceedings. Further details to support the Board's view are thus not necessary.
4. According to the present version of the claims the patent in suit concerns the use of composites as structural components. Such application of epoxy resin compositions is disclosed in document (6) which the Board, like the

Opposition Division, regards as the closest state of the art.

Said document discloses e.g. graphite fibre reinforced composites based on high performance polyfunctional resins, such as the tetraglycidyl derivative of methylenedianiline in combination with diaminodiphenylsulphone, having good elevated temperature (page 289, paragraphs 1 and 3). In spite of a high strength to weight ratio which makes these composites particularly suitable in the aerospace industry, the mechanical properties under hot/wet conditions cannot be regarded as optimal. In view of this shortcoming, the technical problem underlying the patent in suit may be seen in providing a composition leading to composite materials having improved mechanical characteristics under hot/wet conditions for use in the aircraft industry.

The solution proposed according to Claim 1 is (i) to replace the diamine hardener according to document 6), which was characterised by two benzene rings, by diamine hardeners (a) having four benzene rings in the molecule used in specific amounts, and (ii) to combine them with epoxy resins selected from those specified under (b). As structural fibres (c), in addition to glass, graphite, boron and Kevlar fibres, fibres with the specified properties may be used.

According to the data provided by the Appellant with the grounds of appeal, it can be seen that, compared with the control composition containing a diamine with two phenylene groups (diaminodiphenylsulphone) as the hardener component, an otherwise identical composition in which this hardener is replaced by a diamine hardener with four phenylene groups according to the patent in suit shows a marked reduction in water uptake;

furthermore, composites prepared therefrom using carbon fibres have a distinctly better compressive strength under hot/wet conditions. In view of said data, the Board is satisfied that the above problem is effectively solved with the use specified in Claim 1.

5. It remains to be considered whether the given solution involves an inventive step with regard to the disclosure of the cited prior art.

5.1 Document (6) discloses structural composites comprising epoxy resins in combination with glass, graphite, boron and Kevlar fibres in the manufacture of high performance composites to be used e.g. in the aerospace industry. High performance polyfunctional resin, such as the tetraglycidyl derivative of methylenedianiline in combination with diaminodiphenylsulphone are said to be used to provide good elevated temperature properties and humidity resistance.

No suggestion can be read into (6) to the use of tetraaromatic diamine hardeners instead of diaromatic diamine hardeners. Even if it is assumed that the use of such graphite fibre reinforced composites in aerospace industry implies a certain amount of compressive strength and humidity resistance, the skilled person would get no hint from the disclosure of document (6) of modifications that might lead to improved compressive strength and water absorption.

5.2 Since tetraaromatic amine hardeners are known hardeners in the field of thermosetting epoxy resin compositions and composites as shown by documents (1) and (2), the issue of inventive step boils down to the question whether a skilled person seeking to solve the above problem would combine the teachings of (6) with (1) and/or (2).

Document (1) describes a thermosetting resin laminate which is said to have excellent thermal resistance, flexibility and drilling workability. The laminate is made by impregnating a substrate with a thermosetting varnish consisting of an epoxy resin (e.g. Bisphenol A-diglycidyl ether), a tetraaromatic diamine and preferably an organic solvent; drying to obtain prepregs, laminating such prepregs, and, if desired, copper foil, and heating and shaping the laminate under pressure (cf. claim).

A comparison between Example 1 and comparative Example 2 shows that using a binuclear hardener instead of a tetra nuclear hardener, as defined in the patent in suit, gives a product of higher torsional rigidity (5.5 versus 2).

Even accepting that the quality referred to as "flexibility" in document (1) is in fact "lower brittleness" as canvassed by the Respondent BASF and demonstrated by the comparison above using the tetranuclear hardeners, there is no discussion in the citation of the behaviour of these compositions under hot/wet conditions, thus no suggestion that compression strength under hot/wet conditions would be improved when a fibre material such as carbon is impregnated with said varnish.

Consequently, the skilled person would have had no reason to suppose that the technical problem could be solved by using tetranuclear amines.

- 5.3 It remains to be considered whether the disclosure of document (2) alone or in combination with the other documents hints at a solution to the problem underlying the patent in suit. Document (2) mentions an epoxide resin composition which is said to have excellent water and heat resistance. The table on page 4 shows a water absorption of from 0.007 to 0.03% in 24 hours at 25°C for

the epoxy composition i.e. the varnish. It is said in that document that the specified tetranuclear hardener improves water and heat resistance vis-à-vis the binuclear bis(aminoxylyl) sulphone. As far as water absorption is concerned, the results given in the table provide evidence that a marked improvement can be obtained by curing epoxide compounds with a tetranuclear hardener. However, in the absence of any reference in the citation to compressive strength, this beneficial effect cannot be related to the solution of the above-defined technical problem. It follows that the teaching of document (2) cannot encourage the man skilled in the art to replace the binuclear hardeners known from document (6) by a tetranuclear hardener, such as bis(4-aminophenoxyphenyl) sulphone, when seeking to improve hot/wet compressing strengths of laminates.

- 5.4 . Apart from the fact that the prior art relied upon by the Respondents does not provide an incentive to use tetranuclear amines, the choice of specific cycloaliphatic epoxy resins or mixtures based on such polyepoxides must be regarded as a further feature involving an inventive step.

This becomes quite clear from a study document (6), whose teaching boils down to the use of a specific polyfunctional resin, i.e. tetraglycidyl derivative of methylenedianiline, in combination with a specific hardener. This is equally clear from a study of document (1), which mentions a long list of conventional epoxy resins, wherein no cycloaliphatic polyepoxide is quoted (page 3, last paragraph to page 4, paragraph 1). In view of the improved properties demonstrated by the Appellant, the selection of epoxy resins of cycloaliphatic structure, either alone or in admixture with conventional epoxy resins, is a non-obvious choice which contributes to the inventiveness of the solution presently claimed.

- 5.5 For the reasons given above, the subject-matter of Claim 1 must be regarded as involving an inventive step.
6. Claim 1 being allowable, the same applies to dependent Claims 2 to 8, which are directed to preferred embodiments of the use according to Claim 1 and whose inventiveness is supported by that of the main claim.
7. The grounds of opposition being overcome by the claims presently on file and a description suitably adapted having been submitted, the patent can thus be maintained on the basis of these documents.

#### 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 with the order to maintain the patent in suit on the basis of the Claims 1 to 8 and the description submitted during oral proceedings.

The Registrar:

The Chairman:

E. Görgmaier

C. Gérardin