

A		B	X	C	
---	--	---	---	---	--

File Number: T 732/89 - 3.3.3

Application No.: 84 200 106.7

Publication No.: 0 126 494

Title of invention: Impact resistant matrix resins for advanced composites

Classification: C08G 59/50

D E C I S I O N
of 7 October 1992

Proprietor of the patent: AMOCO CORPORATION

Opponent: BASF Aktiengesellschaft

Headword:

EPC Article 56

Keyword: "Inventive step - subsequently demonstrated effect - reformulation of technical problem - affirmed (see points 5.5 to 5.8)."



Europäisches
Patentamt

European
Patent Office

Office européen
des brevets

Beschwerdekammern

Boards of Appeal

Chambres de recours

Case Number : T 732/89 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 7 October 1992

Appellant : AMOCO CORPORATION
(Proprietor of the patent) 200 East Randolph Drive
P.O. Box 5910-A
Chicago
Illinois 60680 (US)

Representative : Weinhold, Peter, Dr.
Patentanwälte Dr. V. Schmied-Kowarzik
Dipl.-Ing. G. Dannenberg
Dr. P. Weinhold, Dr. D. Gudel
Dipl.-Ing. S. Schubert, Dr. P. Barz
Siegfriedstrasse 8
W-8000 München 40 (DE)

Respondent : BASF Aktiengesellschaft
(Opponent) Patentabteilung C6
Carl-Bosch-Strasse 38
W-6700 Ludwigshafen (DE)

Decision under appeal : Decision of the Opposition Division of the
European Patent Office of 21 June 1989, issued on
30 August 1989 revoking European patent
No. 0 126 494 pursuant to Article 102(1) EPC.

Composition of the Board :

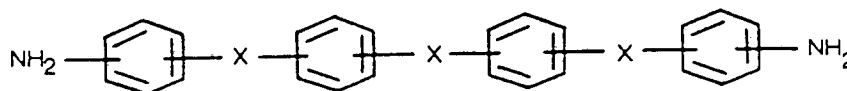
Chairman : F. Antony
Members : R. Young
S. Perryman

Summary of Facts and Submissions

- I. The mention of the grant of European patent No. 0 126 494 in respect of European patent application No. 84 200 106.7 filed on 27 January 1984 and claiming a US priority of 25 May 1983 (US 496 398) was announced on 9 July 1986 (cf. Bulletin 86/28). Independent Claim 1 read as follows:

"A composition comprising:

(a) a diamine hardener represented by the following general formula:



wherein the X's are independently selected from a direct bond, O, S, SO₂, CO, COO, C(CF₃)₂, C(R₁R₂)₂ wherein R₁ and R₂ are independently hydrogen or alkyl of 1 to 4 carbon atoms,

(b) an epoxy resin containing two or more 1,2-epoxy groups per molecule, and

(c) a thermoplastic polymer."

Dependent Claims 2 to 36 related to preferred embodiments of that composition, and Claims 37 to 41 to compositions additionally containing structural fibre, in particular prepreps and composites.

- II. Notice of Opposition was filed on 18 September 1986 on the ground of lack of inventive step, on the basis of inter alia the documents

- (1) JP-A-54 064 599 (Derwent Abstract, later supplemented by a full translation in English); and
- (5) US-A-3 784 433.

III. By a decision which was given at the end of oral proceedings held on 21 June 1989 and issued in writing on 30 August 1989 the Opposition Division revoked the patent on the ground that, on the evidence before it, the subject-matter of Claim 1 did not involve an inventive step.

According to that decision, the compositions disclosed in the closest state of the art document (5) differed from those according to the disputed patent solely in that, instead of diamine (a) containing four linked benzene rings, the hardeners contained only two benzene rings. Moreover it was clear from (1) that diamines of the type (a) of the disputed patent had been known as hardeners for epoxy resins useful in fibre reinforced compositions. The experimental data in the patent specification itself and that supplied by the Patentee failed to form a direct comparison, and in the absence of any surprising effect, it would have been obvious to use the hardeners of (1) as an alternative to those used in (5).

IV. On 30 October 1989 a Notice of Appeal against the above decision was filed, together with payment of the prescribed fee.

In the Grounds of Appeal filed on 29 December 1989, the Appellant (Patentee) argued that the very different requirements placed on resins such as those in (1) which were for use in copper clad printed circuit laminates, and those of the disputed patent intended for use as aircraft parts, as well as the large choice of compositional

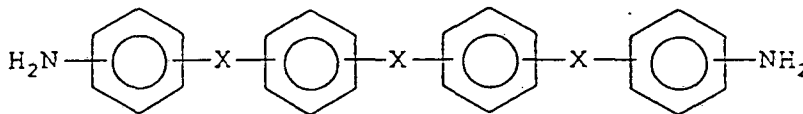
parameters which could be varied by the skilled person meant that it was not obvious to improve one type of resin using a hardener from the other. The effects of improved "flexibility" taught by (1) would in any case lead the skilled person to reject its teaching since flexibility was undesirable in structural applications.

The Appellant filed a new, restricted set of claims together with a submission of 27 March 1991 and new comparative experimental data, intended to prove an unexpected effect. Further amended claims were submitted in August 1992, with supplementary modifications being effected at the oral proceedings held before the Board on 7 October 1992, the final version representing the sole request on the basis of which the patent was to be maintained.

Claim 1 now reads as follows:

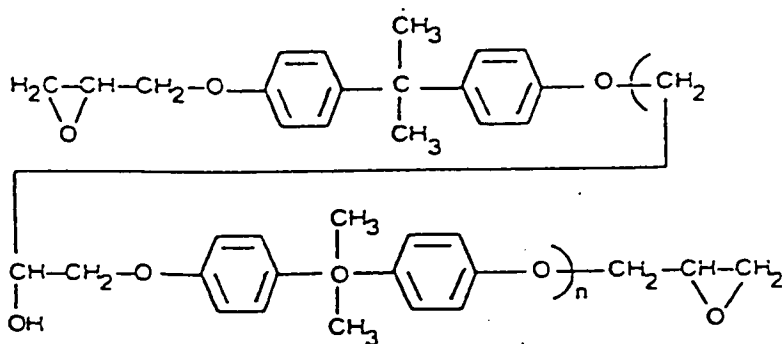
"A composition comprising:

(a) a diamine hardener represented by the following general formula:



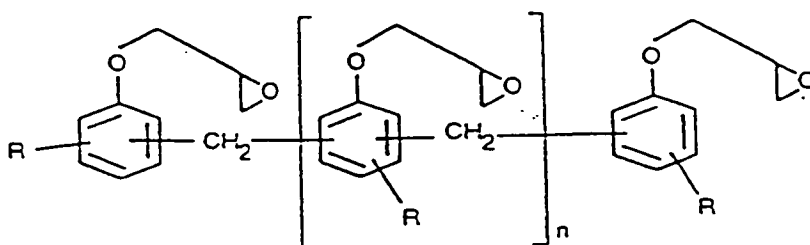
wherein the X's are independently selected from a direct bond, O, SO₂, C(CF₃)₂, S, CO₂, and C(R₁R₂)₂ wherein R₁ and R₂ are independently hydrogen or alkyl of 1 to 4 carbon atoms;

b) 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



II

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



III

where n is 0.1 to 8 and

R is H or CH₃

or with N,N-diglycidyl toluidine or

N,N,N',N'-tetraglycidyl-4,4'-diaminophenyl methane;

c) a thermoplastic selected from one or more of a polysulfone, a polyhydroxyether, a polyether imide a polyarylate, and a poly(ϵ -caprolactone)."

- V. The Respondent (Opponent) on the other hand argued that the resins of (1) were not necessarily restricted in their use to copper laminates, and Claim 1 of the disputed patent was directed to resin compositions per se and not to any particular structural composite or use. As to the teaching of (1), the term "flexible" in the translation of the Japanese original was to be understood as meaning "not brittle". Therefore, the skilled person, starting from (5) and wanting to reduce the brittleness, would in any case have adopted the teaching of (1) and used the claimed

hardeners. Any further effects, such as on water uptake, were consequently merely a "bonus" (not originally emphasised) and were in any case predictable from a consideration of the relative molecular lengths of the different hardeners used. The improved "hot/wet" compressive strength performance of the claimed materials shown by the tests relied upon by the Appellant moreover was not disclosed in the application as originally filed and was therefore not admissible in any reformulation of the technical problem.

- VI. The Appellant requests that the decision under appeal be set aside and that the patent be maintained on the basis of the text finalised at the oral proceedings.

The Respondent requests the dismissal of the appeal.

Reasons for the Decision

1. The appeal is admissible.
2. There are no formal objections under Article 123(2) and (3) EPC since the main request is supported by the original disclosure and manifestly does not extend the protection conferred.

2.1 Present Claim 1 is supported:

- as to the values of X in the hardener component (a), by Claim 1 in the form as originally filed and granted (see also section 2.2 below);
- as to the epoxy resin component (b), by the original description, page 7, line 26 to page 8, line 2, which discloses bis-(2,3-epoxycyclopentyl)ether (hereinafter

abbreviated "cyclo") and its copolymers with ethyleneglycol; furthermore on page 9, line 8 in combination with the last formula on page 4 and the formula on page 5, which discloses mixtures of "cyclo" with bisphenol A epoxy resins and with epoxidized phenol or cresol formaldehyde novolaks respectively; and by the disclosures of Examples 4 and 6 (mixtures of "cyclo" with an N,N-diglycidyl toluidine) and the embodiment on page 15 at lines 11 to 12 (mixtures of "cyclo" with N,N,N',N'-tetraglycidyl-4,4'-diaminodiphenylmethane); and

- as to the thermoplastic resin component (c), by Claims 26-30 as originally filed and granted.

The remaining Claims 2-9 find their basis as follows:

Claim 2 in original and granted Claims 2, 3, 4 and 7; Claims 3, 4 and 5 in original and granted Claims 10, 29 and 26 respectively; Claim 6 in original and granted Claims 33, 34, 35 in combination with the original description on page 12, lines 17 to 22; Claim 7 in original and granted Claim 37 and on page 12, lines 22 to 25; Claims 8 and 9 in original and granted Claims 38 and 40 respectively and Claim 10 on original page 12, lines 2 and 22 to 25.

The description in the patent in suit has been suitably adapted to the revised claims.

2.2 As regards the new level of generality arising through limitations in the definitions of (a), (b) and (c) in Claim 1, the Board is satisfied not only that the limitations taken individually are unobjectionable (cf. section 2.1 above), but also that the smaller number of possibilities embraced by the three groups taken together

does not imply a new invention which, if the granted version of Claim 1 were prior art, could be said to be patentably distinct therefrom. In this connection the meaning of X deleted from the list in component (a) was also absent from the preferred hardeners originally specified in Claims 2 to 9, while the epoxy resins (b) correspond to those originally preferred and exemplified, and the thermoplastic component (c) is derived from the preferred representatives originally set out in Claims 26 to 30.

3. The patent in suit is concerned with matrix resin compositions which may be combined with reinforcing fibres to afford structural composites. The composition according to Claim 1 comprises (a) a specific diamine hardener, (b) a specific epoxy resin, and (c) a specific thermoplastic polymer.

4. Novelty

The claimed subject matter is novel. None of the documents cited discloses a composition of diamine hardener (a), epoxy resin (b) and thermoplastic (c) as claimed. Novelty was in any case not disputed by the Respondent.

5. The Technical Problem

- 5.1 The closest state of the art in the Board's view is represented by document (5).

According to (5), a continuous solid film of a thermosettable resin composition was applied to unidirectional carbon fibres with heat and pressure so that the resin composition flowed about the fibres to form a coherent structure and the resin was converted into its solid, fusible B-stage (cf. Claim 1).

The film was prepared from an epoxide resin, a curing agent therefor and a thermoplastic polysulphone resin which was not heat curable. In examples Ii and Ij, the epoxide resin was a crystallization purified polyglycidyl ether derived from 2,2-bis(4-hydroxyphenyl)propane and epichlorohydrin, and the hardener comprised either bis(4-aminophenyl)methane or bis(4-aminophenyl)sulphone (cf. col. 3, line 6 - col. 4, line 9).

- 5.2 Compared with this state of the art, the technical problem underlying the patent in suit can be stated as being the provision of matrix resins which afford composites with improved stability of mechanical characteristics, especially compressive strength, under conditions of high humidity and elevated temperature, for use in structural applications with exposure to outdoor atmospheric conditions, e.g. in aircraft.
- 5.3 The solution proposed according to Claim 1 was (i) to replace the diamine hardeners of (5), which were characterised by two benzene rings, by a specified range of diamine hardeners (a) having four benzene rings in the molecule, and (ii) to require "cyclo" or a specified copolymer thereof to be present in or as the epoxy resin component (b). As the thermoplastic component (c), a polyhydroxyether, a polyetherimide, a polyarylate or a poly(ϵ -caprolactone) could be used as further alternatives to the polysulphone.
- 5.4 Table I of the experimental data provided by the Appellant in the submission of 27 March 1991 shows that, compared with a Control composition containing a diamine with two phenyl groups (diaminodiphenylsulphone) as the hardener component, an otherwise identical composition in which this hardener is replaced by a diamine hardener having four phenyl groups according to the patent in suit

exhibits a marked reduction in water uptake, and composites prepared therefrom using carbon fibres have a distinctly better compressive strength under "hot/wet" conditions, i.e. after lengthy soaking in hot water (Formulations A and B in comparison with Control Example C). This comparison is a fair comparison in view of the limitation of (b) as indicated in point 5.3 above, resulting in examples Ii and Ij of document (5) no longer presenting the previous high degree of structural approximation.

5.5 The argument of the Respondent that the "hot/wet" performance of the claimed composites, although admittedly better than that of the control, corresponded to a completely new effect which could not be incorporated into the technical problem without contravening Article 123(2) EPC, is not convincing to the Board.

5.6 In the decision T 13/84, (OJ EPO 1986, 253), referred to by the Respondent during the oral proceedings, it is stated that the technical problem may require to be reformulated, since it is established on the basis of objectively established facts, in particular as appearing in the prior art revealed in the course of the proceedings, which may be different from the prior art of which the Applicant was actually aware at the time the application was filed (cf. point 11 of the reasons). Indeed the possibility of such reformulation would appear to be an intrinsic attribute of the problem and solution approach insofar as the problem has to be determined on an objective basis.

5.7 The extent to which such redefinition may be allowed has to be assessed, in the Board's view, on the merits of each particular case. Reference is, however, made in this respect to the decision T 184/82 (OJ EPO 1984, 261) where

the Board allowed a re-definition of the problem regarding the effect of an invention provided that the skilled person "could recognise the same as implied or related to the problem initially suggested" (see point 5 of the reasons).

- 5.8 It is true in the present case that the special test for determining compressive strength under the so-called "hot/wet" conditions was itself apparently developed to become a standard accepted in the aircraft industry after the priority date of the patent in suit (cf. submission of Appellant dated 31 July 1992, referring to the "21st International SAMPE Technical Conference"). Nevertheless, measurements of the crucial parameter of percentage water uptake of the cast (neat) resins after immersion for 2 weeks at 71°C - effectively identical conditions to those of the standard test - as well as a reference to their relatively low water sensitivity compared with many other epoxy castings were originally disclosed (cf. page 21, last paragraph and Table I of the original application).

Furthermore, the quality under examination in the supplementary data - compressive strength - was in any case the same as that originally referred to.

The test for compressive strength under "hot/wet" conditions thus essentially corresponds to the subsequent codification of the originally disclosed conditions of water uptake and the original effect of compressive strength into a standard measure relevant to the circumstances of an originally disclosed use. It cannot therefore be regarded as a new effect unrelated to or not implied by the problem originally suggested.

Nor can the related sub-effect of reduced water uptake of the neat resins, which is clearly inseparable from the

demonstrated effect, therefore be dismissed as a mere "bonus effect".

As this Board has already stated in unpublished decision T 227/89 of 25 September 1991, in paragraph 3.3 of the Reasons for the Decision, "... In determining which effect is crucial and which is merely accidental (so-called "bonus effect"), a realistic approach has to be taken, considering the relative technical and practical importance of those effects in the circumstances of a given case".

It has consequently been considered appropriate to take the demonstrated effect into account in the formulation of the technical problem (see section 5.2 above).

- 5.9 The factual superiority of performance of the tested composites over the Controls was in any case acknowledged by the Respondent at the oral proceedings.

Thus the Board is satisfied that the technical problem is credibly solved.

6. Inventive Step

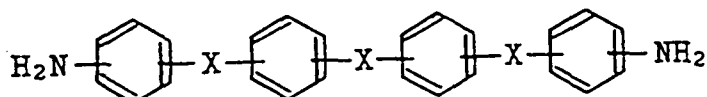
- 6.1 To assess the question of inventive step it is necessary to consider whether the skilled person, starting from (5) would have considered making the combined modifications (i) and (ii) of the solution defined in section 5.3 above, in the expectation of achieving an improved "hot/wet" compression strength in the resulting composites.

- 6.2 There is no reference in (5) to the compression strength of the composites at all, much less any value relating to the particular conditions likely to be encountered in aircraft use. Indeed (5) is not so much concerned with the

stress properties of the composites it discloses, as with the convenience of their manufacture.

Therefore the skilled person could get no hint from the disclosure of (5) of modifications leading to a solution to the technical problem.

6.3 Nor is there any such hint in document (1). According to (1), there is disclosed a thermosetting resin laminate which is to have excellent thermal resistance, flexibility and drilling workability. The laminate is made by impregnating a substrate with a thermosetting varnish consisting of epoxy resin (e.g. bisphenol A diglycidyl ether); a compound of the general formula:



wherein X is selected from O, S, SO₂, CH₂, CO, COO, C(CH₃)₂ and the three X's may be the same or different; and preferably organic solvent; drying to obtain prepreg; laminating prepreps if desired with copper foil; and heating and shaping the laminate under pressure (see Claim).

A comparison between Example 1 and Comparative Example 2 shows that using a binuclear hardener (3,3'-diaminodiphenylsulphone) instead of a 4-nuclear hardener as defined above, gives a product of higher torsional rigidity (about 2x as high).

6.4 Even accepting that the quality referred to as "flexibility" in (1) is in fact "lower brittleness", as canvassed by the Respondent, and is demonstrated by the comparison above using the 4-nuclear hardeners, there is nevertheless no unambiguous pointer in (1) that any

problem having to do with improving compression strength, let alone improving compression strength under "hot/wet" conditions may be solved.

Consequently the skilled person would have had no reason to suppose that the objectively existing technical problem could be solved using the hardeners of (1).

6.5 The remaining documents of the prior art are more remote.

6.6 The allegation of the Respondent at the oral proceedings, that the reduced water uptake of the claimed resins, which is supposedly responsible for the improved "hot/wet" performance, itself arose predictably from the relatively increased molecular lengths between successive hydrophilic -OH bonds when the hardener used had four benzene rings in the molecule rather than two as in the prior art, was disputed by the Appellant and not supported by any factual evidence. The burden of making out such a case is on the Respondent who has not discharged it.

Thus the subject matter of Claim 1 of the Main Request cannot be said to arise in an obvious way from the elements of the prior art.

6.7 On the other hand, the results shown in the supplementary comparative data filed on 27 March 1991 must be regarded as surprising, the more especially so since they show an "inversion", i.e. the compression strength under dry conditions is slightly less good, but under "hot/wet" conditions is considerably better than, the Standard Control.

Thus the subject matter of Claim 1 of the Main Request must be regarded as involving an inventive step.

Since the remaining Claims 2-10 are directly or indirectly dependent on Claim 1, these also by the same token are directed to novel and inventive subject matter.

The description, having been suitably adapted, also meets the requirements of the EPC.

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 following documents:

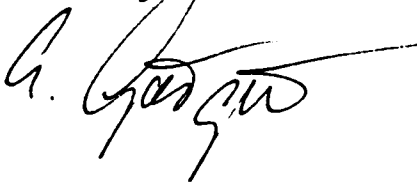
- Claims 1 to 10 submitted during the oral proceedings;

- Description:

pages 1 (title page) and 8 to 11 (ending line 37) as granted;

pages 2 to 7 as submitted during the oral proceedings.

The Registrar:



The Chairman:

