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D E C I S I O N
of 23 November 1995

Case Number: T 0366/91 - 3.3.3

Application Number: 84100716.4

Publication Number: 0130270

IPC: C08G 59/50

Language of the proceedings: EN

Title of invention:
Epoxy compositions containing oligomeric diamine hardeners and high strength composites therefrom

Patentee:
AMOCO CORPORATION

Opponent:
BASF Aktiengesellschaft, Ludwigshafen
Hercules Incorporated

Headword:
-

Relevant legal provisions:
EPC Art. 54, 56

Keyword:
"Novelty (yes) - information available to the public (yes) - disclosure of the invention (no)"
"Inventive step - yes, after change of category of claim"

Decisions cited:
-

Catchword:
-

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Case Number: T 0366/91 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 23 November 1995

Appellant: (Proprietor of the patent)
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Decision under appeal: **Decision of the Opposition Division of the European Patent Office dated 5 March 1991 revoking European patent No. 0 130 270 pursuant to Article 102(1) EPC.**

Composition of the Board:

Chairman: C. Gérardin
Members: H. H. Fessel
W. M. Schar

Summary of Facts and Submissions

I. European patent No. 0 130 270 in respect of European patent application No. 84 100 716.4 filed on 24 January 1984 and claiming a US priority of 30 June 1983 (US 509453) was granted on 29 July 1987 (Bulletin 87/31) to Union Carbide Corporation, Danbury Connecticut 06817 (US) on the basis of a set of 19 claims relating to epoxy compositions containing oligomeric diamine hardeners (Claims 1 to 13) and to prepregs and composites made therefrom (Claims 14 to 19). With effect from 22 June 1988 it was transferred to Amoco Corporation, Chicago Illinois 60680 (US).

II. Notices of Opposition were filed by BASF AG and Hercules INC. on 12 and 25 April 1988, respectively, based on Article 100(a) and (b) EPC. The Opponents requested revocation of the patent in its entirety on the grounds of lack of novelty and inventive step as well as insufficiency of disclosure (Articles 54, 56 and 83 EPC).

The oppositions were inter alia supported by:

D1: Polymer Science USSR vol. 25, No. 6, pp. 1523 to 1529 (1983), translation published by Pergamon Press, May 1984,
D2: US-A-3 895 064,
D8: US-A-4 330 659,
D11: JP Sho 53/98400 (English translation),
D12: JP Sho 54/64599 (English translation),
D14: US-A-3 530 087, and
D18: Communication of U.S.P.O. in respect of US patent application number 182526 together with the set of Claims 1 to 42 of that application.

III. By decision of 15 January 1991 issued in writing on 5 March 1991 the Opposition Division revoked the patent.

(i) It was first stated in the decision that the claims were formally admissible (Article 123(2) and (3) EPC) and that the patent specification contained enough information to be carried out by a person skilled in the art (Article 83 EPC).

(ii) As to novelty, the subject-matter as defined in Claim 1 of the Main Request was held to be anticipated by the teaching given in D18, whereas the subject-matter of independent Claims 12, 14 and 17 was held to be novel over that prior art. These conclusions also applied to the claims according to the Auxiliary Request, wherein the diamine had been defined in a more specific way.

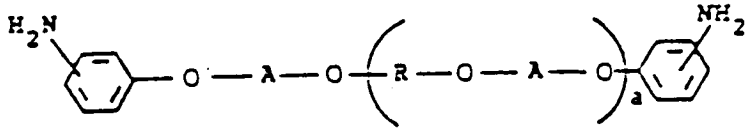
(iii) The subject-matter of Claims 12, 14 and 17 of both requests was considered to be obvious and thus not patentable within the meaning of Article 56 EPC. It was added, however, that a narrower definition of the claimed subject-matter could involve an inventive step.

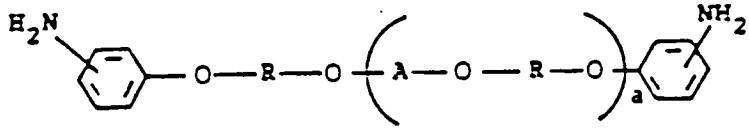
IV. On 3 May 1991 an appeal was lodged against that decision by the Appellants (Patentees) together with payment of the prescribed fee.

(i) Together with the Statement of Grounds of Appeal received on 13 June 1991 the Appellants submitted as sole request a new set of 11 claims directed to the use of the originally claimed compositions, of which Claim 1 reads as follows:

"1. The use of a composition comprising:

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

(i)  or

(ii) 

or mixture thereof, wherein A is the residuum of a dihalobenzenoid compound, R is the residuum of a dihydric phenol and a is 0.05 to 20, and

(b) an epoxy resin containing two or more 1,2-epoxide groups per molecule
as such or in form of prepregs for preparing composites useful as aircraft parts, automotive parts, pressure vessels, tanks, pipes, protective armour on military vehicles and sporting goods."

Dependent Claims 2 to 11 are directed to preferred embodiments of Claim 1.

(ii) In favour of the patentability of these claims the Appellants argued that the reasons given in the decision under appeal no longer applied to the subject-matter of the claims now on file. In particular, D18 was not an information in relation to a technical teaching and, consequently, did not represent a disclosure in accordance with Article 54 EPC, even if the public could have had

access to that communication upon request; as to documents D2, D18, D11 and D12, they did not render obvious the specific uses of the epoxy compositions.

V. In their submissions, on the contrary, both Respondents (Opponents) maintained that D18 had been made available to the public before the date of priority and was thus to be considered as prior art, even if it was not intended to be a technical information. In substance, it was obvious from D14 to use a composition known from D18 for preparing composites useful as aircraft or automotive parts.

Regarding the properties achieved it could be expected from D1 that such composites would show improved strength and chemical resistance, and from D8 that they would show improved water resistance. Moreover it was not surprising that the amount of water absorbed by an epoxy hardened with an amine decreased as the concentration of amino nitrogen atoms decreased.

VI. In a communication dated 18 August 1995 attached to the summons to Oral Proceedings to be held on 23 November 1995, the parties were informed that according to the preliminary opinion of the Board the information given in the US examiner's communication (D18) did not amount to a disclosure of compositions of diamines and polyepoxides, even if that communication could be regarded as prior art document.

VII. The three parties - the Appellants on 18 August 1995, Respondent 1 on 8 August 1995 and Respondent 2 on 17 November 1995 - informed the EPO that they would not attend the oral proceedings.

VIII. The Appellants requested that the decision under appeal be set aside and the patent be maintained on the basis of Claims 1 to 11 as filed on 13 June 1991.

The Respondents requested that the appeal be dismissed.

Reasons for the Decision

1. The appeal is admissible.

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

2.1 The Board confirms the finding of the Opposition Division that the formulae (i) and (ii) can be derived from Claim 1 as filed originally by limiting the meaning of the groups Y and X to oxygen. This limitation does not constitute an inadmissible selection since it is supported by page 3, second paragraph to page 6, first paragraph of the original files (page 3, line 1 to page 4, line 4 of the patent in suit) and by the worked examples.

The wording of Claim 3 corresponds to that of Claim 3 as originally filed and granted, with the exception that three of the original definitions given for B have been deleted.

Apart from the change of category the other claims have the same wording as Claims 2 and 4 to 11 of the application as originally filed and of the patent as granted.

2.2 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 or prepregs for preparing **composites** useful as aircraft 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 Claims 17 to 19 of the patent as granted which corresponded to Claims 17 to 19 as originally filed.

3. Owing to the change of category of Claim 1 the Board is satisfied that the claimed subject-matter is novel over the cited prior art, which has not been disputed in appeal proceedings.

It is, however, necessary to consider the question of novelty of the composition as such with respect to the disclosure of D18, not so much because that citation played a major role in opposition proceedings as well as in the submissions of the Respondents in appeal proceedings, than because that question will be decisive for the relevance of that citation for the issue of inventive step.

3.1 D18 consists of two documents, (a) the text of Claims 1 to 42 of the US patent application Serial No. 182 526, and (b) part of a communication (points 13 and 14) issued during examination of that application. It is undisputed that both were publicly available before the priority date of the patent in suit and that, consequently, they form part of the state of the art within the meaning of Article 54(2) EPC. The statement of the Examiner having been made in a communication issued during examination of the application, the claims of this application and the statement are not separate or independent documents; it was thus correct, as did

the Opposition Division, to consider them in combination as a single disclosure when dealing with the issue of novelty of the compositions as claimed.

3.2 The statement was made in the framework of an objection of lack of unity of invention. The subject-matter as defined in the claims was said to contain in fact two inventions, one concerning polyamide-imides and their method of preparation (Claims 1 to 25), and another concerning amine compounds per se (Claims 26 to 42). These inventions were distinct, since amines were suitable not only to prepare polyamide-imides, but also "as crosslinking agents for polyepoxides and as a reactant in the preparation of polyureas".

3.3 The objection of lack of novelty of the compositions raised in the opposition procedure before the EPO was based on the arguments (i) that these amines, by a proper combination of the structural and compositional parameters in Claims 26, 33 and 39, fell under the definition of the hardeners of the patent in suit, which was not disputed by the Patentees, and (ii) that, consequently, the communication issued by the US Patent Office represented a proper disclosure of compositions within the terms of the patent in suit.

3.4 In the Board's view, however, a combination of the structural and compositional parameters of the diamines specified in Claims 26, 33 and 39 of D18 would not result in a diamine component (a) of the compositions as used in Claim 1 of the patent in suit.

Claim 26 is an independent claim directed to oligomeric diamines defined by a general formula containing many parameters. Claim 39 refers back to Claim 26 and is clearly dependent thereupon, since the value a = 0 required in Claim 39 is a preferred value within the

range a = 0 to 4 specified in Claim 26. However, since Claim 39 does not refer back to Claim 33, there is no basis for the disclosure of a subject-matter resulting from a combination of Claims 26, 33 and 39. Further, although formally related to Claim 26, Claim 33 cannot in substance concern a preferred embodiment of Claim 26, since the requirement in Claim 26 that m = 0 when L has the formula (5) is clearly not compatible with the requirement in Claim 33 that m = 1 when L has the same meaning. Claim 33, thus, deals with compounds not falling under the formula given in Claim 26.

The assumption made in the decision under appeal (point 6 (b)) that this inconsistency was in fact an obvious error, has not been supported by any argument. The reason why a skilled person would interpret Claim 33 in a certain way in order to reconcile the technical features in that claim with the appendancy to Claim 26 has not been substantiated. Moreover, even the argument of an obvious error does not imply that the interpretation followed in the decision under appeal was the only possible solution.

Thus, although it was agreed between the parties that D18 disclosed diamines as used in the patent in suit, this conclusion was not justified on the basis of D18 alone, since this citation does not comprise the description of the whole US application, but only the claims thereof (see point 5.5. hereinafter).

3.5 Moreover, the sole statement that diamines would be suitable as crosslinking agents for polyepoxides cannot be equated with a proper disclosure of a composition comprising an epoxy resin and such diamines. It is nothing more than the general remark that these amines, in addition to other applications in the polymer field,

would have that capability in view of the well known reactivity of amino groups with epoxy groups.

3.6 For these reasons D18 cannot be regarded as an explicit or implicit disclosure of the compositions used in the patent in suit.

4. The patent in suit now concerns the use of particular epoxy compositions for specific applications. The change of category of Claim 1 raises the question of the closest state of the art since, as will appear hereinafter, the documents which played a major role in the discussion of the patentability of the composition in opposition proceedings, in particular D18 and D1, are now much less relevant. In view of the applications listed in Claim 1, the Board takes the view that D8 represents the closest state of the art.

4.1 This citation describes hardeners for epoxy resins which are adducts from diaminodiphenylsulphone and diepoxides, in particular diglycidyl ether of bisphenol A (Claims 1 and 4). The resulting compositions, wherein reinforcing fillers may be incorporated (column 4, lines 33 to 48), exhibit a desirable combination of improved impact resistance, toughness, strength properties as well as high melt flow viscosity properties (column 2, lines 8 to 11 and lines 33 to 42); moreover, these advantageous mechanical properties are substantially maintained even after humidity ageing, which is attributed to increased moisture resistance (column 9, lines 22 to 62). These characteristics make these resin systems suitable as prepreg and laminating resins, in particular in the aerospace industry (column 1, lines 58 to 61; Example III, column 7, lines 1 to 25).

4.2 Since there is no evidence on file that the level of properties achieved with the compositions used in the

patent in suit is any better than in D8, the technical problem underlying the patent in suit has to be seen in the provision of further epoxy resin compositions for preparing composites for the required use.

4.3 According to the patent in suit that problem is solved by using as hardener oligomeric diamines of formula (i) or (ii), as specified in Claim 1.

4.4 In view of the experimental data in the patent in suit, in particular Examples 16 to 25 in conjunction with Table II, Examples 28 and 29, Examples 30 and 31 in conjunction with Table IV, and Example 32 in conjunction with Table V, which show that a balance of properties similar to that reported in D8 is obtained, the Board is satisfied that the above defined technical problem is effectively solved.

5. It remains to be decided whether the use of epoxy compositions as defined in the patent in suit involves an inventive step.

5.1 Although D8 mentions in vague terms that other aromatic polyamines or aralkyl polyamines could be used (column 2, line 67 to column 3, line 3), there is in fact no incentive to operate outside the general teaching of this citation.

First, the use of an aromatic amine which at the same time is a sulphone ensures a better control of the properties of the resins. As specified in column 3, lines 47 to 52, "Concentrations of the sulphone are selected so as to achieve a modified hardener system having from about 2.5 to 100 equivalents of aminohydrogen per equivalent of epoxy. These values are selected into order to avoid gelling at levels lower

than those specified and reduced physical properties at levels higher than those specified."

Secondly, the use of diaminodiphenylsulphone as an adduct must be regarded as an attempt to overcome various shortcomings which had been observed previously when that diamine was used as such as the hardener. As explained in the introduction (column 1, lines 19 to 52), these disadvantages which stemmed primarily from the incompatibility of the diamine with the resin and the poor solubility in it, not only resulted in practical processing difficulties, but also affected some properties of the cured resins. According to D8, both processing characteristics and performance characteristics are improved by using an adduct (column 2, lines 12 to 41).

These considerations show that the above mentioned properties (point 4.1) are closely related to the use of a particular hardener and that, consequently, alternative curing agents which would not combine sulphone groups and adduct structure would entail a loss of properties. It follows that D8 cannot provide any solution to the technical problem.

5.2 Document D14 relates to hardenable compositions containing an epoxide resin, a conventional hardener and a thermoplastic polysulphone resin, which are suitable as structural adhesives in the manufacture of aircraft (column 1, lines 24 to 35). Reinforcing fibres and powdered metals may further be incorporated into these compositions (column 4, lines 49 to 60). The polysulphone resin has to meet specific criteria in terms of average molecular weight and heat deflection temperature (Claims 1 and 2) in order to act as a thickening agent minimising the amount of epoxide resin lost through flowing from the area to be bonded when

pressure is applied (column 4, lines 53 to 68) and to ensure that to a large degree the adhesive strength is retained both at very low and at high temperatures (column 1, lines 56 to 62). By contrast, the curing agent may be any conventional hardener (column 4, lines 21 to 41); even if aromatic polyamines are mentioned among the preferred hardeners for their low reactivity with the epoxy resin in the blending operation and the very high adhesive strength at elevated temperatures of the cured products, there is no indication that oligomeric aromatic diamines would render the polysulphone superfluous.

There would thus be no incentive for a skilled person faced with the above defined technical problem to forego the advantages provided by the thermoplastic polysulphone, thus no incentive to omit that component, let alone choose a type of amine agent not even envisaged in that citation.

5.3 D12 concerns epoxy resin formulations with improved heat resistance and drilling properties which may be used in copper foil laminated board applications (page 2, paragraph 2). These compositions comprise as hardeners aromatic diamines containing four phenylene rings linked by ether bridges, such as di(p-aminophenoxy) diphenyl ether (page 2, paragraphs 4 and 5; page 3, paragraph 2); these diamines may be used together with conventional aromatic diamines, in particular with di(aminophenoxy) benzene. A mixture of these two specific diamines would be very similar to the definition of the hardeners (i) and (ii) in the patent in suit.

Similar epoxy resin formulations are disclosed in D11, wherein aromatic diamines having three or more phenylene rings linked at least partially by ether bridges, such as di(aminophenoxy) diphenyl sulphone (page 2,

paragraph 3), are used. These compositions, which may further contain non-polar type fillers (page 3, paragraph 3), are suitable for use in the insulating parts of electrical parts and semiconductor elements, resin plates for printed circuit boards and heat resistant adhesives (page 4, paragraphs 3).

The properties aimed at in these two citations have nothing to do with the high mechanical properties required for the applications listed in Claim 1 of the patent in suit, so that the skilled person would have no reason to consider higher homologues of the amines described in these two citations for the solution of the technical problem.

5.4 As stated above, D18 does neither describe oligomeric diamines within the terms of the patent in suit, (point 3.4), nor even compositions comprising an epoxy resin and diamines (point 3.5), but at most the suitability of such diamines to cure epoxy resins. In the absence of any mention of the properties likely to be influenced by the use of oligomeric diamines there would be no reason to consider a solution along that line for the solution of the technical problem.

5.5 The same applies to D2, which is a division of Serial No. 182 526, thus of D18. This citation relates to (1) oligomeric aromatic diamines (Claims 1; column 16, line 47 to column 18, line 62) within the terms of the patent in suit, (2) their method of preparation by condensation of an aminophenol, a diphenol and a dihalobenzenoid compound or derivatives thereof (column 9, line 48 to column 10, line 48; Example 2, 3, 7 to 12, 15 and 16), and (3) their use for the preparation of polyamideimides (column 1, lines 9 to 13). Apart from the fact that this method of preparation of the diamines corresponds to the method described in

the patent in suit for the preparation of the oligomeric diamines of general formula (i) (compare patent specifications, page 3, lines 27 to 46), D2 is totally silent about epoxy resins and would thus not be considered by a skilled person.

5.6 The main teaching of D1 is that the properties of polyepoxides cured with oligo-1,5-phenylenesulphide sulphone-α, ω-diamines can be regulated by changing the molecular weight of the hardener. More specifically, it has been found that increasing the distance between the amino end groups on transition from binuclear to tetranuclear diamines is accompanied by an increase in the strength, heat resistance and chemical resistance of polyepoxides (page 1523, Abstract and paragraphs 1 and 2; page 1527, paragraph 3). These effects are explained by the dependence of the glass transition temperature of the cured resins on the content of sulphone groups in the diamine, according to which the glass transition temperature increases together with the proportion of sulphone groups as the result of an increase in chain interaction (page 1527, Table 2; page 1528, Figure 4). This shows that the essential compositional feature of the sulphur containing oligodiamines according to D1 is the presence of the recurring structure {phenylene-SO_2-phenylene-S}, i.e. a combination of sulphone and sulphur linkages (page 1525, formula). By contrast, following the amendment regarding the definition of X and Y in the patent as granted which can now only represent oxygen linkages, the main feature of the oligomeric diamine used in the patent in suit is a sequence of ether bridges.

The Respondents failed to demonstrate why the teaching of D1 disclosed in connection with specific compositional features should apply to the patent in suit where these features, depending upon the definition

of A and R, can at most appear in a limited extent. The improved strength mentioned in D1 in relation with the higher molecular weight of the hardener relied upon by Respondent 1 (Counterstatement of Appeal, page 2, paragraphs 1 to 4) is in any case irrelevant, since the technical problem underlying the patent in suit has been defined in alternative terms with respect to the properties achieved in D8.

In view of these conclusions, the question whether D1, which has a printing date of June 1983, was indeed publicly available before 30 June 1983, is of minor importance and does not thus have to be answered.

5.7 Even the known relationship between the concentration of amine groups in the hardener and the amount of water absorbed by the cured product, which would be an incentive to increase the molecular weight of the diamine (Respondent 2, Counterstatement of Appeal, points 6 and 7), does not render obvious to use diamines of the general formulae (i) and (ii) as defined in the patent in suit.

First, the reference to D11 and D12, for the sole reason that the diamines disclosed there are structurally very close to the diamines (i) and (ii), is not proper since these citations make no reference whatsoever to water resistance properties. The argument that a slight increase of the molecular weight of these amines in order to lower water absorption is thus based on hindsight. Secondly, the solution proposed in the patent in suit does not require an increase of the molecular weight. Rather, a comparison of the molecular weight of the adduct in D8, which represents the closest state of the art, and of the diamines (i) and (ii), although difficult on a general basis because the molar ratio of the reactants used to prepare the adduct is not

specified in D8 and because A and R in the patent in suit may have various meanings, would show there is a large overlap and that either can have the higher molecular weight.

5.8 These considerations show that the subject-matter of Claim 1 does not derive in an obvious manner from the documents relied upon by the Respondents and, therefore, involves an inventive step.

By the same token the subject-matter of dependent Claims 2 to 11, which concern preferred embodiments of Claim 1, also involves an inventive step.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the Opposition Division with the order to maintain the patent on the basis of Claim 1 to 11 as requested and the description yet to be adapted.

The Registrar: The Chairman:

E. Görgmaier C. Gérardin