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BOARDS OF APPEAL OF THE EUROPEAN PATENT OFFICE

CHAMBRES DE RECOURS DE L'OFFICE EUROPEEN DES BREVETS

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File Number: T 198/91 - 3.3.3

Application No.: 84 116 126.8

Publication No.: 0 185 118

Title of invention: Epoxy Resin Composition

Classification: C08G 59/08

D E C I S I O N
of 4 September 1991

Applicant: DAINIPPON INK AND CHEMICALS, INC.

Headword:

EPC Articles 54, 56

Keyword: "Novelty (yes)"
"Inventive step - remittal to the first instance after drastic amendment"

Headnote

Europäisches Patentamt | European Patent Office | Office européen des brevets

Beschwerdekammern | Boards of Appeal | Chambres de recours

Case Number : T 198/91 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 4 September 1991

Appellant : DAINIPPON INK AND CHEMICALS, INC.
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Decision under appeal : **Decision of Examining Division of the European Patent Office dated 12 September 1990 refusing European patent application No. 84 116 126.8 pursuant to Article 97(1) EPC.**

Composition of the Board :

Chairman : F. Antony
Members : C. Gérardin
M. Aúz Castro

Summary of Facts and Submissions

I. European patent application No. 84 116 126.8 filed on 21 December 1984 and published under the publication No. 0 185 118, was rejected by a decision of the Examining Division dated 12 September 1990.

That decision was based for the main request on a Claim 1 essentially directed to a composition drying and curing at room temperature comprising (A) a polyfunctional epoxy resin derived from a low molecular weight epoxy resin and a polyfunctional phenol, and (B) an amine-type curing agent. Claim 1 according to the auxiliary request was directed to the use of an epoxy resin comprising the same components (A) and (B) for producing films drying and curing at room temperature. Both sets of claims, which further contained dependent Claims 2 to 7 directed to preferred embodiments of the main claims, had been filed on 22 May 1989.

II. The main ground for that decision was lack of novelty with regard to the teaching of DE-B-2 521 813 (document (1)), which described the curing with dicyandiamide of an "advanced" epoxy resin identical with component (A) in Claim 1 according to the application. All the compositional features being the same, the property of drying and curing at room temperature could not be regarded as a distinguishing feature, but merely as a result to be achieved. These considerations applied to the composition as well as to the use thereof and, therefore, to the main request as well as to the auxiliary request.

Should, however, the property of drying and curing at room temperature be a distinguishing feature conferring novelty to the claimed subject-matter, the issue of inventive step would boil down to finding a curing agent efficient at

room temperature. From the Handbook of Epoxy Resins, Lee and Neville (1967), pages 7-1 to 7-3 (document (4)), it was readily apparent that primary aliphatic amines would be appropriate for that purpose.

III. On 12 November 1990 a Notice of Appeal was lodged against that decision with payment of the prescribed fee. In the Statement of Grounds of Appeal filed on 15 January 1991 as well as in the later submission of 20 August 1991 the Appellant maintained that the composition according to document (1), which comprised dicyandiamide as a latent curing agent, could not anticipate a composition drying and curing at room temperature. He further argued that, although primary aliphatic amines were generally known from document (4) to cure epoxy resins at room temperature, this did not warrant their suitability for any epoxy resin, as evident from Comparative Examples 1 to 6 in the application in suit.

In accordance with these arguments, the Appellant filed together with that last submission a set of six claims to be considered as main request, of which Claim 1 reads as follows:

"A method for coating a substrate with a coating film of an epoxy resin composition having excellent handling properties, mechanical properties, heat resistance, corrosion resistance and particularly excellent low-temperature curability, said epoxy resin composition comprising as essential ingredients

(A) a polyfunctional epoxy resin having an epoxy equivalent of 185 to 1,500 and a molecular weight of 250 to 4,000 obtained by reacting

(a-1) an epoxy resin having an epoxy equivalent of 100 to 800 and a molecular weight of 200 to 2,000 as well as containing about 2 glycidyl ether groups derived form a diphenol compound having difunctional phenolic hydroxyl groups per molecule, with

(a-2) a molecular weight increasing agent in form of a polyhydric phenol, and

(B) a curing agent for the polyfunctional epoxy resin (A),

characterized by the whole combination of the following features:

- There is used a molecular weight increasing agent (a-2) containing 2.1 to 10 phenolic hydroxyl groups per molecule and having a molecular weight of 200 to 2,000;

- the portions of the epoxy resin (a-1) and of the molecular weight increasing agent (a-2) are selected such that the number of the phenolic hydroxyl groups in the molecular weight increasing agent (a-2) is less than 1 per epoxy group in the epoxy resin (a-1);

- there is used a polyfunctional epoxy resin (A) which does not gel;

- as the curing agent (B) for the polyfunctional epoxy resin (A) there is used an amine-type curing agent; and

- the drying and curing of the epoxy resin composition is carried out at room temperature."

Claims 2 to 6 are dependent claims directed to preferred embodiments of the main claim.

IV. During oral proceedings held on 4 September 1991 the Appellant maintained his previous position and argued additionally along the line of a selection invention. In that sense he filed a set of five claims to be considered as auxiliary request; in Claim 1 thereof the part following the definition of (A) as in Claim 1 according to the main request reads as follows:

"(B) a curing agent for the polyfunctional epoxy resin (A),

wherein

- there is used a molecular weight increasing agent (a-2) containing 2.1 to 10 phenolic hydroxyl groups per molecule and having a molecular weight of 200 to 2,000;

- the portions of the epoxy resin (a-1) and of the molecular weight increasing agent (a-2) are selected such that the number of the phenolic hydroxyl groups in the molecular weight increasing agent (a-2) is less than 1 per epoxy group in the epoxy resin (a-1);

- as the curing agent (B) for the polyfunctional epoxy resin (A) there is used an amine-type curing agent;

characterized in that

- there is used a polyfunctional epoxy resin (A) which does not gel;

- the drying and curing of the epoxy resin composition is carried out at room temperature; and

- as the curing agent (B) there is used a preliminary co-condensation product of triethylenetetramine and "EPICLON 1050-75X" being an epoxy resin with an epoxy equivalent of 475."

Claims 2 to 5 are dependent claims directed to preferred embodiments of the main claim.

V. The Appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of Claims 1 to 6 and an adapted description, both filed on 20 August 1991, as main request, or on the basis of Claims 1 to 5 filed during oral proceedings and a description yet to be adapted as auxiliary request.

Reasons for the Decision

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

Main Request

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

Besides the fact that the valid Claim 1 is drafted as a method claim, the main claim differs from original Claim 1 drafted as a composition claim by the introduction of additional descriptive features and by a more specific

definition of the various reactants. The addition of mechanical properties and corrosion resistance in the preamble is supported by the general reference to these properties on page 15, lines 2/3, as well as by the specific tests carried out for comparative purposes, namely moisture resistance test, salt spray test, pencil hardness test and walkable test, defined on page 15, line 27 to page 16, line 14, and reported in Table 2, pages 17 to 19 and Table 6, page 24, and further by the values of impact strength indicated in Table 7, page 28. The values of the epoxy equivalent of component (A) correspond to the more preferred range originally disclosed on page 5, line 27. The definitions of the epoxy resin (a-1) and of the molecular weight increasing agent (a-2), the relative amounts of these two reactants as well as the requirement that their reaction product does not gel, are to be found in original Claim 6. Last, the use of an amine-type curing agent is supported by the original disclosure on page 8, line 25.

As to the dependent Claims 2 to 6, they correspond respectively to Claims 2 to 4, to the preferred ranges in Claim 5, and to Claim 7 as filed originally, but drafted as method claims.

3. The application in suit concerns a method for coating a substrate with a coating film of an epoxy resin composition. The use of such compositions for that purpose is disclosed in document (1), which the Board, like the Examining Division, regards as the closest state of the art. That citation describes the preparation of advanced epoxy resins derived from a low molecular weight epoxy resin and a polyfunctional phenol (Claim 1). The low molecular weight epoxy resin may be a diglycidyl ether having an epoxy equivalent of 170 to 1000 obtained by

reacting epichlorohydrin with a diphenol (Claim 3 and column 9, lines 56 to 63). The polyphenolic compound is typically a novolak resin containing 3.6 or 4.4 phenolic hydroxyl groups per molecule and having molecular weights of 369 or 433 (column 11, line 30 to column 12, line 43). The relative amounts of diglycidyl ether and novolak resin are such that the resulting advanced epoxy resin has an epoxy equivalent of 200 to 5000 (column 6, lines 56 to 58; Examples 1 to 11). These advanced epoxy resins are subsequently hardened with dicyandiamide, whereby coatings with outstanding mechanical properties and resistance to chemicals are obtained (Example 11, column 15, lines 41 to 52; column 16, lines 10 to 22 and 34 to 41). However, this requires the reactive mixture to be heated to temperatures between 100 and 200°C, which represents a serious limitation of the range of application of such epoxy resin - novolak resin formulations.

In the light of this prior art shortcoming, the technical problem underlying the application in suit can thus be seen in providing a low temperature curing epoxy composition, without impairing the above-mentioned desirable combination of properties of the coating produced therefrom.

According to the main request of the application in suit this problem is solved by (i) using an advanced epoxy resin which does not gel, (ii) selecting an amine-type curing agent, and (iii) carrying out the drying and curing of the epoxy resin composition at room temperature.

In view of the experimental results in the application in suit, in particular the data in Tables 2 to 4 which show acceptable results in terms of low-temperature curability, hardness and corrosion resistance, the Board is satisfied

that the above-defined technical problem is effectively solved.

4. The first question to examine is whether the features (i), (ii) and (iii), in isolation or in combination, confer novelty to the claimed subject-matter with regard to the teaching of document (1).

4.1 Feature (i), which expresses the requirement that the advanced epoxy resin (A) should not gel, cannot be regarded as a distinguishing feature over the disclosure of that citation.

As stated above in point 3, the process described in document (1) aims at the preparation of so-called advanced epoxy resins, i.e. epoxy resins which are still cross-linkable through epoxy groups (column 6, lines 56 to 58); this is achieved by reacting a low molecular weight epoxy resin and a novolak resin, the former being used in molar excess (column 9, lines 23 to 55; Examples 1 to 11). From the general definition of these two reactants, it clearly appears that they correspond to a large extent to the components (a-1) and (a-2) according to the application in suit, and that, consequently, the same applies to their reaction products, as evident from the epoxy equivalent thereof (200 to 5000), which encompasses practically the whole range specified in Claim 1 (160 to 2000). In view of these extensive overlaps, it can reasonably be assumed that the known advanced epoxy resins do not gel and that, therefore, the condition expressed under feature (i) must be implicitly fulfilled in document (1).

4.2 Nor can the choice of an amine-type compound as curing agent according to feature (ii) be regarded as a distinguishing feature. Although dicyandiamide has been deleted from the list of suitable curing agents in the

amended description filed on 20 August 1991 (page 8, line 22 to page 9, line 2), the fact remains that this particular compound is generally classified as an amine-type curing agent for epoxy resins, falling thus within the terms of Claim 1. This is consistent with the complex structure thereof, containing both amino and imino hydrogen atoms, which all play an active role in the curing process, as well as with the type of nitrogen containing compounds which are produced upon curing, namely N-alkylcyanoguanidines and, after rearrangement, guanylureas. It follows that, in default of a more specific definition of the curing agent (B), all the compositional features according to Claim 1 are known from document (1).

4.3 By contrast, the curing temperature is different from that mentioned in document (1), since according to the application in suit that final step must be carried out at room temperature, whereas according to the prior art teaching it is performed between 100 and 200°C. This means that in the framework of the current method claim novelty of the claimed subject-matter can be acknowledged on the basis of feature (iii).

4.4 However, this raises the question of the proper interpretation of that subject-matter, since a composition within the terms of document (1), meeting thus all the compositional requirements specified in Claim 1 of the application in suit, does obviously not cure at room temperature. In the absence of a positive definition of the curing agent (B) in Claim 1, the Board takes the view that the wording of that claim should be regarded as a functional definition insofar as the hardener is concerned; in other words, the curing agent can be any amine-type curing agent provided it exhibits sufficient

reactivity with epoxy groups, thereby ensuring adequate curing of the coating composition at room temperature.

5. Such general solution to the above-defined technical problem, however, does not involve an inventive step.

5.1 The main reason is that, in practice, the Appellant's contribution boils down to finding amino compounds (B) having the desired reactivity at room temperature and to carrying out a preliminary experiment to test the behaviour of that hardener in combination with an advanced epoxy resin (A). As pointed out by the Board during oral proceedings, document (4) provides all the information necessary for that purpose.

It is specified on page 7-2 of that citation that primary aliphatic amines give good cures at room temperature with excellent all-round properties and temperature resistance to 100°C (paragraph 2). Moreover, the epoxy compositions cured with such amines have high solvent and water resistance (paragraph 6), which are among the essential properties aimed at in the application in suit. Triethylenetetramine is exemplified on page 7-3 as particularly suitable; further, alicyclic primary amines, such as isophoronediamine, are mentioned on page 7-3, and aromatic primary amines, such as diaminodiphenylmethane, on page 8-1; all these amines are envisaged in the application in suit (cf. page 8, lines 28 to 33).

5.2 The argument that the structure of the advanced epoxy resin (A) should be regarded as a selection contributing to the inventiveness of the solution cannot be accepted.

In the Board's view, there can be no question of a selection since the main teaching of document (1) is directed to the preparation and the use of an advanced

epoxy resin corresponding practically to component (A) according to the application in suit. As it appears from the definition of the technical problem underlying the application in suit, which takes that teaching duly into account, the curing temperature should be lowered, but the general properties of the coating should not be impaired. There is thus no reason to depart from the advanced epoxy resins disclosed in document (1).

By the same token, the fact that the advanced epoxy resins according to the application in suit (Examples 1 to 18) have a better low-temperature curability than conventional diglycidyl ethers (Comparative Examples 1 to 5) cannot be regarded as surprising, since this finding merely confirms the experimental results reported in document (1) (column 15, lines 60 to 68; column 16, Table). These data clearly show the higher reactivity towards dicyandiamide of epoxy resins which have been modified with novolak resins.

5.3 In conclusion, for the reasons given above, the subject-matter of Claim 1 according to the main request does not involve an inventive step.

6. Claim 1 not being allowable, the same applies to dependent Claims 2 to 6, which are directed to preferred embodiments of the subject-matter of the main claim and thus fall with it.

Auxiliary Request

7. The current wording of the claims is adequately supported by the application as originally filed.

With regard to Claim 1 according to the main request Claim 1 according to the auxiliary request differs by the fact that (i) three features - the definition of the novolak resin (a-2), the relative amounts of the low molecular weight epoxy resin (a-1) and of the novolak resin (a-2), and the amine-type curing agent (B) in general - have been shifted from the characterising part into the preamble of the claims, and (ii) this curing agent (B) is now defined in the characterising part as being a preliminary co-condensation product of triethylenetetramine and "EPICLON 1050-75X", the latter being an epoxy resin with an epoxy equivalent of 475. The amendments (i) have obviously no impact on the scope of the claim. As to the definition of the specific hardener (ii), it is supported by reference to the co-condensation products of amine compounds and epoxy resins originally disclosed on page 8, line 36 to page 9, line 2, and by the specific adduct of 100g of triethylenetetramine and 75g of "EPICLON 1050-75" according to Referential Example 11, which adduct is subsequently used in Examples 1 to 18 (page 14, lines 24 to 30).

As to the dependent Claims 2 to 5, they correspond to Claims 2 to 5 according to the main request and are thus acceptable for the same reasons.

8. Since novelty can be acknowledged on the sole basis of the curing temperature in the case of the main request, the same applies in the case of the auxiliary request, wherein the claimed subject-matter is additionally characterised by the use of a specific hardener which is not mentioned in document (1).

9. Although co-condensation products of aliphatic amines and low molecular weight epoxy resins are well-known curing agents for epoxy resins, it remains to be examined whether

the use of the specific adduct of triethylenetetramine and "EPICLON 1050-75X" in combination with an advanced epoxy resin (A) involves an inventive step. As this question could not yet be examined by the first instance, there being no corresponding claim before it, the Board considers it appropriate, in exercising the discretion conferred to it by Article 111(1) EPC, to remit the case to the first instance for further prosecution.

In that respect, the Board notes inconsistencies in the ranges defining the epoxy equivalent and the molecular weight of the polyfunctional epoxy resin in Claim 5 with regard to the definitions given in Claim 1.

Order

For these reasons, it is decided that:

1. The decision under appeal is set aside.

2. The main request is rejected.

3. The case is remitted to the first instance for further examination on the basis of the auxiliary request.

The Registrar: The Chairman:

E. Görgmaier F. Antony