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D E C I S I O N
of 17 March 1997

Case Number: T 1006/93 - 3.3.3

Application Number: 89101672.7

Publication Number: 0327039

IPC: C08G 59/18

Language of the proceedings: EN

Title of invention:
Barrier coatings

Applicant:
PPG INDUSTRIES, INC.

Opponent:
-

Headword:
-

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

Keyword:
"Novelty (yes) - thermoset resin distinguished from hardener therefor"
"Inventive step (yes) - cited prior art unrelated to field of the invention"

Decisions cited:
T 0799/92

Catchword:
-

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Case Number: T 1006/93 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 17 March 1997

Appellant: PPG INDUSTRIES, INC.
One PPG Place
Pittsburgh, Pennsylvania 15272 (US)

Representative: Sternagel, Hans-Günther, Dr.
Patentanwälte Dr. Michael Hann
Dr. H. -G. Sternagel
Sander Aue 30
D-51465 Bergisch Gladbach (DE)

Decision under appeal: Decision of the Examining Division of the European Patent Office posted 9 September 1993 refusing European patent application No. 89 101 672.7 pursuant to Article 97(1) EPC.

Composition of the Board:

Chairman: C. Gérardin
Members: P. Kitzmantel
J. A. Stephens-Ofner

Summary of Facts and Submissions

I. This appeal, which was filed on 4 September 1993, lies against the decision of the Examining Division dated 9 July 1993, refusing European patent application No. 89 101 672.7 in the name of PPG Industries, Inc. filed on 1 February 1989, claiming the priority of 4 February 1988 from a US-application, and published under No. 327 039. The appeal fee was paid on 2 September 1993 and the Statement of Grounds of Appeal was filed on 15 November 1993.

II. The decision under appeal was based on a set of 22 claims filed on 8 December 1992, independent Claims 1, 2, 13, 14 and 20 reading as follows:

"1. A thermoset polymeric barrier material comprising a reaction product of (i) a polyamine and (ii) a polyepoxide, the polymeric barrier material containing at least about seven percent by weight amine nitrogen, based on the total weight of the polyamine-polyepoxide reaction product."

"2. The barrier material of claim 1 wherein the polyamine (i) is replaced by an ungelled amine-functional polymeric resin (iii) which is a reaction product of a polyamine and a polyepoxide, the ungelled amine-functional polymeric resin being further characterized as containing an average of greater than two polyamine moieties per molecule within said resin."

"13. A coating composition comprising the thermosetting components (i) and (ii) or (ii) and (iii), respectively, of any of claims 1 to 12 in a proportion to yield, after reaction, a thermoset barrier material containing at least about seven percent by weight amine nitrogen, based on the total of the said components."

"14. A multilayer packaging material comprising

(a) at least one layer of a gaspermeable polymeric material; and

(b) at least one layer of a thermoset polymer barrier material of any one of claims 1 to 12."

"20. A container formed from the multilayer packaging material of any of claims 14 to 19."

Claims 2 to 12 were dependent upon Claims 1 and/or 2; Claims 15 to 19 were dependent upon Claim 14; Claims 21 and 22 were dependent upon Claim 20.

III. The decision under appeal held that the subject-matter of Claim 1 of the application in suit was not novel over the respective disclosures in documents

D1: EP-A-177 444 and
D2: DE-B-1 912 485,

because therein polyamine/glycidyl ether adducts suitable as hardeners for polyepoxides were described, which had an amine nitrogen content in excess of seven percent. The decision held furthermore that the subject-matter of Claims 2, 3, 5, 6 and 12 was also anticipated by D1 and/or D2.

The decision also held that the features of the subject-matter of Claims 4 and 7 to 10 could not contribute to an inventive step because they were either known in the art or regarded as trivial.

Apart from these substantial objections, the appealed decision questioned the compliance of Claim 1 with the requirement of Article 123(2) EPC, but stated that this was not a ground on which the decision was based.

IV. In his Statement of Grounds of Appeal the Appellant essentially defended the subject-matter of the claims that had been refused. The Rapporteur, in the course of various contacts with the Appellant's Representative, expressed doubts concerning the compliance of Claim 1 with the requirement of Article 123(2) EPC. In response to a communication of the Rapporteur dated 21 February 1997, in which the latter had commented on four alternative versions of an amended Claim 1, that had been submitted by the Appellant by letter of 3 December 1996, the Appellant, by facsimile dated 4 March 1997, submitted an amended set of claims. After a further intervention of the Rapporteur (communication of 6 March 1997) the Appellant filed on 7 March 1997 the final set of Claims 1 to 22 on which this decision is based.

Independent Claim 1 of this set reads as follows:

"1. A thermoset polymeric barrier material obtainable by reacting (i) an ungelled amine-functional polymeric resin, which is the reaction product of a polyamine and a polyfunctional material selected from polyepoxides, polyacrylates and polyoxalates having at least two functional groups reactive with said polyamine, the ungelled amine-functional resin being further characterized as containing an average of greater than two separate polyamine portions per molecule within said resin; and (ii) a polyepoxide having a 1,2-epoxy equivalency greater than 1.0; said thermoset polymeric barrier material containing at least seven percent by weight amine nitrogen, based on the total weight of components (i) and (ii)."

Claim 2 is dependent upon Claim 1 and is directed to a preferred barrier material derived from a specific polymeric resin (i). The further Claims 3 to 22 essentially correspond to Claims 3 to 22 on which the appealed decision was based.

V. The Appellant's reasoning concerning the issue of novelty of the subject-matter of Claim 1 essentially concentrated on the fact that this claim was directed to a **thermoset** polymeric barrier material, while the adducts disclosed both in D1 and D2, on which the decision under appeal relied, were **not** thermoset, but were only parts of a reactive mixture for the preparation of a thermoset material. It was thus without consequence that these adducts had an amine nitrogen content of above seven percent, because the amine nitrogen content of the thermoset polymeric materials prepared from these adducts was below seven percent. In particular the amine nitrogen content of the thermoset material resulting from Formulation A of Example 5 of D1, although making use of the adduct of Example 1 having 10.4% by weight amine nitrogen, was only 2,9% by weight, and the amine nitrogen content of the thermoset material prepared according to Example 1 of D2, although making use of Adduct A having 11,1% by weight amine nitrogen, was only 3,0% by weight.

With respect to the issue of inventive step the Appellant pointed to the test report filed by letter dated 7 December 1992 which was received on 8 December 1992 (the date of 6 December 1992 mentioned on page 8, line 5 of the Statement of Grounds of Appeal obviously referred to the date of the "additional response"). That report showed that the barrier material according to the application in suit exhibited a reduced permeation of gases, as compared with materials having a lower amine content. Since the state

of the art did not suggest barrier materials having an amine nitrogen content of at least seven percent by weight, the skilled person could not have been aware of their beneficial properties. Hence the subject-matter of Claim 1 was non-obvious. The same conclusion applied to Claims 13, 14 and 20 whose subject-matter comprised the material of Claim 1.

VI. The Appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of the set of claims submitted by facsimile dated 7 March 1997.

As an auxiliary request the Appellant asked for oral proceeding.

Reasons for the Decision

1. The appeal is admissible.

2. *Amendments (Article 123(2) EPC)*

2.1 The various features of Claim 1 are based on the following disclosures in the original application:

"thermoset polymeric barrier material ":	page 1, line 2 to 6; Claim 1;
"obtainable by reacting (i) an ungelled amine-functional polymeric resin, which is the reaction product of a polyamine":	Claim 1, component (b);

"polyfunctional material selected from polyepoxides, polyacrylates and polyoxalates having at least two functional groups reactive with said polyamine":	page 10, lines 12 to 22;
"the ungelled amine-functional resin being further characterized as containing an average of greater than two separate polyamine portions per molecule within said resin":	page 13, lines 26 to 32;
"(ii) a polyepoxide having a 1,2-epoxy equivalency greater than 1.0":	Claim 1, component (a);
"said thermoset polymeric barrier material containing at least seven percent by weight amine nitrogen, based on the total weight of components(i) and (ii)":	page 10, lines 22 to 26; Claim 1.

2.2 Claims 2, 3, 4, 6, 8, 9, 14, 16, 17, 18, 19, 21 and 22 are based, in this sequence, on original Claims 1, 5, 5, 6, 2, 3, 15, 16, 16, 16, 18, 21 and 22.

2.3 The basis in the original application for Claims 5, 7, 10, 11, 12, 13, 15 and 20 is as follows:

Claim 5: page 4, last paragraph to page 6, line 16,
Claim 7: page 18, lines 15 to 25,
Claim 10: page 5, lines 24 to 28,

Claim 11: page 2a, lines 1 to 7,
Claim 12: page 10, lines 25 to 26,
Claim 13: page 14, line 25 to page 15, line 16,
Claim 15: page 9, lines 23 to 28,
Claim 20: page 18, lines 30 to 32.

2.4 The requirement of Article 123(2) EPC is therefore complied with by all claims.

3. *Novelty (Article 54 EPC)*

3.1 Document D1

This document relates to adducts obtained by the reaction of an excess of diethyltoluene diamine with a polyepoxide which are to be used as components of hardener compositions for epoxy and urethane resins (Claim 1; Abstract).

Example 1 discloses the preparation of 800g of an adduct of 531g diethyltoluene diamine (MW 178) and 269g diglycidyl ether of bisphenol A. The nitrogen content of this adduct is 10,4% [(28 x 531 x 100)/(178 x 800)].

According to Example 5, formulation A (see Table Ia on page 16) 45 parts of the above adduct (containing 4,7 parts nitrogen), 50 parts of triethylenetetramine (MW 146; 56 x 50/146 = 19,2 parts nitrogen) and 5 parts of 1-methylimidazole (tertiary amine catalyst not reactive with epoxy groups) are mixed to form 100 parts of hardener composition comprising 23,9 parts amine nitrogen (4,7 + 19,2). 100 parts epoxy resin are then reacted with 13 parts of this hardener composition to a **thermoset polymer having 2,75 wt.-% amine nitrogen** (13 x 23,9/113).

The amine nitrogen content of the thermoset resins prepared according to the further formulations B to G set out in Table Ia is below the above-mentioned value for formulation A.

Fields of application which are mentioned in D1 for these thermoset resins are heat curable laminates, adhesives, coatings, filament winding, surface protection, uses in the electrical and the building industry (page 3, line 16; page 8, lines 1 to 3; page 10, lines 11 to 13).

3.2 Document D2

This document relates to hardeners for polyepoxides which comprise an adduct of two moles ethylenediamine, diethylenetriamine or hexamethylenediamine and one mole glycidylether of bisphenol A having an epoxy equivalent weight of 190, respectively 250 (Claim 1).

According to column 5, lines 35 to 43 an adduct A is prepared from 1200g (605g thereof are recovered after the reaction) ethylenediamine (MW 60) and 1900g polyglycidylether of bisphenol A. The amine nitrogen content of this adduct A is 11,1% [(1200 - 605) x 28 x 100/(60 x 2495)].

According to Example 1 (column 5, lines 45 to 68) 400g of the above adduct A (containing 44,4g nitrogen), 425g 2,2,4-trimethylhexamethylenediamine (MW 158; 28 x 425/158 = 75,3g nitrogen) and 175g phenol are mixed to form 1000g of a hardener composition comprising 119,7g amine nitrogen (44,4 + 75,3).

64,3g (containing 7,7g nitrogen) of this hardener composition are then mixed with 190g of a polyglycidylether of bisphenol A and the mixture is heat cured to a **thermoset polymer having an amine nitrogen content of 3,0%** [(7,7 x 100)/(64,3 + 190)].

The amine nitrogen content of the thermoset resins prepared according to the further examples of D2 is in the same order of magnitude.

According to column 5, lines 5 to 24 the thermoset resins prepared according to D2 have an excellent resistance against water, acids and chemical agents and provide good surface gloss and good elasticity. They may be used for the production of mouldings of large volume, for laminates, adhesives, putties and cements, as materials for coatings and linings (e.g. for surface and corrosion protection), for repairing concrete floors and concrete tubings, as well as for caulking and insulation purposes.

3.3 It follows from the above analysis of documents D1 and D2 that the amine nitrogen content of the **thermoset** resins disclosed therein is considerably below the limit of at least 7 wt.-% required in Claim 1 of the application in suit. Consequently, the subject-matter of this claim is novel over documents D1 and D2.

The same conclusion applies to the subject-matter of independent Claim 13, which relates to a coating composition comprising components (i) and (ii) of Claim 1 in a proportion to yield, after reaction, a thermoset material containing at least seven percent by weight amine nitrogen.

The subject-matter of the further independent Claims 14 and 20 is also novel over documents D1 and D2 because they relate to multilayer articles, comprising at least one layer of a thermoset polymeric material according to Claim 1.

4. *Inventive step (Article 56 EPC)*

4.1 Field of the invention

The application in suit relates to thermoset barrier materials which are prepared from thermosetting compositions, possibly in the form of coatings, which are used for packaging materials and containers (original application: Claim 1; page 1, lines 3 to 6). It is apparent from the application (see the following points 4.2.1 and 4.3.2) that by "barrier" materials such materials are meant, which have low permeabilities for oxygen and carbon dioxide.

4.2 Relevant state of the art

4.2.1 According to what is set out as "Background of the Invention" on page 1 of the original application, plastics materials have become increasingly important as packaging materials for many foods and beverages. Because the gas-barrier properties of common packaging plastics, such as polyolefins and polycarbonates, did not provide sufficient protection from oxygen and/or could not prevent the loss of carbon dioxide, specially designed gas-barrier polymers have been developed. Among those were polymers based on vinylidene chloride and on ethylene-vinyl alcohol. Since each of these materials still had certain drawbacks, there was a need for further gas-barrier polymers.

4.2.2 Documents D1 and D2, although disclosing thermoset polymers which structurally bear some resemblance with the polymers according to Claim 1 of the present application, do not contain the slightest hint at properties or uses of the respective materials which could qualify them as relevant prior art for someone interested in the development of gas-barrier properties. Therefore, these documents cannot contribute anything to the assessment of the problem underlying the subject-matter of the present application or to the solution of that problem, as set out in decision T 0799/92 of 4 July 1996, unpublished in OJ EPO (cf. reasons 4.4 and 4.5).

4.3 Problem and solution

4.3.1 The state of the art to be considered as starting point for the skilled person is thus the one acknowledged in the application itself (see point 4.2.1 supra). In view of this prior art the problem to be solved by the present invention was the provision of **alternative** barrier polymers having good barrier properties for oxygen and carbon dioxide.

4.3.2 From the results set out in Table 1 (page 26) of the application in suit - from which those submitted by the Appellant with his submission dated 7 December 1992 and referred to in these proceedings (cf. point V supra) represent an extract - it can be concluded that the above-stated problem has been solved by the provision of the thermoset polymeric barrier materials according to Claim 1.

4.3.2.1 The materials whose oxygen and carbon dioxide permeabilities are reported in this table are films of polypropylene which comprise a coating prepared by reaction of EPON 828 (diglycicyl ether of bisphenol A: cf. page 20, lines 21 and 22) with adducts of tetraethylenepentamine and EPON 828 (cf. pages 21 to 25: Examples G and J in combination with Examples 1A to 1E).

4.3.2.2 Table 1 essentially shows that with increasing amine nitrogen content of the coating (Example 1A: 4,65% to Example 1D: 10,9%) the oxygen and the carbon dioxide permeabilities of the materials decrease to very low values (from 3.1 and 13.9 cc-mil/100 in^2-day-atmosphere, respectively, to 0,5 and 0,0 cc-mil/100 in^2-day-atmosphere, respectively). For the material of Example 1E, whose coating has an amine nitrogen content of 9,0%, an oxygen permeability of 0,1 cc-mil/100 in^2-day-atmosphere is reported, which is still lower than that reported for Example 1D, whereas its carbon dioxide permeability is a little bit higher (0,2 cc-mil/100 in^2-day-atmosphere).

4.3.2.3 As compared thereto the oxygen permeabilities of uncoated polypropylene films and such polypropylene films which are coated with the reaction product of EPON 828 with a "commercially available epoxy-amine adduct" (cf. Comparative Example 2, page 25) are reported to be 155 and 5,5 cc-mil/100 in^2-day-atmosphere, respectively.

4.3.2.4 The materials for which the best oxygen and carbon dioxide permeability results are reported are, thus, those which have a coating whose amine nitrogen content ranges from 9,0 to 10,9%. These coatings comprise barrier materials that are within the scope of Claim 1 of the application in suit.

4.4 Obviousness

The state of the art which is on file is silent about and does not suggest barrier polymers which are based on polyamine/polyepoxide reaction products. Moreover, this state of the art does not provide any clue that the oxygen and carbon dioxide permeabilities of such polymers could be minimized by increasing contents of amine nitrogen.

The subject-matter of Claim 1 of the application is, thus, non-obvious over this state of the art.

The same conclusion applies to independent Claims 13, 14 and 20 which relate to subject-matter comprising the same thermosetting components and/or thermoset material as specified in Claim 1.

Owing to their dependency on the respective independent Claims 1, 14 and 20, the subject-matter of Claims 2 to 12, 15 to 19, 21 and 22 is also inventive.

Therefore, the subject-matter of all Claims 1 to 22 complies with the requirements of Article 56 EPC.

5. Claims 1 to 22 of the application in suit are also in agreement with the other requirements of the EPC, particularly with those of Articles 84 and 123(2).

6. In these circumstances there is no need to hold oral proceedings.

7. Since the description of the application is not yet in agreement with the claims, the Board decides to make use of its powers under Article 111 EPC and remits the case to the Examining Division for the sole purpose of bringing the description into conformity with the claims.

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 Claims 1 to 22 as filed on 7 March 1997 and after the description has been adapted.

The Registrar:

E. Görgmaier

The Chairman:

C. Gérardin

C. Gérardin