

Veröffentlichung im Amtsblatt	Ja/Nein
Publication in the Official Journal	Yes/No
Publication au Journal Officiel	Oui/Non

Aktenzeichen / Case Number / N^o du recours : T 617/89 - 3.3.3

Anmeldenummer / Filing No / N^o de la demande : 83 108 095.7

Veröffentlichungs-Nr. / Publication No / N^o de la publication : 0 102 566

Bezeichnung der Erfindung: Beta-hydroxy urethane low temperature curing agents

Title of invention:

Titre de l'invention :

Klassifikation / Classification / Classement : C08G 18/80

ENTSCHEIDUNG / DECISION

vom / of / du 6 November 1990

Anmelder / Applicant / Demandeur :

Patentinhaber / Proprietor of the patent /

Titulaire du brevet :

PPG Industries Inc.

Einsprechender / Opponent / Opposant :

Hoechst AG

Stichwort / Headword / Référence :

EPÜ / EPC / CBE Articles 56, 114, 115

Schlagwort / Keyword / Mot clé :

"Inventive step (confirmed after amendment) -
opposite teachings in the prior art in
connection with the same structural feature"

Leitsatz / Headnote / Sommaire

Summary of Facts and Submissions

- I. The mention of the grant of the patent No. 102 566 in respect of European patent application No. 83 108 095.7 filed on 17 August 1983 and claiming priority of 18 August 1982 of an earlier application in the United States, was published on 12 November 1986 on the basis of 14 claims.

Claim 1 read as follows:

"A curable composition comprising:

- (i) an active hydrogen-containing material,
- (ii) a beta-hydroxy urethane, and
- (iii) a cure catalyst."

Claims 2 to 11 were directed to preferred embodiments of the composition according to the main claim and Claims 12 to 14 to a process or method for coating a substrate with such composition.

- II. On 12 August 1987 the Appellant (Opponent) filed a notice of opposition by telex, duly confirmed in writing, against the grant of the patent and requested revocation thereof for non-compliance with the requirements of Article 100a) EPC.

This objection, which was supported initially by document

- (1) EP-A1-12 463,

was emphasised and elaborated in later submissions based mainly on document

- (4) Journal of the American Chemical Society, 79 (1957), 672.

- III. The Opposition Division rejected the opposition in a decision dated 3 August 1989 which was based essentially on the following reasons:

It was first stated that novelty was not under attack. In particular, it was specified that the polyurethanes according to document (4) did not correspond to the cured compositions according to the patent in suit. As to inventive step, document (1) could not suggest the claimed solution, since it concerned an entirely different reaction involving another reaction mechanism. This fundamental difference prevented the man skilled in the art from applying to the chemistry of carbamates the theoretical principles known for esters.

- IV. Observations by a third party were filed on 25 August 1989 in accordance with Article 115 EPC. In these observations it was first said that the claimed subject-matter was not novel with regard to the teaching of US-A-4 096 291 (document (5)). Further, it was stated therein that the technical concept underlying the patent in suit could not be regarded as inventive in view of the combined disclosure of documents (4) and (5).

- V. The Appellant thereafter filed a notice of appeal on 7 September 1989 and paid the prescribed fee at the same time. In the Statement of Grounds of Appeal filed on 5 December 1989 the Appellant in turn raised an objection of lack of novelty relying on the afore-mentioned document (5) and further on a new document, US-A-3 360 504 (document (6)). Regarding the issue of inventive step, he argued that the compositional features of the claimed subject-matter were obvious. On the one hand, document (1) disclosed the beneficial influence, on the temperature of cross-linking involving a transesterification reaction,

of the presence in polyesters of units derived from 1,2-polyols; on the other hand, it was known from document (4) that 2-(hydroxyethyl)-carbamates lost ethylene glycol quite easily.

- VI. During oral proceedings held on 6 November 1990 the Respondent filed as Main Request a new set of 14 claims, wherein Claim 1 was now directed to "a curable, water-based, protective or decorative coating composition" and Claims 12 and 13 to a process, respectively a method, wherein such composition was used. Further, the components (i) in Claim 1 and (c) in Claims 12 and 13 were defined as being "an active hydrogen-containing polymeric material".

Additionally, the Respondent filed two further sets of claims as Auxiliary Requests. In Claims 1, 11 and 12 according to the First Auxiliary Request, the cure catalyst was defined as a metal salt or complex comprising tin or lead; in Claims 1, 11 and 12 according to the Second Auxiliary Request, it was specified that the β -hydroxy urethane was the reaction product of an isocyanate compound or polymer and a 1,2-polyol acting as a blocking agent, the equivalent ratio of -NCO groups to -OH groups being 1:1.5 to 1:2.

- VII. Although thereafter the Appellant conceded the novelty of the subject-matter of the so amended Claim 1 according to the Main Request, he contended that neither the claimed modification of the composition described in Example 16 of document (5), nor the use of such modified composition as a water-based coating composition, could be regarded as inventive features in view of the teaching of documents (1) and (4).

Besides this substantive objection, the Appellant raised an objection under Article 123(2) EPC, arguing that the concept of water-based composition only applied to epoxy resins, and not to any polymeric material.

VIII. In support of the allowability of the present claims, the Respondent first underlined that Example 16 of document (5) did not demonstrate the formation of, nor any intent to form β -hydroxy urethanes. Furthermore, although ethylene glycol had been mentioned therein in a list of suitable blocking agents, there was no incentive to use this specific compound when splitting off or curing reactions at low temperature were desired. As to documents (1) and (4), they would not have led the skilled man to the subject-matter of the patent in suit, since the former document dealt with plain carboxylic acid esters, which were structurally different from carbamates, and the latter disclosed deblocking conditions which would be totally inadequate for the present invention.

IX. The Appellant requested that the decision under appeal be set aside and the patent be revoked.

The Respondent requested that the appeal be dismissed and that the patent be maintained on the basis of Claims 1 to 14 and the description submitted as Main Request during oral proceedings; or, alternatively, on the basis of one of the two sets of Claims 1 to 13 and descriptions submitted as Auxiliary Requests during oral proceedings.

Reasons for the Decision

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

2. As is apparent from paragraph V above, the late-mentioned documents (5) and (6) are regarded by the Appellant as the most relevant prior art, since both documents are relied upon to raise an objection of lack of novelty for the first time in the Statement of Grounds of Appeal.

After examination of these two documents the Board found them to be sufficiently relevant to raise new issues. Using its discretionary power, it has, therefore, decided to admit them into the appeal procedure (Article 114(2) EPC).

In view of the content of document (5), the Respondent has restricted the scope of Claims 1, 12 and 13 during oral proceedings, after which restriction the maintenance of the patent was no longer at risk (see reasons hereinafter).

The question of the remittal of the case to the first instance therefore does not arise (cf. Decision T 326/87 of 28 August 1990 "Polyamide Compositions/DU PONT", to be published).

3. The current wording of the claims according to the Main Request does not give rise to any objections under Article 123 EPC.

In substance, the wording of Claim 1 differs from that as originally filed and granted by the fact that the claimed composition is defined as a "curable, water based, protective or decorative coating composition". This wording is supported by page 1, lines 29 to 31, page 2, lines 11 to 13 and page 8, lines 7 to 15 and 24 to 28 of the description of the application as originally filed;

corresponding to page 2, lines 37/38, page 2, lines 46/47 and page 4, lines 52 to 57 and 63 to 65 of the patent specification. The incorporation of the word "polymeric" in the definition of component (i) is justified in view of the film-forming material quoted in the description on page 2, lines 20 to 26, respectively page 2, lines 53 to 58. Further, Claim 12 has been amended by incorporation of the words "water-based" and "polymeric" in (c) and, likewise, Claim 13 by the incorporation of the word "polymeric" in (c); these additions are acceptable for the same reasons as those carried out in Claim 1. Claims 2 to 11 and 14 have not been amended during examination or opposition procedure, so that their wording is identical to the version as originally filed and granted of these claims.

During oral proceedings, the Appellant argued that only epoxy resins were disclosed in the patent in suit in connection with water-based compositions. The Board cannot share this restrictive interpretation of the description, since the active hydrogen-containing materials are defined as film-forming compositions and exemplified by a non-limiting list of polymers comprising epoxy polymers, acrylic polymers and polyesters, wherein polyepoxides are said to be particularly preferred (page 2, lines 52 to 58). The Board cannot interpret this mere preference as a restrictive statement regarding the suitability of other polymers for water-based compositions, all the more so as the active hydrogen-containing material may comprise units derived from two different polymers, for instance in the case of a reaction of chain extension of the epoxy resin with a polycaprolactone diol (page 3, lines 7 to 9 and Example IB).

For these reasons, therefore, the claims are regarded as adequately supported by the original disclosure.

4. The patent in suit concerns a curable water-based coating composition comprising a β -hydroxy urethane as well as a process and a method for applying such a composition on a substrate. A similar subject-matter is disclosed in document (5) which the Board, like both parties, regards as the closest state of the art. That document describes a process for the production of films and coatings from solvent-free compositions based on combinations of blocked polyisocyanates, wherein at least 40 percent of the isocyanate groups are blocked with an alkanol or cycloalkanol, with compounds containing active hydrogen atoms which are reactive with isocyanate groups, and whereby the coated substrate is heated to a temperature allowing both splitting off of the blocking agent and complete cross-linking (column 2, lines 48 to 69 and Claim 1). More specifically, Example 16 mentions a melt composition containing 500 parts by weight of a polyester derived from dimethyl terephthalate, ethylene glycol and glycerol; 400 parts by weight of a condensation product of tolylene-2,4-diisocyanate, ethylene glycol and trimellitic acid anhydride; and 100 parts by weight of cresol. After solidifying to a vitreous mass and being crushed to a powder at room temperature, this material is introduced into a melting bath, melted at 140°C and applied to a copper wire; the coated wire is then heated to 160°C and eventually exposed to a temperature of 400°C in a stoving oven. Although the resulting coatings exhibit desirable properties, this composition suffers from two major drawbacks: First, the temperature necessary to achieve complete hardening is very high and presently incompatible with the advent of energy conservation; secondly, the composition applied as a melt aims at the production of rather thick layers by successive coating operations, which contributes to making the process overall unattractive.

In the light of this prior art teaching, the technical problem underlying the patent in suit may thus be seen in providing a composition that cures effectively at relatively lower temperatures and is applicable as an aqueous dispersion in a single operation, especially by electrophoretic coating techniques.

According to the patent in suit, this problem is solved by water-based ternary compositions comprising an active hydrogen-containing polymeric material, a β -hydroxy urethane and a cure catalyst.

In view of the experimental results reported in the patent in suit, especially the data in the Table on page 10, which demonstrate that an acceptable curing level of the coating can be achieved at relatively low temperatures, whereas, as will appear hereinafter, a poor reactivity is indicated in documents (5) and (4), wherein the same splitting off reaction is involved, the Board is satisfied that the above defined technical problem is effectively solved.

5. After examination of the cited documents the Board has come to the conclusion that this technical teaching is not disclosed in any one of them and that the subject-matter of the patent in suit according to the Main Request is, therefore, novel. Since the issue of novelty with respect to the teaching of documents (5) and (6) is no longer maintained by the Appellant, it is not necessary to consider this matter in detail.
6. It still remains to be examined whether the subject-matter of the patent in suit as defined in Claim 1 involves an inventive step with regard to the teaching of the cited documents.

6.1 In spite of a superficial similarity between the claimed compositions and the ternary composition according to Example 16 of document (5), there is no incentive for the skilled man to consider any compositional feature of this prior art disclosure for the solution of the above defined technical problem.

6.1.1 First, although the description of this document refers to aliphatic diols as suitable blocking agents and even mentions ethylene glycol and propylene glycol (column 4, lines 49 to 65), this passage contains in fact a fairly long list of conventional alkanol and cycloalkanol blocking agents. There is no indication as to possible advantages in terms of low curing temperature as the result of the use of these specific glycols. In this respect, thus, the choice made by the Respondent must be regarded as a non-obvious selection.

As to the composition according to Example 16, far from pointing to the claimed compositions, the curing temperature of 400°C mentioned there would rather deter the skilled man from looking for a solution along these lines, for it is among the highest temperatures specified in the examples. More promising would appear to be the compositions illustrated in Examples 3, 10 to 12, 14 and 15, which are hardened at temperatures between 230 and 300°C and wherein no such glycol is used as blocking agent.

6.1.2 A second aspect to be considered is the role played by the cresol in the prior art composition.

The necessity to apply the coating composition as a melt requires the viscosity to be within a certain range, in

practice below 40000 cP and preferably below 5000 cP (column 7, lines 29 to 32). This melt may also be used in solvents, such as phenols and cresols (column 8, lines 5 to 11); this would suggest that the role of the cresol in the ternary composition of Example 16 is mainly to adjust the viscosity of the melt. This interpretation is actually confirmed by the wording of Example 16, where it is specified that the polyester is treated with the condensation product in cresol "to form a homogeneous easily stirrable melt" having an appropriate viscosity.

As appears from the description (column 4, line 66 to column 5, line 12), the composition mentioned in Example 16 corresponds in fact to a preferred embodiment of the process according to document (5), wherein mixtures of blocked polyisocyanates are used, in which 40 to 99 equivalent percent of the polyisocyanates are blocked with an alkanol or a cycloalkanol and 60 to 1 equivalent percent are masked with blocking agents which split off at temperatures at least 30°C below the alkanols or cycloalkanols; cresols are quoted as specific examples of such blocking agents. According to this preferred embodiment, thus, the cresol acts as a blocking agent splitting off at a lower temperature than the alkanol or cycloalkanol blocking agent, giving thus rise to a two-step unblocking reaction. This, incidentally, provides evidence that at the priority date of the patent in suit compounds like phenols, lactams and oximes were known to split off at a lower temperature than alkanols and that, consequently, the skilled man had no reason to look among alkanols in order to find a solution to the above defined technical problem.

Without disputing these two interpretations as to the role of cresol in the composition according to Example 16, the Appellant put forward during oral proceedings that cresols

were also quoted among the catalysts for polyurethane cross-linking reactions (column 8, lines 22 to 27 read in conjunction with lines 59 to 60). If so, however, the cresol is only available as a so-called latent catalyst after splitting off, i.e. above a certain temperature at which the free isocyanate group is regenerated as well, and is used accordingly in catalytic quantities only (column 9, lines 16 to 34). By contrast, the amount of cresol in the composition according to Example 16, wherein it represents 10 percent by weight of the ternary composition, cannot be regarded as a "catalytic" quantity. Moreover, the curing temperature of 400°C, which is in the upper range of the preferred temperatures in the stoving oven, must be regarded as high enough to destabilise the isocyanate and, therefore, bring about the cross-linking reaction, making thus the catalyst superfluous, as actually envisaged in the description (column 8, lines 17 to 27).

In conclusion, although the cresol may be regarded in principle as a catalyst for polyurethane cross-linking reactions, the quantities thereof present in the composition according to Example 16 and the temperature of the oven shaft do not suggest a predominantly catalytic function to cresol.

- 6.1.3 A further point to be considered is the structure of the blocked polyisocyanate used in Example 16, which is defined as a condensation product of 1 mol of tolylene-2,4-diisocyanate, 1 mol of ethylene glycol and 1 mol of trimellitic acid anhydride. As argued by the Respondent, these three reactants could, depending upon the reaction conditions, lead to different final products. However, even if one accepted the interpretation proposed by the Appellant during oral proceedings and regarded the condensation product as illustrative of the particular

blocked polyisocyanates referred to at column 4, lines 13 to 27, the teaching of Example 16 would still be restricted to a blocked polyisocyanate having an imide group, i.e. a specific structural feature which is neither claimed in the patent in suit, nor even envisaged in the description thereof.

- 6.1.4 For these various reasons, the teaching of document (5) and, more specifically, of its Example 16, cannot lead towards the claimed subject-matter.
- 6.2 Contrary to the repeated submissions made by the Appellant during oral proceedings, all the experimental figures and results in document (4) concur to show that liberation of ethylene glycol from 2-(hydroxyethyl)-carbamate does not occur so easily.

This document deals with an alternative process for the preparation of polyurethanes without starting from chemically sensitive diisocyanates. For this purpose, 2-(hydroxyethyl)-carbamates obtained from the reaction of primary amines with ethylene carbonate are subjected to vacuum distillation in the presence of barium oxide and/or zinc borate as catalysts (page 672, Abstract to column 2, paragraph 2, in particular reaction (1); page 675, section "Preparation of Polymers"). According to the latter section, a mixture of 0.1 mole of melted monomer is heated together with the above catalyst system under a pressure of 0.5 to 2 mm Hg at the lowest temperature at which ethylene glycol is formed; then the temperature is raised in successive stages to about 220°C, and the heating continued as long as ethylene glycol is liberated, i.e. up to 50 hours, leading to a yield of 80 to 90%. In the Board's view, such figures cannot be regarded as suggesting that ethylene glycol splits off easily from 2-(hydroxyethyl)-carbamates or β -hydroxy urethanes.

Besides the kinetic and quantitative aspects of this reaction, it is essential to observe that the polycondensation reaction is not specific when transesterification is involved additionally (page 672, reaction (2)), as in the case of the patent in suit. When the above type of reaction is extended to the preparation of polyurethanes other than those derived from ethylene glycol, an appreciable transesterification occurs only in the case of high-boiling diols; in this case, however, the polyurethanes obtained are copolymers containing units derived from such diols and from ethylene diol (page 672, column 2, last paragraph to page 673, column 1, paragraph 1). This means that acceptable yields can only be achieved at the expense of the specificity of the polycondensation reaction.

These dissuasive conclusions regarding both the liberation of ethylene glycol and the polyurethane formation by transesterification cannot be an incentive for the skilled man to base the preparation of coatings on compositions involving these reactions.

- 6.3 Nor can the beneficial influence of β -hydroxy groups in poly-carboxylic-acid esters on the course of the transesterification reaction disclosed in document (1) be necessarily transposed to polyurethanes.

This document describes a thermosetting water-soluble binder composition comprising (I) a mixture or a precondensate of (1) a hydroxyl-containing resinous material and (2) as cross-linking agent a non-acidic polyester of a polycarboxylic acid having more than one β -hydroxyester group per molecule and (II) as curing catalyst a transesterification-promoting metal salt or metal complex (Claims 1 and 16). It is specified that the

β -hydroxyester group is an essential structural feature to ensure sufficient cross-linking at acceptable temperatures within acceptable times (page 10, lines 13 to 27). By the process of curing or stoving the coating, β -hydroxyester groups of the polyester transesterify to the effect that ester bonds are formed between carboxyl groups of the polyester and hydroxyl groups of compound (I)(1), whereby a glycol-type compound is released. When, on the contrary, a polyester is used which does not contain such β -hydroxyester groups, the transesterification is too sluggish to effect sufficient cross-linking at acceptable conditions, which impairs the solvent resistance of the stoved coating.

In spite of this promising teaching, there was no incentive for the skilled man to consider β -hydroxyurethane groups for a similar reaction on the sole basis of their functional similarity to β -hydroxycarboxylic-acid ester groups, because he would have known from documents (5) and (4), both published before document (1), that, in the case of β -hydroxyurethane compounds, a splitting off reaction of ethylene glycol only occurs at unacceptable thermal or kinetic conditions. In the Board's view, the difference in ease of splitting off ethylene glycol in document (1) with regard to documents (5) and (4) was thus more likely to be attributed to the β -hydroxy compound itself, i.e. to the specificity of component (I)(1) in the composition described in document (1).

- 6.4 As to document (6), its teaching can no longer be regarded as relevant against the water-based compositions claimed at present.

This document describes the use of hydroxyalkyl carbamates having vinyloxyalkyl or vinylthioalkyl groups substituted

on the nitrogen atom as functional monomers for the preparation of homopolymers and copolymers (column 1, lines 11 to 14). Even though it is specified that the presence of the hydroxyl group(s) increases the versatility of reaction with modifying agents, such as aldehydes, acids and isocyanates (column 1, lines 20 to 24), and even though these polymers can be regarded as β -hydroxypolyurethanes, there is no teaching of a particular crosslinking ability which could be related to this structural feature. This is readily apparent from Example 2, wherein the preparation of a copolymer of 58.2% of butyl methacrylate, 38.8% of methyl methacrylate and 3.0% of 2-hydroxyethyl-N-(2-vinyloxyethyl)carbamate is described; the amount of the latter as reactive monomer may be sufficient to improve the adhesion properties of the copolymer, but not to provide enough crosslinking as required in the patent in suit, all the more so as this compound would have to function both as blocked isocyanate and as hydroxy source. This critical argument put forward by the Respondent (Statement filed on 5 October 1990, page 3, paragraph 2) has not been disputed by the Appellant.

In the Board's view, similar considerations would apply to the copolymer according to Example 4, referred to more specifically by the Appellant, which is obtained from 64 g of ethyl acrylate, 132 g of vinyl acetate and 4 g of 2-hydroxyethyl-N-(2-vinyloxyethyl)carbamate. Further, the numerous references to air-drying of this copolymer at elevated temperatures (column 3, line 73 to column 4, line 10) would not be interpreted by the skilled man as a proper crosslinking induced by unblocking reaction, but rather as a cure involving mainly the removal of residual water and/or solvents.

In view of these differences in composition and reaction, the skilled man had no reason to consider the teaching of document (6) for the solution of the above defined problem.

- 6.5 In conclusion, in view of the foregoing, the subject-matter of Claim 1 is not rendered obvious by the documents relied upon by the Appellant, either in isolation or in combination, and, therefore, involves an inventive step.
7. Claim 1 being allowable, the same applies to the dependent composition Claims 2 to 11, which represent preferred embodiments of the subject-matter of Claim 1, as well as to Claims 12 to 14, which are directed to a process for providing a protective or decorative coating for a substrate by applying and curing a composition as defined in Claim 1 and to a method of coating a conductive surface by electrodeposition comprising the use of a composition as defined in Claim 1, and whose inventiveness is supported by that of the main claim.
8. The Main Request being thus allowable, it is not necessary to consider either of the Auxiliary Requests.

Order

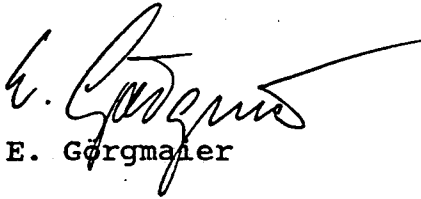
For these reasons, it is decided that:

1. The decision under appeal is set aside.


2. The case is remitted to the first instance with the order to maintain the patent on the basis of the Claims 1 to 14 and the description submitted as Main Request during oral proceedings.

The Registrar:

The Chairman:



E. Gorgmaier



F. Antony