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**Datasheet for the decision
of 17 December 2009**

Case Number: T 0118/05 - 3.3.05

Application Number: 98110334.4

Publication Number: 0884290

IPC: C04B 24/26

Language of the proceedings: EN

Title of invention:

Process for producing a polycarboxylic acid

Patentee:

NIPPON SHOKUBAI CO., LTD.

Opponent:

BASF SE

Headword:

-

Relevant legal provisions:

EPC Art. 56

Relevant legal provisions (EPC 1973):

-

Keyword:

"Inventive step (yes) - evidence for the technical solution in the patent in suit - no pointers to the technical solution in the prior art"

Decisions cited:

-

Catchword:

-



Case Number: T 0118/05 - 3.3.05

DECISION
of the Technical Board of Appeal 3.3.05
of 17 December 2009

Appellant: BASF SE
(Opponent) D-67056 Ludwigshafen (DE)

Representative: Witz, Michael
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Respondent: NIPPON SHOKUBAI CO., LTD.
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Representative: Schön, Christoph
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Decision under appeal: Interlocutory decision of the Opposition
Division of the European Patent Office posted
18 November 2004 concerning maintenance of
European patent No. 0884290 in amended form.

Composition of the Board:

Chairman: G. Raths
Members: E. Waeckerlin
S. Hoffmann

Summary of Facts and Submissions

I. The present appeal lies from the interlocutory decision of the opposition division to maintain the European patent No. 0 884 290 on the basis of claims 1 to 6 filed with letter dated 7 September 2004.

Claim 1 as filed with letter dated 7 September 2004 reads as follows:

*"1. Process for producing a polycarboxylic acid as an effective component for a cement mixture, said process including the steps of:
charging "a" parts by weight of a polyalkylene glycol (A) of the general formula (1) below and "b" parts by weight of a (meth)acrylic acid monomer (B) of the general formula (2) below into a reactor in the range of the equation (1) below;
carrying out an esterification reaction of the resultant mixture in the reactor, thus obtaining a polyalkylene glycol (meth)acrylate (C) of the general formula (3) below; and
copolymerizing the resultant reaction mixture including the polyalkylene glycol (meth)acrylate (C) and the (meth)acrylic acid monomer (B), thus obtaining the polycarboxylic acid;
wherein the general formula (1) is:*



wherein the general formula (2) is:



wherein the equation (1) is:

$$45 \leq \{(a/n^{1/2})/b\} \times 100 \leq 160$$

wherein the general formula (3) is:



wherein: R^1O is one kind of oxyalkylene group or a mixture of two kinds thereof, wherein the two or more kinds of oxyalkylene groups may be added either in block form or at random; R^2 is an alkyl group with 1 to 22 carbon atoms, a phenyl group, or an alkylphenyl group; n is an average molar number of the added oxyalkylene groups and is a number of 1 to 300; and R^3 is a hydrogen atom or a methyl group."

II. The following documents were cited *inter alia* in the opposition procedure:

D1: GB 2 280 180 A;

D2: EP 0 753 488 B1;

D3: GB 874 235 A;

D5: E. Breitmaier, G. Jung: Organische Chemie I.
2. Auflage, Stuttgart: Georg Thieme Verlag, 1986,
page 327.

III. The opposition division held that the closest prior art is represented by either D1 or D2.

The technical problem underlying the invention consisted in providing a process for producing a polycarboxylic acid as an effective component for a cement admixture, whereby the process shows a higher productivity (shorter esterification reaction time, thereby avoiding the formation of diester by-products) and gives rise to a production of a polycarboxylic acid having an improved cement dispersibility.

The opposition division acknowledged that the inventors had for the first time recognised the significant role of the "*K-value*" expressed by equation (1). The examples, as well as tables 1 and 2 of the description demonstrated an unexpected higher productivity of the process. Thus, the formation of diesters was negligible. Moreover, when used as additives to cement, the polycarboxylic acids obtained by the process showed an improved efficiency in terms of low amounts required, excellent cement dispersibility and high slump of the cement admixture.

The combination of D1 or D2 with D3 did not lead to the process according to claim 1. When reading D3, optionally together with D5, the skilled person would be inclined to use "*K-values*" much higher than the upper limit of 160 set out in claim 1.

For these reasons the opposition division concluded that the process according to claim 1 involved an inventive step.

IV. The appellant (opponent) gave notice of appeal on 25 January 2005. Together with the grounds of appeal a further document D6 was introduced into the proceedings:

D6: EP 0 705 854 B1.

V. In reply the respondent (proprietor of the patent) filed its comments with letter dated 2 August 2005.

VI. Summons to oral proceedings were issued on 5 August 2009.
In response both parties filed further comments. In addition the respondent filed four sets of amended claims as auxiliary requests 1 to 4 with letter dated 17 November 2009.

VII. Oral proceedings were held on 17 December 2009. After discussion of the claims according to the main request, the set of claims 1 to 6 of the first auxiliary request filed on 17 November 2009 became the new main request.

In claim 1 of the new main request the number "n" is limited to the range of from 10 to 250. Claim 1 reads as follows:

*"1. Process for producing a polycarboxylic acid as an effective component for a cement mixture, said process including the steps of:
charging "a" parts by weight of a polyalkylene glycol (A) of the general formula (1) below and "b" parts by weight of a (meth)acrylic acid monomer (B) of the general formula (2) below into a reactor in the range of the equation (1) below;
carrying out an esterification reaction of the resultant mixture in the reactor, thus obtaining a polyalkylene glycol (meth)acrylate (C) of the general formula (3) below; and
copolymerizing the resultant reaction mixture including the polyalkylene glycol (meth)acrylate (C) and the*

(meth)acrylic acid monomer (B), thus obtaining the polycarboxylic acid; wherein the general formula (1) is:



wherein the general formula (2) is:



wherein the equation (1) is:

$$45 \leq \{ (a/n^{1/2})/b \} \times 100 \leq 160$$

wherein the general formula (3) is:



wherein: R¹O is one kind of oxyalkylene group or a mixture of two kinds thereof, wherein the two or more kinds of oxyalkylene groups may be added either in block form or at random; R² is an alkyl group with 1 to 22 carbon atoms, a phenyl group, or an alkylphenyl group; n is an average molar number of the added oxyalkylene groups and is a number of 10 to 250; and R³ is a hydrogen atom or a methyl group."

Dependent claims 2 to 5 represent particular embodiments of the subject-matter defined in claim 1.

Claim 6 of the new main request is formulated as an independent claim, but it includes all the features of

claim 1. In addition, the peroxide value of the polyalkylene glycol (A) is specified in claim 6 to have a value of 0.7 meq/kg or less.

VIII. The arguments presented by the appellant, as far as they still apply having regard to the new main request, can be summarised as follows:

The auxiliary requests filed by the respondent with letter dated 17 November 2009 should not be admitted. It would be inappropriate to admit such new requests which are neither immediately allowable nor *bona fide* attempts to overcome objections raised.

The subject-matter of the patent as maintained by the opposition division, although slightly differently worded, is essentially non-distinguishable from the subject-matter of the pending divisional patent application Nr. EP 1 247 824 A (application No. 02 014 548.8).

The closest prior art is represented by D1, and in particular example 1 of D1, wherein methoxy polyethylene glycol monomethacrylate and additional methacrylic acid are used as components. The technical problem to be solved was to provide a process for producing a polycarboxylic acid as an effective component for a cement mixture, which process shows a higher productivity and gives rise to the production of a polycarboxylic acid having an improved cement dispersibility.

A person skilled in the art, in order to simplify the process of D1 and to obtain the starting material

monomethacrylate, would resort to the teaching of D3 as a standard procedure for producing monofunctional and difunctional acrylic or methacrylic esters. D3 discloses that a molar excess of (meth)acrylic acid over poly(alkylene oxide)monoalkylether is desirable. Furthermore it belongs to the common general knowledge as illustrated e.g. in D5, to use an excess of acid or alcohol in order to increase the productivity of esterification reactions. When combining the teaching of D1 and D3 with the common general knowledge, the skilled person would arrive at the claimed process.

Alternatively, the skilled person would combine D1 with the teaching of D6. In D6 a process for producing ethylenically unsaturated biphilic monomers is disclosed. The monomers thus obtained may be acrylate and methacrylate esters of certain nonionic surfactant alcohols obtained by acid catalyzed esterification of the corresponding surfactant alcohols with an excess of an unsaturated carbonic acid monomer such as methacrylic acid. According to D6 the resulting product can be used directly in the copolymerization, in analogy to the process of the patent in suit.

Having regard to either the combination of D1, D3 and the common general knowledge exemplified by D5, or alternatively to the combination of D1 and D6, the claimed process lacks an inventive step.

IX. The respondent dissented with the views expressed by the appellant, presenting essentially the following arguments:

The technical problem solved by the process according to claim 1 was the provision of a process for producing a polycarboxylic acid as an effective component for a cement admixture, which process shows a superior and unexpectedly high productivity of the polycarboxylic acid produced and gives rise to the production of a polycarboxylic acid having a superior and unexpectedly improved cement dispersibility.

Document D1 does not disclose the first step of the claimed process, namely the esterification of the polyalkylene alcohol and (meth)acrylic acid under the conditions set out in claim 1. As far as D3 is concerned, this document does not provide a hint how to carry out the first step of the process, but teaches away from using such an esterification process.

Document D5 does not provide relevant additional information. Document D6 on its part does not teach the specific esterification reaction either. Rather it teaches an esterification reaction wherein only a partial esterification takes place.

Therefore the skilled person would not have considered either the combination of D1 and D3, or the combination of D1 with D6, when trying to solve the technical problem posed.

- X. The appellant requested that the decision under appeal be set aside and that the European patent be revoked.

The respondent requested that the decision under appeal be set aside and that the patent be maintained on the basis of the set of claims 1 to 6 of the main request corresponding to the first auxiliary request filed on

17 November 2009, or on the basis of one of the auxiliary requests 2 to 4 filed on 17 November 2009.

Reasons for the Decision

Admissibility of the auxiliary requests filed with letter dated 17 November 2009

- 1.1 The auxiliary requests 1 to 4 were filed by the respondent on 17 November 2009, i.e. one month before the date of the oral proceedings. They are clearly occasioned by the objection of lack of inventive step raised by the appellant. Moreover they are *prima facie* in conformity with the requirements laid down in Article 123(2) and (3) EPC.

- 1.2 In particular the auxiliary request 1 as filed on 17 November 2009 corresponded to the main request then on file, except that the range of the values of "n" was restricted from "*a number of 1 to 300*" to "*a number of 10 to 250*". A basis for this limitation can be found *inter alia* on page 7, lines 5 - 6 of the application as originally filed. By restricting the range of "n" representing the average molar number of added oxyalkylene groups, the claimed process was confined to the production of polycarboxylic acids having superior characteristics (see respondent's letter dated 17 November 2009, page 4, paragraph II).

- 1.3 The board found no evidence in support of the allegation that the auxiliary requests were not submitted in good faith. Under these circumstances the board concludes that the auxiliary requests 1 to 4 as

filed on 17 November 2009, auxiliary request 1 becoming the main request and the remaining requests becoming auxiliary requests 1 to 3, respectively, are allowable.

Allowability of the amendments - Article 123(2), (3) EPC

2. No formal objections under Article 123(2) OR (3) EPC were raised by the appellant, and none is seen by the board, as all amendments effected to the claims are of a restrictive nature and based on the application as originally filed. Consequently the amendments are allowable.

Novelty - Article 54 EPC

Main request corresponding to auxiliary request 1 of 17 November 2009

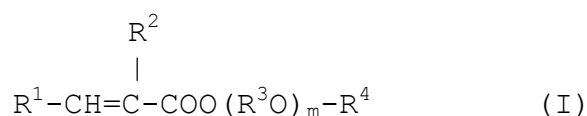
3. No objection of lack of novelty was raised by the appellant, and the board is not aware of any document that could call the novelty of the claimed process into question. Consequently the board is satisfied that the subject-matter of claims 1 to 6 meets the requirements of Articles 52(1) and 54 EPC.

Inventive step - Article 56 EPC

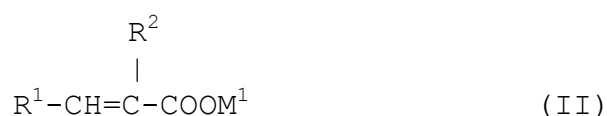
Main request corresponding to auxiliary request 1 of 17 November 2009

4. The present invention relates to a process for producing polycarboxylic acids as effective components for cement admixtures.

- 4.1 At the oral proceedings both parties considered document D1 to represent the closest prior art. The board can accept this.
- 4.2 D1 discloses a cement-dispersing agent for controlling the fluidity of cementitious compositions containing an aqueous solution of a polymer. Suitable polymers include the products resulting from the polymerisation of an alkylene glycol mono(meth)acrylic acid ester monomer of the general formula (I):



wherein R^1 and R^2 are independently hydrogen or methyl, R^3 is an alkylene group of from 2 to 4 carbon atoms, R^4 is hydrogen or an alkyl group of 1 to 22 carbon atoms, and m represents an integer of 1 to 100, with a (Meth)acrylic acid monomer of the general formula (II):



wherein R^1 and R^2 have the abovementioned significances, and M^1 is hydrogen, monovalent metal, divalent metal, ammonium group or an organic amine group (see D1, claims 1 and 3; correct formulae on page 3, lines 7 - 8 and 14 - 15).

The alkylene glycol mono(meth)acrylic acid ester of formula (I) corresponds to the polyalkylene glycol (meth)acrylate of formula (3) as defined in claim 1 of the patent in suit. Thus, D1 discloses the second step

- of the process according to claim 1 of the patent, namely the copolymerisation of polyalkylene glycol (meth)acrylate esters and (meth)acrylic acid to the corresponding polycarboxylic acids.
- 4.3 The technical problem underlying the patent in suit in the light of document D1 was to improve the overall productivity of the process for producing polycarboxylic acids having a good cement dispersibility and a high slump retainability when used as a component in cement admixtures (see patent, page 2, paragraphs [0001] and [0006]; page 7, paragraph [0041]; page 8, paragraphs [0054] and to [0056]).
- 4.4 As the solution to the technical problem the patent in suit proposes a process for producing polycarboxylic acids according to claim 1 of the main request, characterised in that the esterification step comprises the reaction of a polyalkylene glycol of formula (1) with an excess of (meth)acrylic acid monomer of formula (2), under conditions complying with a "K-value", i.e. a value of the parameter $\{(a/n^{1/2})/b\} \times 100$, within the range of from 45 to 160 in accordance with equation (1), and that the esterification step is followed by the copolymerisation of the resultant reaction mixture in a second step.
- 4.5 It has to be examined whether the technical problem has actually been solved.
- 4.5.1 Examples 5 to 8 illustrate the process according to claim 1. In these examples mixtures of methoxypolyethylene glycol having a value of n equal to 25 and methacrylic acid were esterified until a

conversion rate of 99 % was reached. In these examples the "*K-values*" were 56 (example 5), 69.7 (example 6), 101.7 (example 7) and 121.5 (example 8), respectively. It was found that the reaction mixtures thus obtained were virtually free of the undesirable by-product polyethylene glycol dimethacrylate (see pages 12 to 13, examples 5 - 8). In a further experiment with a "*K-value*" of 215 the content of polyethylene glycol dimethacrylate was determined to be 3.5 % (see pages 14 to 15, "*comparative example 1*").

It can be derived from the results that esterification under the conditions set out in claim 1 leads to reaction mixtures containing low or negligible amounts of polyethylene glycol dimethacrylate. The reaction mixtures can therefore be used as starting materials in the copolymerisation step without prior isolation and purification of the polyalkylene glycol (meth)acrylates of formula (3).

- 4.5.2 Slump values of cement compositions containing carboxylic acids produced by the process of claim 1 were measured according to Japanese industrial standards JIS A1101 and A1128. It was found that the slump values varied between 17.5 cm and 19 cm immediately after the preparation of the cement compositions. After 30 minutes the values varied between 13.5 cm and 15.5 cm, and after 60 minutes values between 9 and 13.5 cm were measured, depending on the specific type and amount of polycarboxylic acid used in the experiments (see page 10, paragraph [0068]; Table 1, examples 1, 2, 3, 11).

It follows from the results that the polycarboxylic acids, when used as cement admixtures in low concentrations, exhibit a high cement dispersibility. In fact, the slump values of the cement compositions are relatively high and decrease only slowly with time.

4.5.3 For a number of examples the flow values of cement admixtures containing carboxylic acids produced by the process of claim 1 were determined as a function of time. The results reveal that there is only a relatively small drop of the flow values with time, namely from 104 to 80 mm, 106 to 89 mm, 95 to 85 mm, 96 to 94 mm and 107 to 70 mm, respectively, the first value being measured immediately after the preparation of the cement admixtures, and the last value after 60 minutes (see pages 12 to 14, examples 5 - 9, 11; page 16, Table 2).

On the basis of these examples it may be concluded that the cement admixtures display a high slump retainability, even if the amount of additive is small, for example 0.3 wt % in the case of example 8, the other amounts being smaller (0.25, 0.18, 0.15 wt %).

In a further experiment with an additive based on a "K-value" of the concerned monomer mixture of 215 (see page 15, line 26), a significantly higher amount of not less than 0.5 wt % of additive was required to achieve about the same flow values as those of example 8 (see page 16, Table 2, "comparative example 3").

4.5.4 As far as the measurements of flow values are concerned, the appellant observed that in the case of example 9 a value of 79 mm was found after 60 minutes. This is

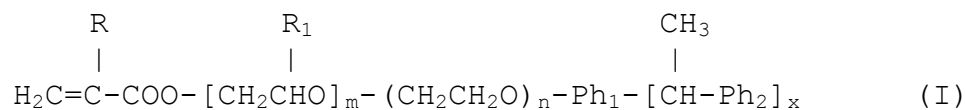
worse than the value of 92 mm of "*comparative example 3*" (see page 16, Table 2). It has to be borne in mind, however, that a considerably higher amount of additive (0.5 wt %) is used in "*comparative example 3*" in comparison to example 9 (0.15 wt %). Therefore the results of the two examples cannot be compared directly.

4.6 In view of the experimental data referred to above, and in the absence of any evidence to the contrary, the board concludes that the technical problem is, in fact, credibly solved by the process according to claim 1.

4.7 It remains to be decided whether the proposed solution is obvious having regard to the prior art.

4.7.1 The technical teaching of D3 does not provide an incentive to produce polycarboxylic acids by a two step process under the conditions set out in claim 1 of the patent. In particular, D3 is silent on the concept of the "*K-value*". It does not address the issue of the suppression of unwanted by-products, and it does not mention either that the reaction mixture resulting from the esterification step is to be used directly as starting material in the copolymerisation step, i.e. without prior isolation of the polyalkylene glycol (methacrylates) of formula (3). For these reasons the skilled person would not have attempted to solve the technical problem by relying on the teaching of D3.

4.7.2 Document D6 discloses polymers useful as pH sensitive thickeners for aqueous compositions. These polymers are prepared by copolymerising three basic monomers (A), (B) and (C), respectively. The monomers (C) are acrylic or methacrylic acid esters of the general formula (I):



wherein R and R₁ represent hydrogen or methyl, Ph₁ represents a phenylene group, Ph₂ represents a phenyl group, n is an average number of from 6 to 100, m is an average number of from 0 to 50, provided that n is ≥ m and (m+n) is an average number of from 6 to 100, and x is an average number of from 2 to 3. The preferred monomers of formula (I) include (1-phenyl ethyl)_x phenyl poly(alkyleneoxy) (meth)acrylates moieties, where x is a number of from about 2 to 3, the preferred poly(alkyleneoxy) moiety being ethyleneoxy, and the repeating alkyleneoxy units being an average number of from about 6 to 100 (see D6, claims 1, 6, 25; page 2, line 47 to page 3, line 13). The monomers of formula (I) can be prepared by the direct acid catalyzed esterification of the corresponding surfactant alcohol with an excess of the carboxylic acids used as component (A) in the final copolymerisation step, preferably acrylic acid or methacrylic acid. The resulting mixture with excess acid can be used directly in the copolymerisation (see D6, page 3, lines 30 - 32; claims 6, 7).

- 4.7.3 The appellant argued that the skilled person had an incentive to apply the teaching of D6 in order to solve the technical problem, because D6 did not only teach to use an excess of (meth)acrylic monomer in the esterification step, but also to use the resulting reaction mixture directly in the subsequent copolymerisation step, i.e. without isolation of the intermediate (meth)acrylate ester.

- 4.7.4 The board notes that the structure of the surfactant alcohols used as starting materials in the process of D6 is significantly different in respect of the terminal group of the oxyalkylene chain, when compared with the polyalkylene glycols of formula (1) of claim 1 of the patent in suit. As a consequence the intermediate (meth)acrylate esters of formula (I) of D6 are likewise structurally different from the polyalkylene glycol (meth)acrylates according to formula (3) and the final products obtained by the process of claim 1 of the patent in suit, i.e. the polycarboxylic acids resulting from the copolymerisation step.
- 4.7.5 At the oral proceedings the appellant admitted that the compounds involved in the process of D6 exhibit "*a slightly different chemistry*" than the corresponding compounds of formula (1) and (3) according to claim 1 of the patent. The appellant argued, however, that in view of the similarity between the structures of the concerned compounds the teaching of D6 applies in the same way to the polyalkylene glycols of formula (1) and the polyalkylene (meth)acrylates of formula (3), respectively.
- 4.7.6 There is no evidence in support of the appellant's allegation according to which the technical teaching of D6 can be generalised and, thus, applied to processes for producing polycarboxylic acids having another structure than the products obtained by the process of D6.

The board considers that in the absence of such evidence the argumentation of the appellant is based on hindsight. Moreover, while D6 suggests that it is suitable to use an excess of (meth)acrylic acid monomer over the surfactant alcohol in the esterification step, nothing in D6 suggests that the amounts "a" and "b", respectively, of the two components have to be chosen in a manner that the resulting "K-value" lies in the range of from 45 to 160, thus complying with equation (1) of claim 1. Nothing suggests either that the polycarboxylic acids obtained by the process of D6 can be used as effective components for cement admixtures, let alone that they possess the specific properties of the polycarboxylic acids produced in accordance with claim 1 of the patent. D6 is concerned with pH sensitive polymeric thickeners for aqueous systems (see D6, page 2, lines 30 - 43; page 6, lines 10 - 13; page 7, example 3). Nowhere in D3 the cement dispersibility or the slump retainability of the polycarboxylic acids is addressed.

4.7.7 In view of the above considerations, the skilled person, faced with the technical problem posed, had no reason to expect that the technical teaching of D6 would contribute to the solution. Therefore there existed no incentive to combine D1 and D6.

4.8 Having regard to the above considerations the board concludes that the process according to claim 1 of the patent involves an inventive step.

Independent claim 6 and dependent claims 2 to 5 of the main request corresponding to auxiliary request 1 of 17 November 2009

4.9 Independent claim 6, although formulated as independent claim, relates to a particular embodiment of the process of claim 1. Therefore claim 6 can be regarded as equivalent to a dependent claim.

4.10 Thus, claim 6 on the one hand, and claims 2 to 5 on the other hand all relate to particular embodiments of the process according to claim 1. They derive their patentability from claim 1.

Pending divisional application

5. The issue whether the subject-matter of the pending divisional application Nr. EP 1 247 824 A (application number 02 014 548.8) is properly delimited against the parent application and the patent in suit, respectively, does not form part of the present appeals procedure. Once a divisional application has been validly filed, it becomes separate and independent from the parent application. Thus, provided that the conditions laid down in Article 76(1) EPC have been met, the divisional application is to be examined separately from the parent application and has itself to comply independently with all the requirements of the EPC.

Auxiliary requests

6. The main request corresponding to auxiliary request 1 of 17 November 2005 being allowable, there is no need to consider the other 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 department of first instance with the order to maintain the patent in amended form on the basis of the set of claims 1 to 6 filed as first auxiliary request on 17 November 2009 and a description to be adapted.

The Registrar:

The Chairman:

C. Vodz

G. Rath