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Bezeichnung der Erfindung: A poly(acrylate) containing composition and process for producing molded articles
Title of invention:
Titre de l'invention :

Klassifikation / Classification / Classement : C08F 220/20

ENTSCHEIDUNG / DECISION
vom / of / du 07 July 1988

Anmelder / Applicant / Demandeur :

Patentinhaber / Proprietor of the patent / Titulaire du brevet : Union Carbide Corporation

Einsprechender / Opponent / Opposant : TVB

Stichwort / Headword / Référence :

EPÜ / EPC / CBE Art. 54(2), 56

Schlagwort / Keyword / Mot clé : "Novelty (denied) - numerical ranges in chemical compositions" - "Inventive step (denied)"

Leitsatz / Headnote / Sommaire

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Case Number : T 25 /87 - 3.3.1

D E C I S I O N
of the Technical Board of Appeal 3.3.1
of 07 July 1988

Appellant : (Opponent)
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Decision under appeal : **Decision of Opposition Division of the European Patent Office dated 07 November 1986 rejecting the opposition filed against European patent No. 0 018 638 pursuant to Article 102(2) EPC.**

Composition of the Board :

Chairman : K. Jahn

Members : R. Spangenberg

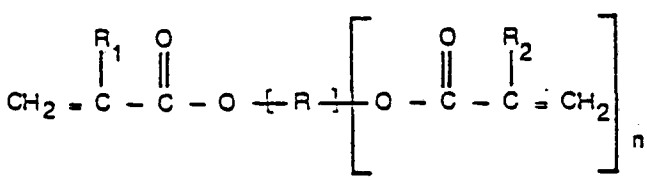
W. Moser

Summary of Facts and Submissions

I. European patent No. 18 638, incorporating 5 claims, was granted on 19 September 1984 on the basis of European patent application No. 80 102 355.7, filed on 30 April 1980 claiming priority of 1 May 1979 and 27 March 1980 from two earlier applications in the USA.

The independent claims read as follows:

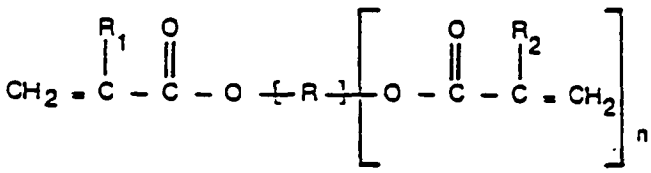
"1. A curable molding composition which comprises: (a) one or more fibers with a melting point or a glass transition temperature above 130°C, (b) a poly(acrylate) characterized by the following empirical formula:



wherein R is the hydroxy-free residue of an organic polyhydric alcohol which contained hydroxy groups bonded to different carbon atoms, R_1 and R_2 are independently hydrogen or methyl, n is 1 to 3, (c) an ethylenically unsaturated monomer which is soluble in and copolymerizable with (b), and (d) an elevated temperature free-radical curing catalyst capable of effecting the co-reaction of (b) with (c), said mixture containing (i) from 15 to 80 weight percent of (a), (ii) from 5 to 60 weight percent of (b), and (iii) from 5 to 60 weight percent of (c), and (iv) from

0.2 to 3 weight percent of (d) and wherein (b) plus (c) has a viscosity of from 1 to 50 mPa.s.

4. A process for producing high performance fiber reinforced thermoset molded articles which comprises:
(I) forming a mixture of:
(a) one or more fibers with a melting point or a glass transition temperature above 130°C, (b) a poly(acrylate) characterized by the following empirical formula:



wherein R is the hydroxy-free residue of an organic polyhydric alcohol which contained hydroxy groups bonded to different carbon atoms, R_1 and R_2 are independently hydrogen or methyl, n is 1 to 3, (c) an ethylenically unsaturated monomer which is soluble in and copolymerizable with (b), and (d) an elevated temperature free-radical curing catalyst capable of effecting the initiation at a temperature not greater than 150°C, of the co-reaction of (b) with (c) and the cure of the mixture within 6 minutes of the initiation of said reaction, said mixture containing (i) from 15 to 80 weight percent of (a), (ii) from 5 to 60 weight percent of (b), and (iii) from 5 to 60 weight percent of (c), and (iv) from 0.2 to 3 weight percent of (d),

(II) providing said mixture into a heatable mold,

(III) molding said mixture at a temperature sufficient to cause the initiation of the co-reaction of (b) and (c) at a temperature not greater than 150°C and

(IV) completing said molding in less than 6 minutes from the time of initiation of said co-reaction.

5. A cured article formed from a composition according to Claims 1 to 3."

II. On 14 June 1985 the Appellant filed a notice of opposition referring to

(1a) Derwent abstract 71-51435 S
(1b) Chemical abstracts 76(1972), 15346 k
(2) DE-B-1078325

and requesting that the patent be revoked in its entirety on the grounds of lack of novelty and inventive step.

In a decision dated 7 November 1986, the Opposition Division rejected the opposition, stating that none of the citations discloses or suggests a composition having all the technical features contained in the above Claim 1.

More specifically, regarding novelty, documents (1a) and (1b) were not found to be relevant because the formula for the acrylate monomer is different and does not refer to the type and amount of catalyst nor to the amount of ethylenically unsaturated monomer to be used.

Regarding inventive step, the Opposition Division found that documents (1a) and (1b) did not constitute a pointer towards the claimed invention, because it relates to copolymerisation leading to products having improved impact and weather resistance, and is not concerned with the inventive concept conceived by the Respondent, which relates to the molding process of Claim 4 made possible by formulating of the compositions as defined in Claim 1.

III. The Appellant appealed against the above decision on 8 January 1987 and paid the appeal fee at the same time.

Together with the Statement of Grounds, which was filed on 11 March 1987, the Appellant further submitted (1) JP-A-71-27247 in the form of an English translation.

(1) is the Japanese patent application abstracted in (1a) and (1b). The Appellant also relied inter alia on (3) Glasfaserverstärkte Kunststoffe (Springer-Verlag 1961), p.390-391, in order to demonstrate the common general knowledge in the technical field concerned.

The Appellant substantially argued that the subject-matter of Claim 1 of the patent-in-suit lacks novelty and, in any case, inventive step when compared with the disclosure of each of documents (1) or (2) as well as the common general knowledge in the art concerned.

The Appellant also submitted the results of viscosity measurements of a resin composition corresponding to that disclosed in Example 1 of (1), thus demonstrating that the compositions tested have viscosities within the range specified in Claim 1 of the patent-in-suit.

IV. The Respondent argued that neither of documents (1) and (2) contain a specific reference to the viscosity of the compositions disclosed therein, and that in any case the examples of (1) do not disclose an amount of catalyst falling within the range specified in Claim 1. He also stated that neither of these documents would suggest to a skilled person that a viscosity of less than 50 mPa.s is important for obtaining the favourable results referred to in the description.

V. At the oral proceedings on 7 July 1988 the Appellant pointed out that the compositions according to Claims 1 to 3 and the articles according to Claim 5 lacked novelty when compared with the whole content of (1), since all the resin mixtures specifically disclosed in (1) have a viscosity falling within the range of the disputed Claim 1.

The claimed process was said to be obvious because the decreased reaction time at the increased reaction temperature could be expected. The Appellant also stated that the expression "molding time" used in the process claim need not necessarily mean "curing time" within the meaning of (1) because a further heating of the molded article after being removed from the mold is not excluded.

VI. The Respondent emphasised that the curing time indicated in (1) was substantially longer than that claimed and that this should be considered as strong evidence for a difference in the compositions themselves.

The Respondent did not question the viscosity measurements made by the Appellant. He also admitted not having any facts or arguments which could rebut the Appellant's statement that similar viscosities must be expected for the other compositions disclosed in (1).

He submitted, however, that any coincidence of parameters disclosed for compositions in (1) with those recited in Claim 1 of the patent-in-suit would only be purely accidental and that, in the absence of any disclosure of viscosity data in (1), this document did not prejudice the novelty of the claimed compositions.

The Respondent considered his process to be inventive. No hint as regards the surprisingly short curing time could be found in the prior art documents, nor was there any hint to be found concerning the particular advantages of the claimed compositions in injection or transfer molding processes.

On the contrary, the long curing time according to (1) would have deterred the skilled person from considering this document at all when looking for short-curing compositions.

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

The Respondent requested that the appeal be dismissed. Furthermore, as an auxiliary request, he submitted that the patent be maintained in amended form on the basis of the process Claim 4 as the only claim. Alternatively, he requested that the patent be maintained on the basis of that process claim further limited to the technique of transfer molding or injection molding.

Reasons for the Decision

1. The appeal complies with the provisions of Art. 106 and 108 and Rule 64 EPC and is, therefore, admissible.

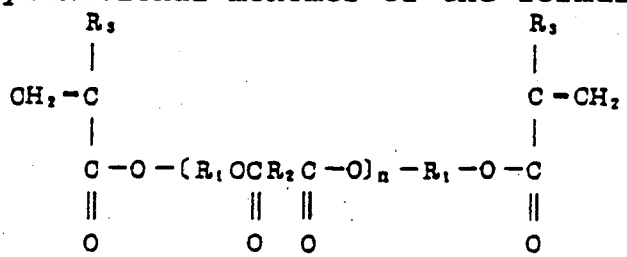
2. Main request

2.1 The patent-in-suit relates to polyacrylate containing compositions, a process for obtaining molded articles and to molded articles so obtained. The polyacrylate compositions contain (a) 15-80 weight percent of

reinforcing fibers (b) 5-60 weight percent of a polyacrylate of the formula described in Claim 1 (see Part I), (c) 5-60 weight percent of an ethylenically unsaturated monomer which is soluble in and copolymerisable with (b) and (d) from 0.2 to 3 weight percent of an elevated temperature free-radical curing catalysts; all weight percentages relating to the total weight of the composition. Such compositions are already known, from (1). The accuracy of the English translation of the original Japanese document was not challenged. This document discloses compositions containing

(a) 10-80% by weight of the molded article of a fibrous material, especially glass fibers (page 6, last 3 lines to page 7, line 3)

(b) 0.5-90% by weight of the resinous components of a di- or polyfunctional monomer of the formula



where R_1 and R_2 represent an aliphatic, alicyclic or aromatic hydrocarbon radical or a derivative thereof, R_1 may be interrupted by - O - in the main chain thereof and R_2 may be interrupted by - O -, - S - or - SO_2 - in the main chain thereof, R_3 represents hydrogen or methyl group, and n is an integer of from 1 to 20 (see page 6, second paragraph and the paragraph bridging pages 2 and 3)

(c) monofunctional vinyl monomers (see page 5, second complete paragraph) and

(d) a heat curing free radical catalyst in an amount of 0.02-4.0% by weight, preferably 0.05-3.0% by weight of the resinous components (see the paragraph bridging pages 5 and 6 and page 6, lines 16 to 27).

It is clear from the description, page 3, lines 7-9 that the amount of component (c), which is more specifically described on page 5, second paragraph, of (1), must be the remainder of the resinous material which is not made up by (b). Consequently the amount of (c) disclosed in (1) is from 10 to 99.5% by weight of the resinous material, preferably 20 to 99%.

2.2 Contrary to the opinion expressed in the decision under appeal, it is not disputed in the appeal proceedings that the polyfunctional monomers (b) disclosed in (1), specifically the bis(ethylenglycol)phthalatedimethacrylate used in the worked examples of (1), fall within the group defined in Claim 1 of the patent-in-suit, i.e. that the group $-[R_1-O-C(O)-R_2-C(O)-O]_n-R_1-$ of (1) falls within the definition of "R" in that claim.

2.3 The ranges disclosed in (1) are not all based on the weight of the whole composition but partially on the weight of the resinous components only. A calculation of these ranges on the basis of the total weight of the composition, using various amounts of fibers has therefore been made. For example, with 20 and 65% of fibers, based on the total weight of the composition, which correspond to the limits of the preferred weight range of fibrous material disclosed in (1), page 7, line 4, the weight ranges of difunctional monomer (b), based on the total weight of the composition are 0.4-72% and 0.175-31.5% respectively, the weight ranges for the monofunctional component (c) are 8-79.6% and 3.5-

34.8% respectively, and the amounts of catalyst (d) are from 0.016-3.2% and 0.007-1.4% respectively.

2.4 From this analysis it is clear that at least the upper limits of the weight ranges disclosed in (1), calculated according to the patent-in-suit, fall within the weight ranges claimed therein, i.e. that the ranges disclosed in (1) and the ranges claimed overlap to a great extent. Moreover, it is not disputed, that the values calculated from the worked examples of (1) are all within the claimed ranges except for the catalyst concentration. However, Example 4 of (1) specifies a composition consisting of 45% glass fibers (a), 33% bis(ethylenglycol)phthalatedimethacrylate (b), 22% methylmethacrylate (c) and 0.55% (1% of the resinous components as in the worked examples of the patent-in-suit) of a catalyst (d), methyl ethyl ketone peroxide, which however in (1) is said to already initiate curing at room temperature, all these values falling within the ranges specified in Claim 1 of the patent-in-suit. There cannot, therefore, be any doubt that the examples read in context with the broader disclosure given in (1) teach the skilled person to use the whole of the disclosed ranges.

2.5 With respect to the viscosity range of 1 to 50 mPa.s, which is a characterising feature of the claims under consideration, the Board considers that the value of 10 mPa.s found by the Appellant, for a mixture of 30 parts of styrene (c) and 70 parts of bis(ethyleneglycol)-phthalatedimethacrylate (b), can fairly be taken as representative for at least all compositions (b) and (c) specifically disclosed in (1), because the example chosen is the one with the highest content of the more viscous component (b) of these compositions.

The Board therefore takes the view that the indication of the range of viscosities of 1 to 50 mPa.s, a parameter which was already inherent to known compositions, cannot be used to distinguish the claimed compositions from the known ones.

2.6 The Respondent's submission that the difference in the curing times mentioned in (1) and those applied according to the patent-in-suit is a strong indication of the novelty of the claimed compositions cannot be taken into account when assessing the novelty of the claimed composition. The Respondent obviously relies upon the earlier decision T 205/83 (OJ EPO 1985, 363) of this Board where it is said that a chemical product which can only be defined by process parameters instead of structural features may be regarded as novel if evidence is provided that the modified process results in a product having different properties.

However, in the present case the claimed composition as well as that disclosed in (1) are made by simply mixing the components, i.e. there is no difference in this respect. Moreover the claimed composition is not characterised by process parameters but by its components. In addition, the Respondent has not established that the known compositions cannot be cured in a short time by applying appropriate temperatures, nor has he demonstrated that there is a different characterising parameter which is responsible for this alleged difference in properties. Therefore the generally accepted principle has to be applied according to which mixtures of identical composition must inevitably possess identical properties.

2.7 In another earlier decision T 17/85 (OJ EPO 12/1986, 406), this Board has already held that if the preferred range of numbers in a citation anticipates a range claimed in an

application, the said claimed range cannot be regarded as novel, at least in cases where the values in the examples given in the citation lie just outside the claimed range and teach the skilled person that it is possible to use the whole of this range.

In the present case the ranges of numbers contained in Claim 1 under consideration and those disclosed in (1) overlap considerably and the claimed ranges include the values disclosed in the examples of (1). The Board considers that the principles applied previously to a single range of numbers used to define a chemical product also apply to the present case where four ranges of numbers are used to define the claimed compositions and where these ranges are combined in the same way as in the prior art document (1).

Consequently, in the Board's judgement the subject-matter of Claim 1 of the patent-in-suit lacks novelty. This finding applies equally to the molded articles claimed in Claim 5, because (1) also discloses the production of reinforced molded articles having high shock resistance. Therefore the main request must fail.

3. First auxiliary request

3.1 This request is based on the process Claim 4 (see paragraph I).

This process comprises

(i) forming mixtures according to Claim 1 of the main request,

(ii) providing these mixtures into a heatable mold

(iii) molding said mixtures at temperatures sufficient to cause the initiation of the co-reaction of (b) and (c) at a temperature not greater than 150°C and

(iv) completing said molding in less than 6 minutes from the time of initiation of said co-reaction.

3.2 In (1) there is disclosed a process of polymerisation of the compositions mentioned therein to produce reinforced plastic products. According to page 7, lines 7 to 10 of (1) any known process for preparing reinforced plastic products of the unsaturated polyester type may be applied without modification. However, a curing time of less than 6 minutes is not mentioned in (1). The claimed process, therefore, is novel.

3.3 There is also no specific disclosure in (1) relating to the use of a heatable mold. However, as the compositions of (1) according to the worked examples (see especially Example 1, page 7, paragraph B) are subjected to a "matrix polymerisation" at 60°C a heating device, which is to be regarded as a "mold" because the article so obtained must have had a defined shape, must have been used.

3.4 The Board is therefore satisfied that the only technical difference between the claimed process and the process disclosed in (1) consists in the upper limit of the reaction temperature. Whereas in (1) on page 6 it is stated that curing should take place at 20-100°C, preferably 50-90°C for 10 to 50 hours, followed by an additional heat treatment at 100-140°C, preferably 100-120°C for several hours, the claimed process provides for molding at a temperature below 150°C for not more than 6 minutes.

As the worked examples in the patent specification under consideration show, in order to achieve sufficiently rapid curing, it is necessary to perform the molding at a temperature of 130-140°C, which is well above the 100°C mentioned in (1). The molding time at 130°C is 5 minutes; at 140°C it is 3 minutes. It is easily derivable from these data that this reaction cannot be performed in less than 6 minutes if the reaction temperature is substantially lower than 130°C. Moreover, these data show that this reaction essentially follows the well known approximative rule according to which an increase in reaction temperature of 10°C reduces the reaction time by about a half. No additional heat treatment is mentioned in the patent specification under consideration.

3.5 Turning now to the specific disclosure of reaction time and temperature in (1), one finds in Examples 1 to 3 and 5 to 7 a curing time of 60°C for 20 hours, followed by a heat treatment of 2 hours at 120°C to complete the polymerisation. In these examples the catalyst concentration is 0.05 parts per 100 parts of resin, i.e. much less than the catalyst concentration used in the patent-in-suit. In Example 4 of (1) a catalyst concentration of 1 part per 100 parts of resin is used which corresponds to the amount used in the examples of the patent-in-suit. In this example, curing is performed for 35 minutes at room temperature, followed by heat treatment at 120°C for 2 hours, thus showing that the curing conditions mentioned on page 6 of (1) are not mandatory. This also follows from the wording of Claim 1 on page 14 of (1).

Since the curing time specified in (1) as well as the molding time specified in the process claim under consideration depend on the reaction temperature, the molding time defined in the patent-in-suit is not an

independent technical feature, but is closely related to the reaction temperature, provided that there is no change in the resinous composition to be cured.

3.6 Thus the technical problem to be solved by the claimed process, starting from (1) as the closest prior art, may be seen in avoiding the long curing times which were considered to be disadvantageous.

The solution to this problem was to increase the reaction temperature.

As already pointed out previously, a person skilled in the art, on the basis of his common general knowledge, is able to estimate the effect of temperature increase on reaction times by employing the rule that a temperature increase of 10°C approximately doubles the reaction rate. The above estimation shows that, starting from 60°C and 20 hours (1200 minutes) as used in the worked examples 1-3 and 5-7 of (1), a reaction time from 5 to 10 minutes at 130-140°C can be expected. Moreover, as previously mentioned the disclosure of (1) (see page 6 and the claim on page 14) shows that the curing conditions (time and temperature) are in no way critical. The curing temperatures mentioned in (1), page 6 cannot therefore be interpreted as a warning against the use of higher temperatures.

Therefore, the Board is satisfied that a skilled person, confronted with the problem as set out above, would inevitably have considered increasing the reaction temperature. Therefore this feature must be regarded as obvious and the Respondent's first auxiliary request cannot be allowed either. This position cannot be changed by the Respondent's argument that it was surprising that according to the claimed process no additional heat treatment is

necessary. On the one hand, this additional treatment is in no way excluded by the wording of the claim under consideration and, on the other hand, such an additional advantage cannot render a clearly obvious process unobvious.

4. Second auxiliary request

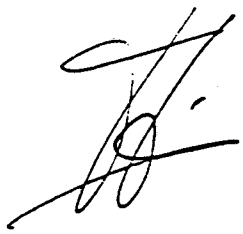
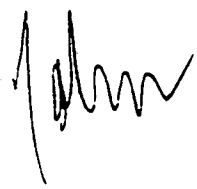
4.1 As a second auxiliary request the Respondent requests that the process claim according to the first auxiliary request be further limited to the molding methods referred to in the description on page 1 line 7. However, it was not disputed that these molding techniques are well known in the art. Injection molding for instance is known from (3). No specific adaption of these molding processes is necessary to use the compositions of (1), which were stated to be suitable for any known process without modification. Thus the selection of these processes out of the limited number of possibilities falls within the routine duties of the skilled person and does not involve an inventive step.

Therefore, the second auxiliary request must also fail.

Order

For these reasons, it is decided that:

The decision of the Opposition Division is set aside.

The patent is revoked.

The Registrar: The Chairman: