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Bezeichnung der Erfindung: Process for making emulsions of sequential polymer  
Title of invention: particles useful as opacifiers in coating or  
Titre de l'invention : impregnating compositions

Klassifikation / Classification / Classement : C08F 265/00

### ENTSCHEIDUNG / DECISION

vom / of / du 6 March 1990

Anmelder / Applicant / Demandeur : Rohm and Haas Company

Patentinhaber / Proprietor of the patent /  
Titulaire du brevet :

Einsprechender / Opponent / Opposant :

Stichwort / Headword / Référence :

EPÜ / EPC / CBE Articles 56, 123(2)

Schlagwort / Keyword / Mot clé : "Inventive step (denied) - obvious  
alternative - skilled man as a team - late  
filed claims not admitted because not clearly  
allowable under Article 123(2) EPC"

Leitsatz / Headnote / Sommaire

Europäisches  
Patentamt

Beschwerdekammern

European Patent  
Office

Boards of Appeal

Office européen  
des brevets

Chambres de recours



Case Number : T 51/88 - 3.3.3

D E C I S I O N  
of the Technical Board of Appeal 3.3.3  
of 6 March 1990

Appellant : Rohm and Haas Company  
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Decision under appeal : Decision of Examining Division 001  
of the European Patent Office  
dated 14 September 1987 refusing  
European patent application  
No. 84 301 489.5 pursuant to  
Article 97(1) EPC

Composition of the Board :

Chairman : F. Antony  
Members : C. Gérardin  
G. Paterson

## Summary of Facts and Submissions

- I. The European patent application No. 84 301 489.5, filed on 7 March 1984, claiming priority of 9 March 1983 from an earlier application in the United States of America and published on 19 September 1984 under the publication No. 119 054, was rejected by a Decision of the Examining Division dated 14 September 1987.

The rejection was based on a set of eleven claims of which Claim 1 reads as follows:

"A process for making an aqueous dispersion of water-insoluble core/sheath polymer particles comprising sequentially emulsion polymerizing at a temperature of from 10°C. to 100°C. in an aqueous medium containing a free radical initiator:

- (a) a core monomer system comprising one or more monoethylenically unsaturated monomers at least one of which monomers has an amine group and the amount of amine monomer in the core being sufficient to render the core swellable on at least partial neutralization thereof by acid comprises at least 5% by weight of the core monomer system; thereby forming dispersed core particles having an average diameter of from 0.5 to 1 micron, and
- (b) polymerizing in the presence of the dispersed core particles a sheath monomer system comprising at least one monoethylenically unsaturated monomer having no ionizable group to form a sheath on the core particles, any monoethylenically unsaturated amine in the sheath monomer mixture being present in an amount of no more than 10% by weight of the sheath monomer(s), the proportion of amine in the sheath

monomer mixture being less than 1/3 the proportion thereof in the core monomer mixture, the resultant core-sheath particles having an average diameter before acidification and swelling of from 0.07 to 4.5 microns, the relative amounts of core-forming monomer(s) and sheath forming monomer(s) being such that the ratio of the weight of the core to the weight of the total polymer in the resulting dispersed particles is from 1:4 to 1:100, said sheath being permeable to an aqueous acid and having a  $T_g$  of greater than 50°C, and

- (c) at least partially neutralizing with acid so as to swell said core and form particles which, when dried, contain a single void."

Claim 8 was an independent composition claim directed to an aqueous dispersion of water-insoluble core/sheath polymer particles and Claim 11 was a use claim of these compositions; all the other claims were dependent claims concerning specific embodiments of process Claim 1 (Claims 2 to 7) and composition Claim 8 (Claims 9 and 10).

II. The ground for this Decision was that the subject-matter of the application in suit did not involve an inventive step with regard to the teaching of the following documents:

- (1) EP-A2-22 633
- (2) FR-A-2 072 664.

In the Decision, it was stated that the skilled man faced with certain difficulties associated with the basic dispersions of polymer particles described in document (1) would have readily realised that their basic character resulted from the use of a base in at least stoichiometric amounts to neutralize the acid core polymer. Having knowledge of document (2) which also concerned the

manufacture of cellular material, he would have seen that the alternative means described there would provide a suspension without the said undesirable basic character.

III. A Notice of Appeal was lodged against this decision on 16 November 1987 with payment of the prescribed fee. The Statement of Grounds of Appeal was filed on 16 January 1988. The arguments presented by the Appellant in the Statement of Grounds, in the submission filed on 28 November 1989 in reply to a communication of the Board, and during oral proceedings which took place on 6 March 1990, can be summarised as follows:

In attempting to overcome the difficulties which arise when using the core/sheath polymers described in document (1), the skilled man would not have contemplated making any major change to the composition, since the slightest change in composition could result in a major change of properties. Having recognised that the acidity of the core and the subsequent addition of a volatile base for the swelling process were the cause of these difficulties, he had the choice between several solutions which did not involve compositional changes at all. There was thus no reason to consider document (2) which made only a very general, speculative and passing reference that polymers containing ionisable ionic groups may be swollen by water at pH values lower than 7, all the more as the polymers described therein were not even prepared by a suspension polymerisation process. Even if the user of the polymers known from document (1), who should be regarded as the skilled man in the present case, had come to the idea to consider the alternative structure mentioned in document (2), he would have been faced with coagulation problems, which the Appellant could only solve satisfactorily by using 3-(methacrylamido)propyl trimethyl ammonium chloride in the second step of the process as a comonomer with surfactant activity. This argument was

supported by experimental results submitted during the oral proceedings showing that in the absence of this particular compound coagulation occurred, making it impossible to obtain an acceptable core/sheath polymer dispersion.

On the basis of this argument, the Appellant filed two additional sets of claims during the oral proceedings as auxiliary requests. In the first set, it was specified in Claim 1 that in step (b) the sheath monomer system was polymerised in the presence of the dispersed core particles and the above specific monomer. In the second set, it was additionally specified in step (a) that the monomer with an amine group was selected from 3-(dimethylamino)propyl(meth)acrylamide and 2-(dimethylamino)ethyl(meth)acrylamide.

- IV. The Appellant requested that the impugned decision be set aside and that a patent be granted, as a main request, on the basis of the claims as considered by the Examining Division in its decision, or, as first and second auxiliary requests, on the basis of either of the two sets of claims as filed during the oral proceedings.

At the end of the oral proceedings, the Decision was announced that the appeal was dismissed.

#### Reasons for the Decision

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

#### Main request

2. The application in suit concerns a process for making an aqueous dispersion of polymer particles, a composition comprising such an aqueous dispersion and the use of such a

composition as an opacifier in coating and impregnating compositions. Such process, composition and use are generally known from document (1), which in the Board's view represents the closest prior art. The process described there comprises sequentially emulsion polymerising at a temperature from 10 to 100°C in an aqueous medium containing a free radical initiator, first a monoethylenically unsaturated core monomer containing a carboxylic group, optionally together with a hydrophilic monomer, so as to form dispersed core polymer particles which have an average diameter of from 0.05 to 1 µm and are swellable by neutralisation with a volatile base, then at least one monoethylenically unsaturated sheath monomer without ionisable group to form on the core particles a sheath polymer with an inflection temperature greater than 50°C, the sheath being permeable at 20°C to an aqueous volatile base selected from ammonia and amines, the resultant core/sheath particles having an average diameter before neutralisation and swelling of from 0.07 to 4.5 µm and the ratio of the weight of the core to the weight of the total polymer in the resulting dispersed particles being from 1:4 to 1:100, and at least partially neutralising these particles with a volatile base as above defined so as to swell said core and form particles which contain microvoids upon drying (Claim 1). In practice, the aqueous core/sheath heteropolymers are used as aqueous dispersions in coating compositions comprising film-forming polymers or binders, pigments and extenders; the swelling process is achieved by addition of the volatile base, which raises the pH to 8 to 12 or even more (page 58, line 27 to page 59, line 8). Although the resulting heteropolymer particles exhibit advantageous properties as opacifiers, the relatively high pH of their dispersion has a detrimental effect on some binders in that it affects their stability, like in the case of poly(vinyl acetate) latices, or interferes with curing reactions, like in the case of acid-catalysed thermosetting coatings.

In the light of this prior art, the problem underlying the application in suit may thus be seen in providing a process for preparing swellable core/sheath polymers, whose swelling process does not require the use of basic reactants, without impairing the opacifying properties thereof.

According to the application in suit, this problem is solved by polymerising at least one monoethylenically unsaturated monomer with an amine group in the first step and by at least partially neutralising the core/sheath heteropolymer with an acid in the final step.

In view of Example 2 of the application in suit, the Board is satisfied that the above defined technical problem is plausibly solved.

3. After examination of the prior art documents, the Board has reached the conclusion that the aforementioned solution is not disclosed in any of them and that the subject-matter of the application in suit is, therefore, novel. Since the issue of novelty has not been raised by the Examining Division, it is not necessary to consider this matter in detail.
4. It still remains to be examined whether the subject-matter of the application in suit involves an inventive step with regard to the teaching of the cited documents.
  - 4.1 The alternative structure claimed by the Appellant is suggested in document (2), which describes the two-step preparation of swellable polymer particles particularly suitable as opacifiers in coating compositions (Claim 1; page 1, lines 14 to 25). The first step comprises the dispersion or emulsion polymerisation of a mixture of core



monomers which may be more or less polar (page 8, lines 36 to 38; page 9, lines 21 to 24); when the reaction product is a strongly polar polymer having ionisable groups, the swelling is achieved by neutralisation with aqueous solutions having the appropriate pH (page 7, lines 11 to 37). The second step consists in the preparation of a vesiculated polymer by converting to a solid polymer a liquid medium containing dispersed therein the polymer product previously obtained (page 1, lines 20 to 25).

Regarding the polar groups more particularly, document (2) explicitly specifies that when the polymer contains acid groups, swelling occurs by addition of aqueous solutions of ammonia or low molecular weight amines; conversely, when the polymer contains amine groups, swelling occurs by addition of aqueous solutions with pH values lower than 7, such as aqueous solutions of chlorhydric or formic acid (page 7, lines 31 to 37). This teaching would clearly suggest that the nature of the polarity of the core polymer is not a feature which is essential for the opacifying properties of the heteropolymers; rather, that it can be changed, provided the polarity of the neutralising agent is changed accordingly and that the mere permutation of functional polar groups does not affect the swelling process, since the reaction remains basically the same.

Contrary to the Appellant's submission in the Statement of Grounds of Appeal (page 2, point 5.b), the teaching of document (2) cannot merely be regarded as a very general, speculative and passing reference about the possibility to swell polymers containing ionisable basic groups with acidic aqueous solutions. This document specifically mentions among the monomers suitable to confer a basic character to the core polymer, in particular vinylpyridine, 2-(dimethylamino)-ethyl methacrylate and 2-(tert-butylamino)ethyl methacrylate (page 7, lines 28 to 30), all monomers which are clearly envisaged in the application in

suit (compare description, page 8, lines 11 to 16) and whose use is even claimed in Claim 2.

In conclusion, although document (2) may be regarded as a broad teaching encompassing several embodiments, it explicitly mentions within the same class of compounds core polymers with amine groups and the neutralisation thereof with acidic aqueous solutions and core polymers with acid groups and the neutralisation thereof with aqueous solutions having pH values higher than 7. Therefore, in the light of the problem underlying the application in suit, core polymers with amine groups as claimed represent an obvious alternative.

- 4.2 The Board cannot accept the argument put forward by the Appellant during oral proceedings that the buyer and user of the prior art opacifying agents was the appropriate notional skilled person in this case, and that such a person, having noticed that there remained unreacted ammonia in the core/sheath heteropolymer dispersions, might have tried to remove the ammonia, but would not have thought of looking outside the field of acid core polymers, e.g. of changing the polarity of the core polymer, because this was beyond his skill as a skilled man.

In the first place, in the Board's view, the notional skilled man to be considered in the present case is not simply the user of the opacifying agent who first identifies the acidity of the core polymer as the cause of the shortcomings in the known coating products. The notional skilled addressee of the application in suit is clearly an industrial chemist skilled in the manufacture of coating compositions of the type in question as well as in the manufacture of components of such coating compositions, and could in reality consist of a team of more than one person.

In fact, the large excess of ammonia referred to by the Appellant would only be present in the case of total neutralisation of the acid core polymer with an excess of reactant, not in the general situation of partial neutralisation only. Besides, the three possibilities mentioned by the Appellant - i.e. neutralisation by addition of acid, removal of the base by dialysis, centrifugation or ion exchange, and drying out of the dispersion - would hardly be practical solutions as they would require an additional chemical step or a lengthy physical post-treatment of the polymer dispersion, thus, in any case, a modification of the process.

Furthermore, such a notional skilled man would be aware of the alternative suggested in document (2) and of the fact that such alternative does not require any fundamental modification of the process disclosed in document (1), but only the exchange of the functionalities of the core monomer and the neutralising agent. Thus, both for practical reasons as well as simply reasons, there was an incentive to change the polarity of the core polymer. As to the alleged difficulty, i.e. risk of coagulation in preparing a core polymer from amine functional monomers by emulsion polymerisation, document (2) does not mention any such difficulty, although the same monomers are mentioned (page 7, lines 27 to 30) as in the application in suit; it can thus be assumed that the skilled man would have been able to carry out such a process at the date of priority of the application in suit.

- 4.3 The argument repeatedly put forward by the Appellant according to which the notional skilled man would not have contemplated making any major change to the composition of the core/sheath polymers described in document (1), let alone making a fundamental change of the magnitude provided by the application in suit, cannot be accepted.

The comparison between the descriptions of document (1) and the application in suit shows that in reality such differences are not envisaged at all. If one excepts the amino groups which have replaced the acid groups as functional groups of the core monomers, there is identity between the various compounds which are used in the prior art process and the claimed process. This applies to the definition of nonionic monoethylenically unsaturated core and sheath monomers and polyethylenically unsaturated core and sheath monomers as well as to the various embodiments suitable to carry out the first stage of emulsion polymerisation, including the appropriate radical initiators and emulsifiers; likewise, there is correspondence between the general properties, especially the apparent second order transition temperature, as well as between the use as opacifying agents in coating and impregnating compositions. In fact, entire passages in the prior art document and in the application in suit are absolutely identical (compare in particular page 11, line 20 to page 18, line 11; page 22, line 19 to page 24, line 6; page 24, line 13 to page 25, line 25 of document (1) with page 8, line 25 to page 17, line 24 of the application in suit) and from the wording of Claim 1 the change of polarity of the core monomer and the choice of a neutralising agent of the opposite polarity appear to be the only modifications.

- 4.4 In conclusion, for the reasons given above, the solution claimed by the Appellant is obvious in the light of the problem to be solved and the advantageous properties of the products obtained could be expected to result from the change of polarity of the core monomers; hence the subject-matter of the application in suit does not involve an inventive step.

5. Claim 1 not being allowable, the same applies to the dependent process Claims 2 to 7, which merely represent preferred embodiments of the subject-matter of Claim 1 and thus fall with it. Under these circumstances, it is not necessary to consider the patentability of the subject-matter of the independent composition Claim 8 or dependent composition and use Claims 9 to 11.

#### Auxiliary requests

6. As mentioned above, both Claims 1 of the first and second auxiliary requests incorporate the use of 3-(methacrylamido)propyl-trimethylammonium chloride as monomer with surfactant activity in the preparation of the sheath polymer.

There is no specific requirement in the description of the application in suit regarding the conditions to carry out the second polymerisation step giving rise to the sheath polymer. The description indicates that "it is generally unnecessary to add emulsifier unless a polymodal product is desired, but in certain monomer/emulsifier systems for forming the sheath, the tendency to produce gum or coagulum in the reaction medium may be reduced or prevented by the addition of about 0.05 to about 0.5% by weight, based on sheath forming monomer weight, of emulsifier without detriment to the deposition of the polymer formed on the previously formed core particles" (page 12, line 31 to page 13, line 5). Moreover, there is no example of any specific emulsifier in the general part of the description; only Example 2, which illustrates the preparation of a sheath polymer in the presence of the core polymer prepared in Example 1, mentions the use of this particular monomer with surfactant activity.

In the Board's view, this feature was only disclosed in the application as filed in the context of the particular

process features of Example 2, not as a general feature in isolation. There is, thus, no adequate support for the generalisation made by the Appellant, so that the wording of both Claims 1 is objectionable under Article 123(2) EPC, and therefore is certainly not clearly allowable within the meaning of the decision T 153/85 of 11 December 1986 published in OJ EPO 1988, 001. Furthermore, the experimental comparative work by which the amended claims in both auxiliary requests are supported from the point of view of inventive step, was admittedly only carried out by the Appellant around a month before the oral hearing. Such a change in the centre of gravity of the alleged invention would require considerable further investigation and conceivably a further search, and in the Board's view, for this reason also, the auxiliary requests presented at the oral hearing constitute amendments which are not appropriate within the meaning of Rule 51(1) EPC, and again are certainly not clearly allowable. Following the principles set out in Decision T 153/85 regarding the filing of alternative claims at a late stage (points 2.1 and 2.2), the Board refuses thus to admit the two sets of claims constituting the auxiliary requests which were submitted during the oral proceedings.

For this reason, it is not necessary to consider any contribution to the inventiveness of the above new composition feature in the light of the Comparative Examples filed and the arguments presented during the oral proceedings.

Order

For these reasons, it is decided that:

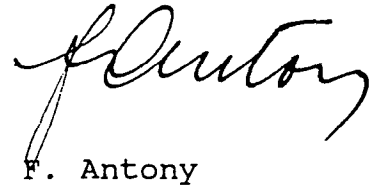
The appeal is dismissed.

The Registrar:

The Chairman:



M. Beer



F. Antony

CG  
hmf