BESCHWERDEKAMMERN DES EUROPÄISCHEN PATENTAMTS | BOARDS OF APPEAL OF THE EUROPEAN PATENT OFFICE | CHAMBRES DE RECOURS DE L'OFFICE EUROPEEN DES BREVETS

Internal distribution code:
(A) [ ] Publication in OJ
(B) [X] To Chairmen and Members
(C) [ ] To Chairmen

D E C I S I O N
of 27 August 1998

Case Number: T 0609/95 - 3.3.2

Application Number: 91302894.0

Publication Number: 0450922

IPC: A61L 15/00

Language of the proceedings: EN

Title of invention:
Method for production of fluid stable aggregate

Applicant:
NIPPON SHOKUBAI KOGYO CO. LTD.

Opponent:
-

Headword:
Fluid stable aggregate/NIPPON SHOKUBAI

Relevant legal provisions:
EPC Art. 52(1), 54, 56, 123(2)

Keyword:
"Novelty (yes)"
"Inventive step (yes): improved properties of the product resulting from unobvious modifications of the reaction parameters of the process for its preparation"
"Amendment (yes): notional amendment by the skilled person"

Decisions cited:
-

Catchword:
-

Europäisches Patentamt | European Patent Office | Office européen des brevets

Beschwerdekammern | Boards of Appeal | Chambres de recours

Case Number: T 0609/95 - 3.3.2

D E C I S I O N
of the Technical Board of Appeal 3.3.2
of 27 August 1998

Appellant: NIPPON SHOKUBAI KAGAKU KOGYO CO. LTD.
1-1, Koraibashi, 4-chome
Chuo-ku
Osaka-shi
JP-Osaka-fu 541 (JP)

Representative: Rees, David Christopher
Kilburn & Strode
20 Red Lion Street
GB-London WC1R 4PJ (GB)

Decision under appeal: **Decision of the Examining Division of the European Patent Office posted 27 February 1995 refusing European patent application No. 91 302 894.0 pursuant to Article 97(1) EPC.**

Composition of the Board:

Chairman: P. A. M. Lançon
Members: G. F. E. Rampold
M. B. Günzel

Summary of facts and submissions

I. European patent application No. 91 302 894.0, published as EP-A-0 450 922, was refused under Article 97(1) EPC by the decision of the examining division dated 27 February 1995. The decision was based on claims 1 to 23 for all designated contracting states filed on 27 December 1994.

Independent claim 1 read as follows:

"A method for the production of a fluid stable aggregate, which comprises mixing (A) 100 parts by weight of an absorbent resin powder possessing a carboxyl group, (B) 0.01 to 30 parts by weight of a cross-linking agent having at least two functional groups capable of reacting with the carboxyl group of the absorbent resin powder, (C) 0 to 50 parts by weight of water, and (D) 0 to 60 parts by weight of a hydraulic organic solvent in a high-speed stirring type mixer; the mixer having an inner surface formed substantially of a substrate (I) possessing a contact angle of not less than about 60° with respect to water and a heat distortion point of not less than about 70° under stirring conditions in which the leading-end of a stirring blade has a peripheral speed of not less than 600 m/min and thereafter completing the reaction of the adsorbent resin powder (A) with the cross-linking agent (B) under the condition that the total kinetic energy F added to the mixture during the reaction is satisfied by the following equation:

$$0 \leqq F \leqq 36{,}000 \text{ joule/kg},$$

and in which the kinetic energy Fa per minute added during the reaction is not more than 600 joule/kg."

Dependent claims 2 to 22 related to specific embodiments of the method claimed in claim 1.

Independent claim 23 read as follows:

"Substantially water-insoluble, absorbent, hydrogel forming, polymer fluid stable aggregate produced in accordance with the method of any of the preceding claims."

II. The stated ground for refusal was lack of inventive step.

The examining division found that citation (1), viz. GB-A-2 162 525, was already concerned with the preparation of aggregates of water absorbent resin particles in which the molecular chains near the surface were cross-linked. The aggregates in (1) were basically prepared in the same way as described in the present application, that is to say, by mixing and reacting an adsorbent resin powder possessing a carboxyl group with a cross-linking agent, preferably in a high speed stirring type mixer. There was no doubt that the cross-linked products were obtained in the prior art of (1) also in the form of aggregates as a result of the particular process for their preparation.

The difference between the claimed method for the preparation of the aggregates according to the application and the prior art of (1) was seen by the examining division essentially in the determination of certain ranges of suitable reaction conditions and operating parameters, which were merely the result of trial and error, and in the use of a particular type of mixer for carrying out the claimed process.

As the aggregates of the present invention showed also about the same properties and capabilities as those

described in (1), the examining division considered the subject-matter of the present application as an obvious alternative to the prior art of (1).

III. The appellants (applicants) lodged an appeal against the above decision and submitted a statement setting out the grounds for appeal within the time limit set in Article 108 EPC.

IV. During the oral proceedings, held on 27 August 1998, the board raised certain doubts as to the patentability of "product-by-process" claim 23, on the one hand, and the clarity of dependent claims 9, 10, 20 and 21, on the other. Moreover, it found that, on the basis of the experimental data provided in Table 1 of the application, the range of temperature required to induce cross-linking would have to be considered as an important technical feature which is essential to the proper implementation of the claimed invention and should therefore be incorporated into claim 1 to comply with the requirements of Article 84 in conjunction with Rule 29(1) and (3) EPC.

As a consequence of this, the appellants filed a revised statement of claim with an amended claim 1 as the sole independent claim reading as follows:

"A method for the production of a fluid stable aggregate, which comprises mixing (A) 100 parts by weight of an absorbent resin powder possessing a carboxyl group, (B) 0.01 to 30 parts by weight of a cross-linking agent having at least two functional groups capable of reacting with the carboxyl group of the absorbent resin powder, (C) 0 to 50 parts by weight of water, and (D) 0 to 60 parts by weight of a hydrophilic organic solvent in a high-speed stirring type mixer; the mixer having an inner surface formed substantially of a substrate (I) possessing a contact

angle of not less than about 60° with respect to water and a heat distortion point of not less than about 70°C **under stirring conditions in which the leading-end of a stirring blade has a peripheral speed of not less than 600 m/min** and thereafter completing the reaction of the adsorbent resin powder (A) with the cross-linking agent (B) **at a temperature in the range of 90° to 250°C** under the condition that the total kinetic energy F added to the mixture during the reaction is satisfied by the following equation:

$$0 \leqq F \leqq 36{,}000 \text{ joule/kg}$$

and in which the kinetic energy Fa per minute added during the reaction is not more than 600 joule/kg.

Claim 1 is followed by dependent claims 2 to 17, all relating to specific embodiments of the method of claim 1.

V. The appellants submitted in support of their appeal both in the written procedure and at the oral proceedings essentially the following arguments:

The refusal of the application by the examining division was based upon a basic misunderstanding of the teaching of citation (1). In particular, by the process defined in claim 1 of (1) the particles of the carboxyl-containing, water-absorbing resin were subjected to a cross-linking agent which affected the surface layers. When the particles were heated cross-linking occurred in the surface areas and the particles remained discrete.

According to the process of claim 10 of (1) the discrete particles with surface-cross-linking were then treated with an aqueous liquid in the form of a binder. The binder affected the surface of the particles and

when these were stirred, the particles adhered together by a mechanical adhesion. The adhered particles represented an aggregate.

However, when water was added to the aggregated particles connected by mechanical adhesion, separation and degradation occurred upon swelling of the particles by water, because the inter-particle bond was relatively weak. Such aggregated particles were not fluid stable.

According to the present invention, a cross-linking agent was added to the particles of the carboxy-containing , water-absorbing resin and the particles were subjected to mixing/stirring. The cross-linking agent penetrated the surface layer of the particles and the particles were stuck together by adhesion. The adhered particles represented an aggregate. This was effected as a result of the specific operating parameters of the mixing step.

When these aggregated particles connected by mechanical adhesion were then submitted to the heating step, inter-particle cross-linking occurred between the particles. When water was added to the aggregated particles according to the application, which had inter-particle cross-linking, the aggregated structure was maintained, even on absorption of water. The aggregates of the present invention could therefore be described as fluid-stable and showed improved properties, in particular, as far as their particle size distribution, their swelling rate and their liquid permeability were concerned.

The production of fluid stable aggregates according to the invention possessing improved characteristics was possible by the claimed combination of particular process parameters as defined in present claim 1,

including the particular properties of the inner surface of the mixers used, the stipulated formulation of the raw material, the stipulated operating conditions of the mixer, and finally the particular range of kinetic energy stipulated.

VI. The appellants requested that the impugned decision be set aside and a patent be granted on the basis of claims 1 to 17 as filed during oral proceedings before the board and a consequentially amended description.

Reasons for the decision

1. The appeal is admissible.

2. Amendments

2.1 The amended claims are acceptable as complying in formal respects with the provisions of Article 123(2) EPC. Furthermore, such claims are adequately supported by the description as required by Article 84 EPC.

In particular, claim 1 is based on originally filed claim 1 with the following amendments:

(i) the range of the reaction temperature required to induce cross-linking, namely "a temperature in the range 90° to 250°C", has been inserted into claim 1 from page 20, line 5 of the originally filed documents;

(ii) the wording "under the stirring conditions of not less than 600m/min of a peripheral speed of a leading-end of a stirring blade" has been amended from a linguistic point of view to improve readability and comprehensibility so as to read

"under stirring conditions in which the leading-end of a stirring blade has a peripheral speed of not less than 600m/min".

Dependent claims 2 to 6 correspond to originally filed claims 2 to 6;

Dependent claim 7 corresponds to originally filed claim 9;

Dependent claims 8 to 10 correspond to originally filed claims 13 to 15;

Dependent claims 11 and 12 correspond to originally filed claims 17 and 18;

Dependent claims 13 to 16 correspond to originally filed claims 20 to 23;

Dependent claim 17 corresponds to originally filed claim 26.

2.2 The consequential amendments to the description on pages 5, 19 and 20 are also acceptable.

2.3 In the second paragraph on page 18 of the description as originally filed reference is made to four equations by stating: "The time of completion of the reaction is the time which satisfies the following equations (a-1), (a-2), (b-1), (b-2)".

For example, equation (a-1) reads as follows:

$$30 \leqq \frac{(100 + R)}{100} \times \frac{Q}{P} \times 100 \leqq 95$$

wherein P is the absorption capacity of absorbent resin powder (A) using physiological saline solution, Q is

the absorption capacity of the reaction product using physiological saline solution, and R is the amount, in parts by weight, of cross-linking agent (B) to be used based on 100 parts by weight of absorbent resin powder (A).

From a comparison of the above statement with the respective equations it appears evident to a person skilled in the art that this statement is clearly defective and inconsistent with said equations for at least two reasons:

(i) none of the equations (a-1) to (b-2) shown in the description contains a coefficient or a variable to which the unit "time" could be attributed;

(ii) under given conditions for the reaction the only variable appearing in the above-mentioned equations is Q representing the absorption capacity of the reaction product expressed in g/g.

Consequently, the above statement has been amended during oral proceedings so as to read:

"The reaction may be considered as completed when Q reaches a value so that one of the following equations (a-1), (a-2), (b-1) and (b-2) is satisfied."

In the board's judgment, the proposed amendment would be regarded by the skilled reader after a closer study of the application documents as clearly implied by the disclosure of the application as filed, since the absorption capacity of the product Q is the only coefficient which under the predetermined conditions of the reaction may vary to satisfy the above equations.

The board takes the position that where a drafting defect or inconsistency in an application would be

evident to a reader skilled in the art, the person to whom the application is addressed, it is reasonable to suppose that he would, in the light of the content of the application, attempt to formulate a notional amendment which would enable him to make sense of what he reads, and to the extent that the amendment might be said to leap to the mind of the reader, although perhaps only after close study of the document, it can be regarded as implicit in the application and would not contravene Article 123(2) EPC, if effected in practice.

In the light of the above considerations, the board adopts the amendment proposed by the appellants as acceptable under the terms of Article 123(2) EPC.

3. The problem and the solution

3.1 Citation (1) discloses a water-absorbing agent composed of a water-absorbing resin powder having cross-linked the molecular chains near its surface. This water-absorbing resin powder is essentially produced by mixing (A) 100 parts by weight of a resin powder possessing a carboxyl group with 0.001 to 10 parts by weight of a polyhydric alcohol, 0.01 to 8 parts by weight of a hydrophilic organic solvent and 0 to 8 parts by weight of water, and heating the mixture at a temperature of at least 90°C to react the water-absorbing resin powder with the polyhydric alcohol and thereby to induce cross-linking of the molecular chains near the surface of the resin (see especially page 2, line 12 to page 3, line 52, Example 1).

According to a preferred embodiment of the prior art of (1) the water-absorbing resin powder with surface cross-linking, which is obtained by the manufacturing process described above, is subsequently subjected to agglomeration or size enlargement by the use of an

aqueous liquid serving as a binder. This binder affects the surface of the particles and when these are strongly stirred, the particles adhere together. The adhered particles undoubtedly represent an aggregate (see especially page 4, line 4 to page 5, line 40, Example 6). In aggregates of this type the particles adhere together by mechanical adhesion.

The method of preparing such aggregates, designated products (III) in citation (1), including both the steps of cross-linking and water agglomeration, represents the state of the art in (1) which comes closest to the claimed subject-matter in the application.

Although the method used in (1) comprising cross-linking of the absorbent resin powder followed by water agglomeration has the advantage of allowing the absorbent resin powder to acquire improved handling as a particulate substance, the agglomerates obtained by the method according to (1) still show certain properties capable of improvement, in particular as far as their particle-size distribution, their swelling rate and their liquid permeability are concerned.

3.2 Given this closest state of the art, the technical problem underlying the present application may therefore be seen as that providing a modified process allowing the preparation of an absorbent resin showing the desired advantages such as an improved swelling rate and improved liquid permeability in addition to a decrease of fine powder content.

As is emphasised in the present application, water-absorbing resins having these properties and capabilities are advantageously usable as absorbents in sanitary articles such as sanitary napkins and disposable diapers and are also suitable for a wide

variety of products such as coagulants for sludge, dew-drop proofing agents for building materials, and water retaining agents or desiccants for agriculture and horticulture (see especially page 21, lines 1-7).

3.3 The appellants propose to solve this problem by the process of present claim 1 which affords, as a result of certain specific reaction conditions and operating parameters used in the said process, an absorbent resin (polymer) with a high proportion of fluid-stable aggregates in the range of from 60% (see Table 1, Example 2) to 100% (see Table 1, Example 7). Fluid-stable aggregates within the meaning of the present invention are formed by having a plurality of the resin powders (A) inter-particle cross-linked by co-valent bonds with the cross-linking agent (B).

During the appeal proceedings the appellants have submitted that, if the manufacturing process disclosed in (1) is carried out, a certain proportion of the product may already be fluid stable aggregates. However, apart from the fact that the presence of such fluid stable aggregates is neither disclosed nor suggested in (1), their proportion in (1) is far too small to provide products with the desired improved properties and capabilities.

3.4 In fact, the comparison of the particle size distribution given in Table 1 on pages 30 and 31 of the present application for a representative number of products prepared according to the process of the invention with the particle size distribution given in Table 2 on page 10 of (1) for a product prepared according to the technique disclosed in (1), including both the cross-linking and water agglomeration step, demonstrates that the products of the invention can be prepared with a significantly decreased quantity of fine particles, determined as the weight percent of

particles which passed through the 100-mesh wire gauze, as compared to the product obtained in (1);

see present application, Example 1: 8%; Examples 2, 3, 4: 6%, Example 5: 9%; *vs.* Example 6, water absorbing agent (5) in (1): 13.3%.

As is convincingly explained in the description (see especially page 20, line 26 to page 22, line 12), the absorbent resin obtained by the method of the present invention contains abundantly fluid stable, tenacious aggregates of a structure sparingly disintegrable on absorption of liquid and, as a result of this, exhibits notably improved liquid permeability and an increased rate of fluid uptake (swelling rate), as compared with known water-absorbing resins containing only a minor proportion of fluid stable aggregates.

In view of the foregoing, the board is satisfied that the stated technical problem has been solved in its entirety by the claimed process.

4. *Novelty*

4.1 The method claimed in the application for preparing an absorbent resin having a high proportion of fluid-stable aggregates involves two distinct procedural steps:

(i) in the first step a cross-linking agent (B) is added to the absorbent resin powder (A) and the particles are subjected to mixing/stirring using a particular type of mixer and specific operating parameters; the cross-linking agent penetrates the surface layer of the particles and the particles are stuck together to form aggregates by mechanical adhesion;

- the mixer used in this step has an inner surface formed of a substrate which possesses a contact angle of not less than 60° with respect to water and a heat distortion point of not less than 70°C;

- the stirring conditions are such that the leading-end of a stirring blade has a peripheral speed of not less than 600 m/min;

(ii) in the second step the aggregates from step (i) are subjected to heating to induce cross-linking whereby the

- total kinetic energy added to the mixture in this step is limited to 36,000 joule/kg maximum, while the

- kinetic energy added to the mixture per minute is limited to 600 joule/kg maximum.

4.2 In the present application (see especially page 6, line 12) and in citation (1), as well (see especially page 4, lines 42-43), the use of mixers designated "Turbulizer" or "Sand Turbo" is recommended in order to achieve agglomeration of the particles by mechanical adhesion. The appellants have, however, plausibly submitted during the oral proceedings that the particular inner surface of the mixer used in the present application does not belong to the standard equipment of such type of mixers but was specifically designed and ordered to enable completion of the present invention.

In the absence of any evidence to the contrary and any possibility of verifying the particular material used for the inner surface of the mixers mentioned in (1) in their standard version, the board cannot but accept the

appellants' submissions as a basis for the decision to be taken in the present proceedings. As (1) is also entirely silent about the material of the inner surface of the mixer used, the board takes the view, that this feature, which appears from a technical point of view particularly relevant to the proper implementation of the invention, is not derivable from the cited state of the art.

4.2 In conclusion, neither the particular inner surface of the mixer and the exact stirring conditions used in the first step nor the limitation of the kinetic energy added to the mixture during cross-linking to the particular values specified in the second step of the claimed process are disclosed in citation (1) or in any other document cited in the search report. The processes of present claim 1 and dependent claims 2 to 17 are accordingly deemed to be novel within the meaning of Article 54(1) EPC.

5. *Inventive step*

5.1 As the appellants themselves have submitted, a minor proportion of the product obtained in (1) may be a fluid stable aggregate. Although this is neither disclosed nor suggested in (1), it appears, in the board's judgment, nevertheless reasonable to assume that the skilled person would have tested the product obtained in (1), for example, for absorption capacity, swelling rate or particle size distribution and that, in doing so, he would have realised the presence of fluid stable aggregates in the said product and would also have recognised the advantageous properties of such aggregates. This is particularly so, since such fluid stable aggregates would appear to be readily separable from the remaining product by their increased stability vis-à-vis the influence of fluids.

The relevant question to be answered under Article 56 EPC appears therefore whether the skilled person having studied the relevant state of the art and being guided by the technical problem would have been aware, from his common general knowledge and also his familiarity with the related art, what kind of modification of the reaction conditions and operating parameters of the known process could make the desired increase in the proportion of fluid stable aggregates in the product available. As has already been noted in point 3.4 (*supra*), the improved properties and capabilities of the water adsorbing agent produced by the process of the invention undoubtedly have their origin in the higher proportion of fluid stable aggregates in the product which in turn is a result of the specific reaction conditions and operating parameters which distinguish the invention from the closest state of the art.

5.2 The appellants have convincingly argued during the oral proceedings that the properties of the inner surface of the mixing apparatus used (see points 4.1(i), 4.2 *supra*) represent essential operating conditions in order to produce the required fluid stable aggregates, although the exact reasons for this are admittedly not fully understood.

In any case, (1) and the other documents cited in the search report, as well, are entirely silent about the relevance of the material of the inner surface of the mixing apparatus to the properties of the water adsorbing agent produced in the said apparatus. Consequently, the person skilled in the art could not derive or glean from this state of the art any clue as to the substances usable as the substrate for the formation of the inner surface of the mixer, let alone as to the properties of such substances required to

achieve the desired high proportion of fluid stable aggregates.

5.3 There is also nothing in (1) or any other cited document suggesting to a person skilled in the art that the total kinetic energy added during the cross-linking reaction as well, as the kinetic energy added per minute should be limited to a certain value in order to promote interparticle cross-linking of a plurality of the absorbent resin powder through the reaction of the absorbent resin powder with the cross-linking agent and thereby to increase the proportion of the fluid stable aggregates in the product.

5.4 Notwithstanding the board's finding that the reaction parameters mentioned in points 5.2 and 5.3 (*supra*), when taken individually, are inventive, the particular combination of process parameters, which distinguish the process of present claim 1 from the closest prior art of (1), similarly cannot be considered obvious to a person skilled in the art faced with the solution of the stated technical problem, and more specifically, with the preparation of fluid stable aggregates.

This combination includes:

- the particular properties of the inner surface of the mixing apparatus;

- the specific operating conditions of the mixer (the relevance of this parameter to the invention is clearly derivable from control experiment 2 in Table 1 of the application); and

- the particular range of kinetic energy added during cross-linking.

5.5 Having regard to the foregoing, the board considers that the process of present claim 1 involves an inventive step. The non-obviousness of claim 1 also imparts an inventive step to dependent claims 2 to 17 relating to specific embodiments of the process according to claim 1.

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 grant a patent in the following version:

Description: pages 1-4, 6-17, 21-31, as originally filed
pages 5 and 18-20, as received during oral proceedings of 27 August 1998

Claims: 1-17, as received during oral proceedings of 27 August 1998

Drawing(s): Sheet 1/1 as originally filed.

Der Geschäftsstellenbeamte: Der Vorsitzende:

P. Martorana P. Lançon