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D E C I S I O N
of 25 October 1995

Case Number: T 0424/93 - 3.4.2

Application Number: 85302719.1

Publication Number: 0162577

IPC: G03G 9/08

Language of the proceedings: EN

Title of invention:
Process for producing toner for electrophotography

Patentee:
Hitachi Chemical Co., Ltd.

Opponent:
Canon Inc.

Headword:
-

Relevant legal provisions:
EPC Art. 56, 84, 123(2)

Keyword:
"Inventive step - (second auxiliary request) yes"

Decisions cited:
-

Catchword:
-

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Case Number: T 0424/93 - 3.4.2

D E C I S I O N
of the Technical Board of Appeal 3.4.2
of 25 October 1995

Appellant: (Proprietor of the patent)
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Decision under appeal: **Decision of the Opposition Division of the European Patent Office dated 12 February 1993, posted on 4 March 1993, revoking European patent No. 0 162 577 pursuant to Article 102(1) EPC.**

Composition of the Board:

Chairman: E. Turrini
Members: W. W. G. Hofmann
B. J. Schachenmann

Summary of Facts and Submissions

I. The Appellant (Proprietor of the patent) lodged an appeal against the decision of the Opposition Division on the revocation of the patent No. 0 162 577 (Application No. 85 302 719.1).

Opposition had been filed against the patent as a whole and based on Article 100(a) EPC.

The Opposition Division had held that the grounds for opposition mentioned in Article 100(a) EPC prejudiced the maintenance of the patent, having regard to the documents

D1: JP-A-58-50545 and an English translation thereof,
D2: GB-A-2 091 435, and
D3: GB-A-2 112 538.

During the appeal procedure, the Respondent (Opponent) furthermore cited

D4: JP-A-57-207259 and an English translation thereof.

II. Oral proceedings were held, at the end of which the Appellant requested that the decision under appeal be set aside and the patent maintained in amended form

as a main request:
on the basis of Claims 1 to 15 ("main request"), filed with the letter of 22 September 1995,

as a first auxiliary request:
on the basis of Claims 1 to 15 ("first auxiliary request"), filed with the letter of 22 September 1995, and

as a second auxiliary request:
on the basis of Claim 1 ("second auxiliary request"), submitted at the oral proceedings of 25 October 1995, and Claims 2 to 15 as filed for the main request with the letter of 22 September 1995.

The Respondent requested that the appeal be dismissed.

III. The wording of Claim 1 on file at the time of the present decision reads as follows:

Main request:

"1. A process for producing a toner for electrophotography by emulsion polymerizing one or more polymerizable monomers stably dispersed by emulsification in the presence of a colorant and/or a magnetic powder to prepare a principal resin component, and then effecting the coagulation of the resulting polymerization liquid, to finally obtain imperfectly spherical particles,
characterized by
(a) conducting the coagulation by the addition of a coagulating agent while subjecting the coagulating mixture to heat treatment at a temperature at least 24°C higher than the glass transition point of the principal resin component and not higher than 150°C so as to increase the bulk density of the coagulated particles and/or
(b) after coagulation by the addition of a coagulating agent, subjecting the coagulated particles to heat treatment at a temperature at least 24°C higher than the glass transition point of the principal resin component and not higher than 150°C so as to increase the bulk density of the coagulated particles."

First auxiliary request:

"1. A process for producing a toner for electrophotography by emulsion polymerizing one or more polymerizable monomer stably dispersed by emulsification in the presence of a colorant and/or a magnetic powder to prepare a principal resin component, adding a coagulating agent to the resulting polymerization liquid so as to obtain coagulated toner particles, and subjecting the coagulating mixture and/or the coagulated particles to a heat treatment at a temperature not higher than 150°C so as to increase the bulk density of the coagulated particles,
characterised in that coagulation is conducted after the principal resin component has been produced by the polymerization step and that the heat treatment is conducted at a temperature at least 24°C higher than the glass transition point of the principle resin component."

Second auxiliary request:

"1. A process for producing a toner for electrophotography by emulsion polymerizing one or more polymerizable monomers stably dispersed by emulsification in the presence of a colorant and/or a magnetic powder to prepare a principal resin component, adding a coagulating agent to the resulting polymerization liquid so as to obtain coagulated principal resin particles, and subjecting the coagulating particles and/or the coagulated particles to a heat treatment at a temperature not higher than 150°C so as to increase the bulk density of the coagulated particles, the polymerisation being continued until the conversion reaches at least 99% and coagulation being conducted after the principal resin component has been produced by this conversion and the heat treatment being

conducted at a temperature at least 24°C higher than the glass transition point of the principal resin component."

According to all requests, Claims 2 to 15 are dependent on Claim 1.

IV. The Appellant essentially argued as follows:

D1 teaches neither conducting the coagulation after the resin has been prepared, nor the use of a coagulating agent, nor a temperature range for the heat treatment from 24°C above the glass transition temperature (Tg) to 150°C so as to increase the bulk density of the coagulated particles. There is nothing in D1 or any of the other cited documents which could suggest this combination of process features. The improvement of the properties of the produced toner achieved by the process according to the patent in suit is demonstrated by comparative tests. The potassium persulfate added according to D1 is not a coagulating agent but a polymerization initiator.

D3 is concerned with producing core-and-shell particles and aims at obtaining highly spherical particles, which should be contrasted with the deliberately imperfectly spherical particles of the present invention. A person skilled in the art would consider modifying the process of D1 by means of the features of D3 in order to achieve highly spherical core-and-shell particles, but not for achieving imperfectly spherical particles. Moreover, the process features of D3 do not correspond to those of the present invention. The process of D3 does not provide what a skilled person would understand to be "effecting the coagulation of the polymerization liquid" in which "a principal resin component" has been prepared. It is clear from the text of D3 that the objective is to avoid

agglomeration of the core particles with other core particles and of shell particles with other shell particles whilst securing the growth of spherical shells on individual core particles. Moreover, the shell particles could not be regarded as the "principal resin component" which is specified in Claim 1. Regarding the heating step, D3 mentions a sintering step after formation of the shell, at a temperature between Tg+5 and Tg+20, and states that the temperature is very crucial; it thus teaches against the adoption of step (b) of present Claim 1.

V. The Respondent's arguments may be summarized as follows:

Claim 1 (main request) is unclear since the expression "imperfectly spherical particles" gives no indication as to what extent the produced toner particles are "imperfectly spherical". Because of its obscurity, this feature also cannot provide a difference from the prior art. The fact that an imperfectly spherical shape improves the cleaning property is, moreover, known from D4.

No invention can be seen in the fact that according to Claim 1 the polymerization and the coagulation of the polymerization liquid are carried out in two separate steps since this follows from the skilled person's basic experimental knowledge and the additional hints from D3. D3 discloses core-and-shell toner particles. However, Claim 1 of the patent in suit does not exclude the production of core-and-shell toner particles (eg in Example 27, particles of low molecular weight polypropylene are coated with polymer prepared by emulsion polymerization). The shell polymer particles according to D3, which are emulsion polymerized in the presence of a colorant and coagulated by the addition of a coagulating agent on the surface of the base

particles, may constitute the principal resin component in the sense of Claim 1 since the volume ratio between shell and core may rise up to a value of 10 and since they provide important properties for the toner material.

Adding a coagulating agent is an immediately evident option for the skilled person when the coagulation of particles is intended. According to D3, flocculation, ie coagulation, can be accomplished by the addition of salts or acids. Moreover, potassium persulfate (KPS) which is added to the emulsion according to D1, may be considered not only as an initiator but also as a coagulating agent.

The claimed temperature range of 24°C above Tg to 150°C is not originally disclosed as a general feature; the general disclosure only refers to 25 to 60°C above Tg. Moreover, this choice of the heat treatment temperature corresponds to what is chosen in D1 and approximately in D3, and also does not give rise to a surprising effect.

Reasons for the Decision

1. The appeal is admissible.

2. *Main request*

Claim 1 according to the main request contains the feature that "imperfectly spherical particles" are obtained. The term "imperfectly spherical particles" lacks a clear definition. The shape of real objects is never "perfect" and, on the other hand, there is also no clear borderline between completely non-spherical and imperfectly spherical particles. More or less all particles might in some way be referred to as "imperfectly spherical". There is not the slightest

indication in the prior art that the above-mentioned term might have a commonly agreed meaning. Thus, contrary to the Appellant's argument, the Board is not convinced that the term would have a well-defined meaning for a person skilled in the art of polymer dispersions.

The further feature of Claim 1, that monomers are emulsion polymerized "and then" coagulation is effected, also lacks clarity. The polymerization of the monomers in emulsion is a continuously proceeding process, and the above-mentioned expression does not unambiguously indicate at what stage of polymerization the coagulation should be performed.

Claim 1 according to the main request therefore lacks clarity in the sense of Article 84 EPC.

The main request is therefore not allowable.

3. *First auxiliary request*

Certainly, Claim 1 according to the first auxiliary request no longer contains the feature "imperfectly spherical", but it still does not unambiguously define the polymerization stage at which coagulation should be performed. The modified wording "after the principal resin component has been produced" insofar does not go beyond the expression "and then" used in Claim 1 according to the main request.

For these reasons, Claim 1 according to the first auxiliary request also lacks clarity in the sense of Article 84 EPC. The first auxiliary request is therefore nor allowable.

4. *Second auxiliary request*

4.1 The Respondent has argued that the range of 24°C higher than the glass transition point Tg to 150°C for the heat treatment temperature (contained in Claim 1 according to all three requests) was not originally disclosed. However, Claim 5 and the second paragraph of page 22 of the original application already relate quite in general to a continuous temperature range for the heat treatment reaching from Tg to 150°C, and Examples 10, 12 and 15 use a temperature of 100°C for treating a principal resin having a Tg of 76°C, which means a heat treatment temperature of 24°C above Tg. Since it is thus originally disclosed to append the lower limit of the temperature range to Tg of the resin component concerned (see also the lower limit of 25°C above Tg mentioned in Claim 6 and on page 22, lines 27/28), the Board sees no reason why the range should not be restricted by shifting the lower limit - while maintaining the idea of making it dependent on Tg - from 0°C above Tg to a value of 24°C above Tg, in correspondence with specific examples.

Further amendments with respect to the granted Claim 1 relate to the following added features:

"Emulsion polymerizing" is disclosed in the Examples 1 to 15, 27, 29, 31 to 34 (see eg the original page 33, line 23; page 35, line 2; page 36, line 2; page 40, lines 3/4; etc.).

"Stably dispersed" is disclosed on page 13, line 14 and page 14, lines 12/13.

The addition of a coagulating agent to the resulting polymerization liquid so as to obtain coagulated principal resin particles is eg disclosed on page 20, line 24, to page 21, line 5.

The alternative "subjecting the coagulating particles and/or the coagulated particles to a heat treatment" corresponds to the alternatives (a) and (b) of the granted Claim 1 and is based on the original page 22, lines 6 to 17 (in correspondence with the examples).

The conversion until a rate of at least 99% is reached, and coagulation after this conversion, is disclosed on page 20, lines 24 to 28; page 9, lines 25/26; and in the examples which, in the respective descriptions of the coagulation step, clearly refer to the use of the polymerization liquid obtained as described in the respective preceding paragraphs and having led to a certain percentage of conversion.

The temperature limit of at least 100°C additionally introduced into Claims 2 and 3 is disclosed in the Examples 3, 5, 6, 10, 12, 15, 27, 29, 31 to 34.

The above-mentioned amendments to Claim 1 only restrict the protection conferred.

The amendments therefore meet the requirements of Article 123(2) and (3) EPC.

4.2 Novelty

4.2.1 D1 (see in particular Claim 1; page 5, lines 1 to 3; page 7, lines 4 to 23; and Examples 1 to 3 on pages 8 to 11 of the English translation) describes a process for producing a toner for electrophotography by emulsion polymerizing one or more polymerizable monomers

dispersed by emulsification in the presence of a colorant to prepare a principal resin component. The statement on page 7, lines 15 to 18, that the particles are combined with each other, can only be understood in the sense that there is coagulation of the principal resin particles. As is described in the examples, at the time that the particles are coagulating, the solution is heated up to 90°C (which is thus lower than the upper limit of 150°C set in Claim 1 of the patent in suit).

The percentage of conversion reached for the polymerization is not explicitly mentioned in D1.

There is no indication in D1 that - as is the case according to the present Claim 1 - the coagulation is conducted after the principal resin component has been produced to a conversion rate of at least 99%. On the contrary, the only statement of D1 relating to coagulation (page 7, lines 15 to 17) indicates that the particles are combined with each other "as the polymerization proceeds", which does certainly not point to a combination of particles after the polymerization has been completed up to 99%.

In view of this clear difference of the claimed process from the teaching of D1, the remaining differences put forward by the Appellant, but contested by the Respondent, are of minor importance.

The Respondent argued that Claim 1 does not exclude coagulation during the polymerization step. However, in the view of the Board, the indication in Claim 1 that "stably dispersed" monomers are polymerized, must be understood as excluding major changes in the particle distribution of the emulsion, contrary to what occurs according to D1. An addition of a coagulating agent is not mentioned in D1, and the two sentences on page 7,

lines 18 to 23, indicating that the particle size could be controlled by selection of the amount of surfactant and stabilizer and of the agitation speed, rather point to the fact that it is these factors which are responsible for the "combination" of the particles. It cannot be fully excluded that the potassium persulfate added as an initiator implicitly leads to the presence of salts capable of acting as coagulating agents. However, such contribution would not be decisive for the coagulation of the emulsion and would not form an addition of coagulating agent to which the functional feature "so as to obtain coagulated principal resin particles" could be attributed.

According to D1, a temperature of 90°C is used for the heat treatment, however no data are given regarding Tg of the chosen resins, so that the difference between heat treatment temperature and Tg remains open. At the oral proceedings, the Respondent submitted calculations intended to show that the monomer mixtures used in Examples 1 and 2 of D1 resulted in polymers having Tg values more than 24°C below 90°C. The Appellant objected to the submission of these calculations so late in the appeal procedures. No reason was given by the Respondent why these calculations had not been submitted earlier. In fact, the question whether D1 disclosed a heat treatment temperature more than 24°C higher than Tg had been open since the filing of new claims by the Appellant with the letter of 7 July 1993. The Board would consider it unjust to admit such calculations into the proceedings at a time too late for a thorough examination by the Appellant. Moreover, as is apparent from the reasons given above in this paragraph and in paragraphs 4.3.2 and 4.3.3 below, neither novelty nor inventive step depend decisively on the implicit Tg values of the polymers according to D1.

Thus, the Board considers the Respondent's allegation, that D1 implicitly discloses a heat treatment temperature of more than 24°C above Tg, to be still unproven. This question may remain open.

4.2.2 D3 describes a process for producing a toner for electrophotography (see title and abstract). For preparing a shell of the toner particles, one or more polymerizable monomers, stably dispersed by emulsification in the presence of a colorant, are emulsion polymerized (see in particular page 4, lines 37 to 61; page 6, lines 25 to 41; page 9, lines 53 to 59).

According to D3, a second polymerization liquid including another dispersed resin component (base particles) is prepared and mixed to the first polymerization liquid, to which combined polymerization liquid a coagulating agent is added so as to obtain "flocculation" of the shell-polymer particles on the surface of the base particles, which flocculation thus occurs after polymerization (see page 8, line 56, to page 9, line 22). While it is not excluded in present Claim 1 that a second polymerization liquid might be prepared and mixed with the first one, the mixture then being called "resulting polymerization liquid", the Board holds the view that obtaining "coagulated principal resin particles" is distinctly different from flocculation of shell resin particles on the surface of base resin particles. In the view of the Board, coagulation of the "principal resin particles" (ie the same particles as those prepared by the emulsion polymerization step mentioned precedingly in Claim 1) means that these particles are coagulated with each other, while the flocculation on the surface of the base particles (small shell particles on much larger base particles, cf D3, page 9, lines 32 to 35) means that only particles of the first (shell) type are coagulated

with particles of the second (base) type. It is clearly expressed in D3 that "agglomeration" of both of these types of particles among each other is to be avoided (eg page 6, lines 26/27, 40, 46 to 50, and 62 to 64; page 7, lines 4/5 and 59 to 62; page 8, lines 56/57; page 9, lines 16/17). This difference between the subject-matter of Claim 1 and the disclosure of D3 has nothing to do with the question whether, in D3, the shell or the base particles form the "principal" resin component in the sense of Claim 1, which question, in the view of the Board, is quite irrelevant.

On page 8, line 11, of D3, sintering "at between about 5 to 20 centigrades above the glass temperature" is mentioned, which temperature is thus outside the temperature range defined in Claim 1.

No values are given for the percentage of conversion reached.

4.2.3 D2 describes a process for producing a toner for electrophotography by polymerizing one or more polymerizable monomers stably dispersed by emulsification in the presence of a colorant.

However, this process neither relates to emulsion polymerization, nor is there any indication that coagulation occurs. No conversion of at least 99% and no heat treatment temperature of more than 24°C above Tg are mentioned.

4.2.4 D4 describes a process for producing a toner for electrophotography including a magnetic substance. An "age-treatment" at a temperature of 140°C is mentioned (page 8, lines 4/5).

However, the toner is not produced by emulsion polymerization, but by spray drying. No coagulation or conversion rate of at least 99% are mentioned.

4.2.5 The subject-matter of Claim 1 is thus novel in the sense of Article 54 EPC.

4.3 Inventive step

4.3.1 There is no controversy about the fact that D1 represents the prior art closest to the present subject-matter. This document indeed discloses an emulsion polymerization process, coagulation of the particles of the emulsion, and heating of the coagulating particles above the glass transition point.

The object of the present subject-matter is to produce a toner excellent in image density, resolution, gradation, cleaning property, charge stability, caking resistance and durability (see page 4, lines 9 to 14, and page 9, line 63 to page 10, line 2, of the patent specification), ie in other words: good properties in practically all essential respects.

4.3.2 As stated above, the process according to Claim 1 differs from the teaching of D1, inter alia, by the fact that coagulation is conducted after the production of the principal resin component has been practically completed. In the view of the Board, this is not merely an equivalent to the known coagulation during polymerization. The properties of the dispersed particles change substantially when they are

polymerized, and it is therefore credible that a totally different structure of the coagulated particles is achieved, depending on coagulation during or after polymerization. There is no reason to doubt that the overall properties of the particles coagulated after polymerization (and treated in accordance with the other features of Claim 1) are substantially improved.

Since there is no easily detectable path leading from the conditions of polymerization and coagulation to the properties of the particles, a person skilled in the art could not derive from D1 any suggestion regarding coagulation after polymerization.

4.3.3 The other cited documents also do not provide any suggestions in this respect.

Although D3 certainly also tries to optimize the toner particles in all essential respects (see page 1, lines 11 to 26), the path which it follows for reaching this goal is incompatible with the process according to D1 and would not lead, even if combined with the teaching of D1, to the process according to Claim 1.

D3 puts the emphasis on the production of very spherical particles having an exceptionally narrow size distribution (see page 1, lines 54/55; which is contrary to the patent in suit where perfectly spherical particles are considered disadvantageous, cf page 3, line 50, and page 7, lines 60/61, of the patent specification) and, to this end, causes precipitation, in a combined dispersed system, of small dispersed "shell" polymer particles on the surface of large dispersed "base" polymer particles which had been polymerized in a particular manner in an emulsion separate from the emulsion of the "shell" polymer in order to obtain particularly monodisperse and spherical

base particles. Coagulation of the particles of one species among themselves, as it is claimed in present Claim 1 and mentioned in D1, is expressly to be avoided (see the above discussion in paragraph 4.2.2). The teaching of D3 therefore points in the direction opposite to that followed in Claim 1 and D1.

From D4 it might be learned that the spherical shape of particles is not optimal regarding cleaning property. However, the way of obtaining these non-spherical particles is quite different from the method of Claim 1 since there is no emulsion polymerization and no coagulation of the resultant dispersed polymer particles, but a deposition by means of spray drying of a number of small "protuberances" on the surface of large "developer" particles produced by grinding and spray drying.

D2 also does not teach the coagulation of the polymer particles, and moreover, does not relate to emulsion polymerization, but to suspension polymerization.

4.3.4 Since the indicated main idea of the process according to Claim 1, together with the necessary features for rendering it realizable, is not obvious having regard to the cited prior art, the process according to Claim 1 of the second auxiliary request involves an inventive step in the sense of Article 56 EPC.

4.4 Claim 1 according to the second auxiliary request is therefore allowable (Article 52(1) EPC).

Claims 2 to 15 are allowable due to their dependence on Claim 1.

4.5 The description of the patent is not yet adapted to the new Claim 1. Substantial amendments will be required for bringing it in agreement with the invention now claimed (among others, many of the Examples are no longer covered by the conditions of Claim 1 regarding emulsion polymerizing and heat treatment temperature) and for acknowledging the closest prior art documents.

Since this adaptation could not be carried out at the oral proceedings before the Board, the Board makes use of its power under Article 111(1) EPC to remit the case to the Opposition Division for further prosecution.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the Opposition Division with the order to maintain the patent in amended form as follows:

Claim 1 as presented at the oral proceedings as second auxiliary request,

Claims 2 to 15 corresponding to Claims 2 to 15 of the main request filed on 22 September 1995,

and the description to be adapted.

The Registrar: The Chairman:

P. Martorana E. Turrini