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) [ ] To Chairmen and Members
(C) [X] To Chairmen
(D) [ ] No distribution

D E C I S I O N
of 5 July 2005

Case Number: T 0429/02 - 3.4.2

Application Number: 87906449.1

Publication Number: 0323513

IPC: G03G 9/08

Language of the proceedings: EN

Title of invention:
Toner for electrophotography

Patentee:
Mitsui Chemicals, Inc.

Opponent:
Sanyo Chemical Industries, Ltd

Headword:
-

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

Keyword:
"Novelty (yes)"
"Inventive step (yes)"

Decisions cited:
-

Catchword:
-

Europäisches Patentamt | European Patent Office | Office européen des brevets

Beschwerdekammern | Boards of Appeal | Chambres de recours

Case Number: T 0429/02 - 3.4.2

D E C I S I O N
of the Technical Board of Appeal 3.4.2
of 5 July 2005

Appellant: (Opponent)
Sanyo Chemical Industries, Ltd.
11-1, Ikkyo Nomoto-cho Higashiyama-ku
Kyoto 605 (JP)

Representative:
Wilhelms, Rolf E.
WILHELMS, KILIAN & PARTNER
Patentanwälte
Eduard-Schmid-Strasse 2
D-81541 München (DE)

Respondent: (Proprietor of the patent)
Mitsui Chemicals, Inc.
5-2, Higashi-Shimbashi 1-Chome
Minato-ku
Tokyo (JP)

Representative:
Kirkham, Nicholas Andrew, et al
Graham Watt & Co LLP
St Botolph's House
7-9 Botolph's Road
Sevenoaks
Kent TN13 3AJ (GB)

Decision under appeal:
Decision of the Opposition Division of the European Patent Office posted 19 February 2002 rejecting the opposition filed against European patent No. 0323513 pursuant to Article 102(2) EPC.

Composition of the Board:

Chairman: A. G. Klein
Members: F. J. Narganes-Quijano
C. Rennie-Smith

Summary of Facts and Submissions

I. The appellant (opponent) has lodged an appeal against the decision of the opposition division rejecting the opposition against European patent No. 0 323 513 (based on European patent application No. 87 906 449.1).

The opposition filed against the patent as a whole was based on the grounds of lack of novelty and lack of inventive step (Article 100(a) together with Articles 52(1), 54 and 56 EPC).

In the decision under appeal the opposition division referred *inter alia* to the following documents:

D2: JP-A-60150056 together with a partial English translation thereof and the post-published patent document US-A-4727010 (document D5) of the same patent family,

D3: JP-A-61124953 together with a partial English translation thereof,

D4: JP-A-60230666 together with a partial English translation thereof and the patent application document GB-A-2159824 (document D6) of the same patent family, and

D7: JP-A-58-100859 together with a partial English translation thereof,

and to the following experimental test report:

Report A: Certificate of experimental results submitted by the patent proprietor with its letter of 21 September 2000.

The opposition division held in its decision that the polymer resins disclosed in document D7 were not cross-linked and that none of documents D2, D3, D4 and D7 allows the conclusion that the corresponding resins have a Z average molecular weight Mz satisfying the conditions required by the invention. The opposition division concluded that the invention defined in the patent as granted was novel and involved an inventive step over the prior art.

II. During the appeal proceedings the appellant and the respondent (patent proprietor) referred to the following additional experimental test reports submitted during the appeal proceedings:

Report B: Certificate of experimental results submitted by the appellant with the statements of grounds of appeal, and

Report C: Certificate of experimental results submitted by the respondent with its letter of 6 June 2005.

III. In reply to the summons to oral proceedings issued by the Board, the respondent requested as its main request that the appeal be dismissed, or that the patent be maintained as amended according to one of auxiliary requests 1 to 5 filed with its letter dated 6 June 2005.

IV. Oral proceedings were held before the Board on 5 July 2005 in the presence of the parties.

The appellant requested that the decision under appeal be set aside and that the patent be revoked in its entirety. The respondent withdrew its previous main request and auxiliary requests 1, 2, 4 and 5 and

requested maintenance of the patent as amended according to auxiliary request 3 filed with its letter dated 6 June 2005 together with page 3 of the description of the patent as amended during the oral proceedings.

At the end of the oral proceedings the Board gave its decision.

V. Independent claims 1 and 11 according to the respondent's single request read as follows:

"1. An electrophotographic toner which comprises a resin and a coloring agent as primary ingredients, said resin being a non-crosslinked polymer of a vinyl monomer or a mixture containing said non-crosslinked polymer, and said resin having a number average molecular weight (Mn) of 2,000-15,000, a Z average molecular weight (Mz) of not less than 400,000, a ratio of the Z average molecular weight to the number average molecular weight (Mz/Mn) of 50-600, and wherein said resin is a mixture obtainable by mixing a high molecular weight polymer with a low molecular weight polymer in a state of solution."

"11. A method for the preparation of an electrophotographic toner resin having a number average molecular weight (Mn) of 2,000 - 15,000, a Z average molecular weight (Mz) of not less than 400,000, and Mz/Mn of 50 - 600, which comprises mixing in a state of solution (1) 30 - 70 parts by weight of non-crosslinked high molecular weight polymers as solid component obtained by heating a vinyl monomer at 60 - 150°C, conducting a bulk polymerisation to a conversion of 30 - 90 % by

weight, successively adding a solvent to reduce the viscosity of reaction mixture and carrying out a solution polymerisation at 60 - 150°C, with (2) 70 - 30 parts by weight of non-crosslinked low molecular weight polymers as solid component obtained by polymerizing a styrene type vinyl monomer with another vinyl monomer at 190 - 230°C, followed by removing the solvent from the resulting mixture."

Claims 2 to 10 and claims 12 to 14 are dependent claims referring back to claims 1 and 11, respectively.

VI. The arguments of the appellant in support of its request were substantially the following:

According to document D7 the polymer contains an amount of 0.05 to 2 % by weight of a compound having at least two non-conjugated double bonds. In addition, the document specifies that an amount exceeding 2 % by weight results in insoluble polymer. This is a clear indication that the polymer should be soluble. Thus, the polymers of document D7 are soluble in tetrahydrofuran and are therefore non-cross-linked within the meaning of the patent (page 4, lines 4 and 5).

The polymer resins of document D7 have a number average molecular weight Mn between 2000 and 15000. Furthermore, as evidenced by the experimental tests shown in Report B, resins A and B of examples 1 and 2 of document D7 have inherently a Z average molecular weight Mz satisfying the claimed conditions. As to resin C of example 3 having a value Mn of 5600 and a value of the ratio Mw/Mn of 98, it is well known from

the definition of the quantities Mn, Mw and Mz that $Mn \leq Mw \leq Mz$; it follows that the value of Mz/Mn of resin C should be above 98. In addition, it is also known that the values of Mn, Mw and Mz are correlated, and that values of Mz/Mn exceeding 600 are not attainable by copolymerisation of monomers without blending any other polymer. The tests shown in Report A of the patentee, however, are based on blending a small amount of high molecular weight copolymer with a large amount of a low molecular weight polymer. The patent itself acknowledges that the claimed upper limit of 600 for the ratio Mz/Mn is difficult to achieve (page 4, lines 21 to 23). Since the resins of document D7 were prepared using the same divinyl compound of the invention to broaden the molecular weight distribution, the value of Mz of the resins of D7 is also increased by virtue of the same mechanism explained in the patent.

Solution polymerisation and the mixing of low and high molecular weight polymers in a state of solution as used in the patent are well known processes in the prior art as shown in documents D2 (example 3 of document D5), D3 and D4 (examples 8 to 11 of document D6), and no special or surprising effect can be seen in the fact that the polymer resin is produced as specified in claim 1. The advantages associated with the use of a mixture of high and low molecular weight polymer components and with the absence of insoluble material are also well known in the prior art as shown in documents D2, D3 and D4.

VII. The arguments of the respondent in support of its request can be summarised as follows:

Document D7 follows a standard polymerisation process in which the use of 2 % by weight of a cross-linking compound at the start of the reaction results inevitably in cross-linking and therefore in the formation of insoluble matter. All the experimental tests carried out on the basis of example 1 of document D7 and shown in Report C confirm the production of significant quantities of a THF-insoluble gel component. The polymers according to the patent, however, are produced following a different polymerisation process in which the use of small amounts of a cross-linking agent helps in building single chains but does not result in the formation of insoluble, cross-linked material. Moreover, contrary to the patent, document D7 does not properly require the absence of some insoluble component.

As shown in Report A, the values of Mz and Mz/Mn of a polymer depend on the precise molecular weight distribution of the polymer and are not unconditionally determined only by the values of Mn and Mw. Document D7, however, is silent as to the value of Mz and it is doubtful whether the polymers obtained in document D7 satisfy the claimed relationship $Mz/Mn \leq 600$. Some of the tests shown in Report C and based on the disclosure of document D7 result in resins having a ratio Mz/Mn clearly above 600. Also documents D2, D3 and D4 are silent as to the value of Mz.

In addition, contrary to document D7, the production of the polymers according to the invention involve solution polymerisation. As can be inferred from a comparison of the GPC charts shown in Figures 12 and 14 of Report C, the polymers obtained according to the

claimed invention have a peak in the charge distribution that is absent in the polymers obtained by the polymerisation process of document D7. There is no suggestion in the prior art that the production of the resins of document D7 following a process as that disclosed in the invention, while keeping the molecular weight distribution of the resins within predetermined limits, improves the charge characteristics of the resins. In particular, there is no suggestion that the use of small amounts of a cross-linking agent allows for predetermined values of Mz and Mz/Mn that result in a toner with reduced variation of the electrostatic charge during duplication to a level of 10 % or less.

Reasons for the Decision

1. The appeal complies with the requirements mentioned in Rule 65(1) EPC and is therefore admissible.

2. *Amendments*

The amendments to the patent according to the single request of the respondent and denoted during the appeal proceedings as auxiliary request 3 only affect claim 1 and page 3 of the description as granted.

Present claim 1 differs from claim 1 as granted in that the claim further specifies that the resin is a mixture obtainable by mixing a high molecular weight polymer with a low molecular weight polymer in a state of solution. This feature is based on the passage on page 3, lines 25 and 26 of the patent specification and on the corresponding passage on page 7, lines 15 to 18

of the publication under Article 158(3) EPC of the original application filed as an international application under the PCT (Articles 123(2) EPC). In addition, the additional feature does not extend the protection conferred by claim 1 as granted (Article 123(3) EPC).

The statements of invention on page 3 of the patent specification have been amended during the appeal proceedings to make them consistent with the amended claim 1 (Article 84 and Rule 27(1)(c) EPC).

Thus, the Board is satisfied that the amendments made to the patent according to the respondent's request are admissible (Rule 57a EPC) and comply with the formal requirements of the EPC.

3. *Claim 1 - Novelty*

3.1 The assessment of the novelty of the subject-matter of claim 1 has been confined during the appeal proceedings to the prior art represented by the disclosure of document D7. This document discloses an electro-photographic image developing toner comprising a resin of styrene polymer and a colouring agent of carbon black as primary ingredients, the resin having a number average molecular weight $Mn = \sum MiNi/\sum Ni$ in the range 2000 to 15000 (last two paragraphs of page 1 and second paragraph of page 3 of the partial English translation of the document). In particular, the resin C according to example 3 has a value Mn of 5600 and a ratio of the weight average molecular weight $Mw = \sum Mi^2Ni/\sum MiNi$ to Mn of Mw/Mn = 98 (central paragraph of page 2) and therefore a value of Mw of 548800. The document does

not specify the value of the Z average molecular weight defined in the patent as $Mz = \sum Mi^3Ni / \sum Mi^2Ni$ (page 11, lines 1 to 10 of the patent specification). However, it follows from the definition of Mw and Mz that - as stated by the appellant - $Mz \geq Mw$; therefore, the value of Mz of resin C satisfies $Mz \geq 548800$ and $Mz/Mn \geq 98$.

3.2 During the proceedings the appellant submitted experimental evidence (Report B) and substantive arguments in support of its contention that polymer resins according to document D7 have intrinsically a value of the ratio Mz/Mn below 600 as claimed and, in addition, contain no component insoluble in tetrahydrofuran (THF) and are therefore non-cross-linked within the meaning of the patent (page 4, lines 4 and 5, and page 5, line 51 to page 6, line 1). The respondent for its part disputed the appellant's contention in this respect and filed evidence (Reports A and C) in support of its submissions that the polymerisation process of document D7 results in THF insoluble matter and that the information content of document D7 is inconclusive as to the value Mz of the resins.

Nonetheless, the supplementary feature specified in the amended claim 1 further requires that the resin is a mixture obtainable by mixing a high and a low molecular weight polymer in a state of solution. Although this feature of the claimed toner is formulated as a product-by-process feature, the mixture of polymers can be detected in the resulting product itself and, in addition, this feature cannot be derived from the partial English translation of document D7 on file which is silent as to a mixture of high and low

molecular weight polymers as claimed. Moreover, Report C submitted by the respondent shows that the gel permeation chromatography (GPC) chart of a resin as claimed and obtained by mixing a high and a low molecular weight polymer in a state of solution exhibits a peak in the charge distribution (Figure 12 of Report C) that is absent in the corresponding chart of resins obtained following the process of document D7 and which chart presents a broader, more uniform charge distribution (Figure 14 of Report C). Thus, in the absence of evidence or substantive arguments to the contrary on the part of the appellant, the Board accepts the respondent's contention that the product-by-process feature specified in the claim endows the claimed toner with structural features in the form of identifiable and reliable technical characteristics that can be detected in the toner itself and that, in addition, distinguish the claimed toner from the toners disclosed in document D7 (see "Case Law of the Boards of Appeal", 4th edition, EPO, 2001, chapter I, section C-3.2.7).

It follows that the product-by-process feature specified in claim 1 confers novelty on the claimed electro-photographic toner over the toners disclosed in document D7, independently of whether or not the document anticipates intrinsically the claimed features relating to the non-cross-linked state and to the characteristics of the molecular weight distribution of the polymer resin.

3.3 The remaining documents considered during the proceedings are less relevant or at least less conclusive as far as the issue of novelty is concerned.

In particular, documents D2, D3 and D4 all disclose electro-photographic toners comprising a vinyl polymer resin; however, as was held by the opposition division and as was uncontested by the appellant during the appeal proceedings, none of the documents provides sufficient information that would allow the conclusion that the resins intrinsically satisfy the claimed average molecular weight relationships.

3.4 In view of the above, the Board concludes that claim 1 amended according to the respondent's request defines novel subject-matter over the documents considered during the proceedings (Articles 52(1) and 54 EPC).

4. *Claim 1 - Inventive step*

4.1 In view of the disclosure of documents D2, D3, D4 and D7 and the submissions of the parties during the proceedings (points 3.1 and 3.2 above), the Board concurs with the appellants' view that the disclosure of document D7 constitutes the appropriate starting point in the assessment of inventive step of the claimed subject-matter.

According to the submissions of the respondent, the technical effect achieved over the toners of document D7 by the product-by-process feature specified in claim 1 in combination with the remaining claimed features, and in particular with the non cross-linked state of the polymer and the molecular weight distribution of the polymer resin, is that the claimed electro-photographic toner has a reduced variation of the electrostatic charge during duplication to a level of 10 % or less as supported by the patent

specification (page 3, lines 52 to 58, page 4, lines 10 to 21 and 35 to 48, and page 8, lines 12 to 18 together with Tables 2 and 3 of the patent specification) and by the experimental results reported in Report C (point 3.2 above).

Accordingly, the problem solved by the claimed subject-matter over the disclosure of document D7 can be seen in improving the charge characteristics of the toner and in particular in reducing the variation of the electrostatic charge of the toner during duplication.

4.2 Document D2 discloses toner resins obtained from a mixture of low and high molecular weight vinyl polymers in a xylol solvent and mentions a variation of not more than 20 % in the triboelectric charge of the toners after 10000 copies (claim 1 and central paragraphs on page 4 of the English translation). Nonetheless, the document focuses on the number average molecular weight Mn of the individual polymer components (claim 1 and example 3 of the English translation) and is silent as to the molecular weight characteristics of the resulting resins. In addition, the disclosure of the document, and in particular a comparison of the examples and the comparative examples (Table 1) which all involve mixing low and high molecular weight polymers, does not allow the conclusion that the fact of mixing high and low molecular weight polymer components in a state of solution might itself have a beneficial effect on the charge characteristics of the toner resins. Thus, there is no teaching in the document that would hint at improving the charge characteristics of the toners of document D7 or at mixing polymer components having different molecular

weights while keeping the molecular weight distribution of the resulting resin within predetermined limits as claimed.

Document D3 is directed to toner binder resins of a styrene-acrylate copolymer comprising high and low molecular weight polymer components and having a ratio Mw/Mn of the weight to the number average molecular weight of not less than 3.5 (claims 1 and 2 and example 3 of the English translation). There is, however, no teaching in the document that any of these features, and in particular the use of a mixture of high and low molecular weight polymers, may improve the charge characteristics of the toner.

Document D4 mentions, among others, the problem of the triboelectric and charge controlling characteristics of toner resins (page 1, lines 30 to 47 of document D6) and proposes the use of a toner binder polymeric resin having three peaks in the GPC chromatogram satisfying predetermined requirements (claim 1 and Figure 1 of document D4). The resins are either obtained by controlling the polymerization conditions so as to adjust the molecular weight distribution of the resulting polymer (document D6, page 5, line 59 *ff*.), or by mixing polymers having different molecular weights in a state of solution (document D6, page 6, line 2 *ff*.). However, the first alternative involves cross-linking (page 3, lines 22 to 26, and page 6, lines 14 to 20 and lines 51 to 61 together with examples 1 to 7 of document D6) that results in THF insoluble components, i.e. teaches away from the non-cross-linked polymer resins of the claimed electro-photographic toner. Furthermore, the second alternative

specifies value ranges of the average molecular weights and of the relative amounts of the polymer components (page 5, line 12 *ff*. and examples 8 to 11 of document D6) required to obtain the GPC chromatogram having the appropriate characteristics (page 3, lines 44 to 55), but none of these characteristics would ensure that the molecular weight distribution of the resulting resin satisfies the claimed conditions. In addition, none of these alternatives are disclosed as specifically improving the charge characteristics of the toners.

Thus, none of the documents suggests improving the charge characteristics of the electro-photographic toners of document D7, and in particular reducing the variation of the electrostatic charge during duplication, by using a toner resin having the characteristics of a resin obtainable from a mixture of a high and a low molecular weight polymer in a state of solution while avoiding cross-linking and at the same time keeping the molecular weight distribution of the resulting polymer resin within the value ranges required by the claimed subject-matter.

4.3 In view of the above, the Board concludes that the subject-matter of claim 1 involves an inventive step over the prior art considered by the parties during the proceedings (Articles 52(1) and 56 EPC).

5. *Claim 11*

Although the opposition filed by the appellant also encompassed - at least formally - claim 11 as granted, during the proceedings the appellant did not provide

facts or substantive arguments in support of the objections of lack of novelty and lack of inventive step of the preparation method defined in claim 11 as granted. In addition, no prior art has been identified during the proceedings that would anticipate or at least render obvious the production of a polymer resin by bulk polymerisation followed by solution polymerisation under the polymerisation conditions defined in claim 11.

Consequently, claim 11 defines patentable subject-matter within the meaning of Articles 52(1), 54 and 56 EPC.

6. *Dependent claims*

The same conclusions drawn in points 3, 4 and 5 above with regard to independent claims 1 and 11 apply to claims 2 to 10 and 12 to 14 by virtue of the dependence of these claims on independent claims 1 and 11, respectively.

7. The Board is therefore satisfied that the patent as amended according to the request of the respondent and the invention to which it relates meet the requirements of the EPC. Accordingly, the contested decision is to be set aside and the patent maintained in amended form on the basis of the patent documents according to the respondent's request (Article 102(3) EPC).

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 maintain the patent as amended on the basis of claims 1 to 14 of auxiliary request 3 filed on 6 June 2005 and the description pages 2 and 4 to 18 as granted and page 3 as filed during the oral proceedings.

The Registrar: The Chairman:

P. Martorana A. G. Klein