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Bezeichnung der Erfindung:
Title of invention:
Titre de l'invention : Method of producing barium-ferrite series powder

Klassifikation / Classification / Classement : H01F 1/11

ENTSCHEIDUNG / DECISION

vom / of / du 18 July 1990

Anmelder / Applicant / Demandeur :

Patentinhaber / Proprietor of the patent /
Titulaire du brevet : Kabushiki Kaisha Toshiba

Einsprechender / Opponent / Opposant : BASF AG

Stichwort / Headword / Référence :

EPÜ / EPC / CBE Article 56

Schlagwort / Keyword / Mot clé : "Inventive step - denied"

Leitsatz / Headnote / Sommaire

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Case Number : T 262/88 - 3.5.2

D E C I S I O N
of the Technical Board of Appeal 3.5.2
of 18 July 1990

Appellant : (Opponent)
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Decision under appeal :
Decision of Opposition Division of the European Patent Office dated 19 April 1988 rejecting the opposition filed against European patent No. 0 039 773 pursuant to Article 102(2) EPC.

Composition of the Board :

Chairman : W.J.L. Wheeler
Members : C. Black
M. Lewarton

Summary of Facts and Submissions

I. The grant of European patent No. 0 039 773 on the Respondent's European patent application No. 81 102 435.5, which was filed on 31 March 1981 claiming priority from a previous application in Japan dated 8 May 1980, was published on 4 September 1985.

II. Claim 1 of the granted patent reads as follows:

"A method of producing barium-ferrite series powders from an alkaline aqueous solution containing metal ions in amounts sufficient to provide the metals contained in the final product of the barium-ferrite series powder, said barium-ferrite series powder having a general formula of $BaO.n(Fe_{1-m}M_m)_2O_3$, where "M" is at least one trivalent metallic substituent or an equal amount of metallic substituents of valences of 2 and 4 selected from the group consisting of Co, Ti, Ni, Mn, Cu, Zn, In, Ge and Nb, "m" is 0 to 0.2, and "n" is 5.4 to 6.0, the alkaline aqueous solution having an alkaline normality of at least 0,01 N, and being prepared

(1) by adding a first aqueous solution to a second aqueous solution of strong alkali, said first solution being prepared in advance by dissolving water-soluble compounds of barium, iron (III) and substituent component M in water,

(2) by heating the alkaline aqueous solution at 150 to 300°C at constant volume to precipitate a precursor of the barium-ferrite series powder; and

(3) by baking the precursor at 700 to 1,000°C to crystallize the precursor."

III. On 30 May 1986 the Appellant filed an admissible opposition requesting revocation of the patent on the ground that its subject-matter did not involve an

inventive step having regard in particular to the disclosure in

T. Takada und M. Kiyama, Ferrites: Proceedings of the International Conference, July 1970, Japan, pages 69-71 (D1)

M. Kiyama, Bulletin of the Chemical Society of Japan, Vol. 49, pages 1855-1860 (1976) (D2)

G. Heimke, Berichte der Deutschen Keramischen Gesellschaft, Vol. 39 (1962) pages 326-330 (D3).

IV. The opposition was rejected by a decision of the Opposition Division dated 19 April 1988. The reasons for the decision were, in summary, that although all of the features of Claim 1 to the end of step (2) were to be found in D2, none of the cited documents contained a teaching which would lead the average skilled person to bake the thus formed material as required by step (3) of Claim 1. In fact a reference in D2 to calcining would lead the skilled person away from contemplating step (3) because in D2 no change in saturation, magnetisation and coercive force occurred.

V. On 16 June 1988 the Appellant filed a notice of appeal against that decision and paid the appeal fee. The statement of grounds was filed on 17 August 1988. The Respondent filed a first auxiliary request on 22 June 1990. Oral proceedings were held on 18 July 1990, at which the Respondent filed a second auxiliary request.

VI. Claim 1 of the first auxiliary request reads as follows:

"A method of producing substituted barium-ferrite series powders from an alkaline aqueous solution containing metal

ions in amounts sufficient to provide the metals contained in the final product of the barium-ferrite series powder, said barium-ferrite series powder having a general formula of $BaO.n(Fe_{1-m}M_m)_2O_3$, where "M" is at least one trivalent metallic substituent or an equal amount of metallic substituents of valences of 2 and 4 selected from the group consisting of Co, Ti, Ni, Mn, Cu, Zn, In, Ge and Nb, $0<m \leqq 0.2$, and "n" is 5.4 to 6.0, the alkaline aqueous solution having an alkaline normality of at least 0,01 N, and being prepared

(1) by adding a first aqueous solution to a second aqueous solution of strong alkali, said first solution being prepared in advance by dissolving water-soluble compounds of barium, iron (III) and substituent component M in water,

(2) by heating the alkaline aqueous solution at 150 to 300°C at constant volume to precipitate a precursor of the barium-ferrite series powder; and

(3) by baking the precursor at 700 to 1,000°C to crystallize the precursor."

Claim 1 of the second auxiliary request reads as follows:

"A method of producing substituted barium-ferrite series powders from an alkaline aqueous solution containing metal ions in amounts sufficient to provide the metals contained in the final product of the barium-ferrite series powder, said barium-ferrite series powder having a general formula of $BaO.n(Fe_{1-m}M_m)_2O_3$, where "M" is at least one trivalent metallic substituent or an equal amount of metallic substituents of valences of 2 and 4 selected from the group consisting of Co, Ti, Ni, Mn, Cu, Zn, In, Ge and Nb, $0<m \leqq 0.2$, and "n" is 5.4 to 6.0, the alkaline aqueous solution having an alkaline normality of at least 0,01 N, and being prepared

(1) by adding a first aqueous solution to a second

aqueous solution of strong alkali, said first solution being prepared in advance by dissolving water-soluble compounds of barium, iron (III) and substituent component M in water,

(2) by heating the alkaline aqueous solution at 150 to 300°C at constant volume for a heating time of about 20 minutes to 2 hours to precipitate a precursor of the barium-ferrite series powder; and

(3) by baking the precursor at 700 to 1,000°C to crystallize the precursor."

VII. The argumentation of the Appellant can be summarised as follows: D2 describes a method of producing a barium-ferrite having the formula set out in Claim 1 with n=6, m=0 and having all the features required by Claim 1 except for step (3), as is apparent from Fig. 1 and the corresponding description. The product of this method must therefore correspond to the "precursor" according to the wording of Claim 1, which apparently has the desired crystal contour, but crystallisation is not complete and the precursor is low in coercive force and saturation magnetism (see column 5, lines 22 to 26 of the opposed patent). From D3 it is known that the coercivity and crystallinity of barium ferrite can be improved by annealing at 700 to 1 000°C (page 328, Fig. 2 and description). For the skilled person it is obvious to combine this teaching with that of D2 and thus to arrive at the subject-matter of Claim 1 (main request).

Moreover the disclosure in D2 as regards calcining does not lead away from the subject-matter of the patent in suit. The reason why calcining does not lead to an increase in saturation magnetisation and coercive force in D2 is that the samples, prepared by a different hydrothermal method, already had high values.

To the first auxiliary request the Appellant argues that it is common knowledge to alter the coercivity of barium-ferrite by substitution, and substantiates this by referring to G. Winkler, Z. angew. Phys., 21 Bd., Heft 4, 1966, pages 282-288 (D5) in particular page 284, right column, last two paragraphs, and page 285, Fig. 6, which indicates that the coercivity may be reduced by replacing Fe by Co and Ti. In the patent in suit the aim of the dopants is also to reduce coercivity.

To the second auxiliary request the Appellant argues that there is nothing in the patent in suit to suggest that the heating time in step (2) is important for the invention, nor does D2 suggest that the 5-hour heating time in the corresponding step is essential.

VIII. The Respondent argues essentially as follows: The subject-matter of Claim 1 (main request) differs from the D2 disclosure not only in the requirement for step (3) but also in that step (2) results in a precursor of the final barium ferrite powder. There is no suggestion of such a precursor in D2. The fact that calcining the samples referred to on page 1859, right column, leads to no improvement in magnetic properties indicates that these samples were already fully crystallised because they had been autoclaved for a relatively long time (5 hours). Since such samples are unchanged by further treatment these are not precursors. There is no teaching in D2 that calcining could bring about an improvement in the magnetic properties of a poorly crystallised precursor having substantially no magnetic properties.

D3 relates to barium ferrite powders that have been prepared by a sintering process, not a hydrothermal process and examines the reduction in coercive field strength resulting from grinding. It is shown that the

coercive field strength of the ground material can be increased by heat treatment at about 1 000°C. However the samples treated already had a fairly high coercive field strength, and therefore are different from the precursor in the patent in suit which has substantially no magnetic properties. There is therefore no incentive to combine the teachings of D2 and D3.

As to the first auxiliary request, D5 indeed discloses that incorporation of Co and Ti into a barium ferrite lowers the coercive force. However, the purpose is to make barium ferrite cores which are suitable for high frequency applications, whereas in the patent in suit the purpose is to render the ferrite suitable for the head of a magnetic recording apparatus. Moreover D5 does not disclose a method of preparation of barium ferrite corresponding to that of the patent in suit.

The heating time of about 20 minutes to 2 hours which is a feature of Claim 1 according to the second auxiliary request is much less than the 5-hour heating time referred to in D2, page 1855, right column. Since for the subsequent baking step one hour is sufficient, a product with satisfactory magnetic properties is obtained in a shorter time.

IX. The Appellant requests that the decision under appeal be set aside and the patent revoked. The Respondent requests that the appeal be dismissed and

(1) that the patent be maintained as granted (main request), or

(2) that the patent be maintained in amended form on the basis of the auxiliary request filed on 22 June 1990, or

(3) that the patent be maintained in amended form on the basis of the auxiliary request submitted at the oral proceedings.

Reasons for the Decision

1. The appeal is admissible.

2. **Novelty**

D2 discloses a method of producing barium ferrite series powders from an alkaline aqueous solution containing metal ions in amounts sufficient to provide the metals contained in the final product of the barium ferrite series powder, the barium ferrite series powder having the general formula $BaO.6Fe_2O_3$ (see page 1855 under "Experimental"). The alkaline aqueous solution has an alkaline normality of at least 0.01 N (see pages 1855, 1856, Results and Discussion, pH=11 for suspension with R=0.9 and Fig. 1; according to the patent in suit, column 4, line 32, 0.01 N corresponds to pH12). It is prepared by adding to a first aqueous solution a second aqueous solution of strong alkali, the first aqueous solution being prepared in advance by dissolving water-soluble compounds of barium and iron (III) in water (page 1855, process (1) under "Procedure"). The alkaline aqueous solution is heated at 150 to 300°C (see Fig. 1) at constant volume (page 1855, right column "to prevent the reduction in volume of the suspension") to form a precipitate of $BaO.Fe_2O_3$ (page 1855, 1856 Results and Discussion).

The subject-matter of Claim 1 (main request) differs from this disclosure in that the precipitate, called a precursor of the barium ferrite series powder, is baked at 700 to 1 000°C to crystallise the precursor. A further

difference is that according to Claim 1 the first aqueous solution is added to the second aqueous solution of strong alkali rather than vice versa. None of the other citations comes closer to the subject-matter of Claim 1, which is accordingly novel. Novelty is moreover not disputed by the Appellant.

3. **Inventive step**

3.1 The argumentation of the Respondent relies in part on his contention that D2 does not disclose a precursor which is the product of step (2) of Claim 1. In the Board's view the word precursor does not define the substance in question, but merely indicates that it is to be submitted to a subsequent treatment which will bring about some sort of modification. If therefore it can be shown that it is obvious to submit the precipitate which is the product of process (1) of D2 to a baking step then the said product will also be describable as a "precursor", in which case no distinction would be seen in this respect.

3.2 A related argument of the Respondent is in effect that the barium ferrite powder resulting from the process disclosed in D2 cannot be equated with the precursor according to Claim 1 because the said powders already have a high saturation magnetisation and coercive field strength which cannot be increased by baking, as evidenced by page 1859, penultimate paragraph. He suggests this is because of the relatively long autoclave treatment of 5 hours, whereas according to the patent in suit the corresponding step (2) of Claim 1 can be carried out in 20 minutes to 2 hours. This cannot be followed. The passage in question refers to a limited group of samples prepared under various conditions, though it is not clear what conditions. From the context it would seem that process (2) (page 1855, right column) is being employed, which differs from

process (1) and the process according to Claim 1 in starting from a suspension of insoluble iron oxygen compounds. It is true that page 1855 refers to a reaction time of 5 hours for both process (1) and process (2), but in a further investigation into process (2), (page 1857), reaction times of 5 minutes to 8 hours are quoted, with 5 minutes being in some cases sufficient for complete conversion to $BaO.6Fe_2O_3$. There is no reason to suppose that in the above-mentioned passage on page 1859, "under various conditions" is restricted to a reaction time of 5 hours rather than the shorter times referred to on page 1857 or for that matter any other times. It is therefore incorrect to conclude the good magnetic properties of some samples resulted from the relatively long reaction time. The passage on page 1857 which discusses variation in reaction time refers only to its effect on the composition of the product and not to magnetic properties. As pointed out by the Appellant and not disputed by the Respondent, the most likely effect of prolonging reaction time would be an undesired increase in crystal growth. For the person of average skill in the art there is nothing in D2 to suggest that the reaction time of 5 hours referred to on page 1855 is essential; he derives rather from a general reading of the document that shorter times can be sufficient. Moreover a reaction time of 20 minutes to 2 hours was not a feature of any of the claims as originally filed or of any of the claims of the main request, but only of Claim 1 of the second auxiliary request.

3.3 It is appropriate at this point to deal with the question of the significance of whether the first aqueous solution is added to the strong alkali or the strong alkali to the first aqueous solution (see paragraph 2 above), even though this was not put forward by the Respondent. It is conceivable that the physical structure of the precipitate

might be different if this was formed immediately on contact, because it is forming in a different environment in the two cases. However this is something of which the laboratory chemist is aware and it is within his competence to choose which of the two options gives him the better product. The Board is therefore satisfied that adding the first aqueous solution to the alkali as in Claim 1 does not contribute to inventivity.

3.4 The conclusion to be drawn from the foregoing paragraphs 3.1 to 3.3 is that the person of average skill in the art, following the teaching of D2 with particular reference to process (1) described therein, will be carrying out a process which does not differ materially from the method of Claim 1 up to the end of step (2). He will therefore have on his hands a product substantially equivalent to the precursor which according to the patent in suit, column 5, lines 24 to 29, since crystallisation is not completed, is very low in coercive force and saturated magnetisation.

3.5 According to D3, page 328, second complete paragraph, defects in crystal structure can be healed to a great extent by treatment at elevated temperatures. In the Board's opinion, this statement is to be seen in its context as being of general applicability and not just to the case of barium ferrites whose coercive strength has been reduced by grinding. This is borne out by the sequence of statements on page 328, left column. First of all the reduction in coercive strength of the ground material is referred to, then the assumption that this is due to defects caused by grinding, then the above-mentioned statement concerning a possible remedy, then the conclusion that the coercive strength ought to be increased by heat-treatment, confirmed experimentally - see Fig. 2. Moreover the figure indicates that the

appropriate temperature range for the heat treatment is 700 to 1 000°C just as in step (3) of Claim 1. It is noted too that curve (c) shows an increase in coercive strength of a barium ferrite material which does not appear to have been submitted to grinding.

For the person of average skill in the art it is therefore obvious to apply the teaching of D3 to an incompletely crystallised material of insufficient coercive force obtained by process (1) of D2, and thus arrive at the subject-matter of Claim 1 (main request).

In contrast to the view of the Opposition Division and the Respondent, the passage in D2, page 1859, right column, to the effect that there was no remarkable difference in magnetic properties between calcined and original samples, is seen as corroborating this. As pointed out by the Appellant, the samples in question already had high values. In the Board's opinion, the person of average skill in the art would interpret this passage as indicating that the samples had been calcined in the expectation of improving their magnetic properties.

The subject-matter of Claim 1 (main request) therefore does not involve an inventive step.

3.6 Claim 1 of the first auxiliary request differs from Claim 1 of the main request only in that m=0 is excluded, that is a substituent metal must be present. In the patent in suit this serves to decrease the coercive force to a level suitable for the head of magnetic recording apparatus. However, it is known from D5, page 284, right column and Fig. 6 to reduce the coercive strength of barium ferrites by just such a measure. Although the stated purpose is different, the generality of the

teaching is unaffected, and this feature cannot introduce inventivity.

3.7 Claim 1 according to the second auxiliary request contains the additional feature that the reaction time in step (2) is 20 minutes to 2 hours. This has been considered in paragraph 3.2 from which it follows that the Board sees no contribution to inventivity in this feature. In deciding on the second auxiliary request the Board is using its powers under Article 111(1) EPC because it considers that a remittal to the Opposition Division is unjustified, even though the feature in question has apparently not previously been considered.

3.8 Since ground (a) in Article 100 EPC prejudices maintenance of the patent with any of the forms of Claim 1 specified in the Respondent's requests, the patent must be revoked. The Board need not consider the dependent claims.

Order

For these reasons, it is decided that:

1. The decision under appeal is set aside.

2. European patent No. 39 773 is revoked.

The Registrar: The Chairman:

M. Kiehl W.J.L. Wheeler

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