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D E C I S I O N
of 4 May 1999

Case Number: T 0208/97 - 3.5.2

Application Number: 87107963.8

Publication Number: 0248402

IPC: G11B 5/706

Language of the proceedings: EN

Title of invention:
Magnetic recording medium and production thereof

Patentee:
Hitachi Maxell Ltd.

Opponent:
BASF Akiengesellschaft

Headword:
-

Relevant legal provisions:
EPC Art. 56

Keyword:
"Inventive step (no) "

Decisions cited:
-

Catchword:
-



Case Number: T 0208/97 - 3.5.2

D E C I S I O N
of the Technical Board of Appeal 3.5.2
of 4 May 1999

Appellant: Hitachi Maxell Ltd.
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Decision under appeal: Decision of the Opposition Division of the
European Patent Office posted 11 December 1996
revoking European patent No. 0 248 402 pursuant
to Article 102(1) EPC.

Composition of the Board:

Chairman: W. J. L. Wheeler
Members: M. R. J. Villemin
A. C. G. Lindqvist

Summary of Facts and Submissions

- I. The appeal contests the decision of the opposition division to revoke European patent No. 248 402. The reasons given for the revocation were that the subject-matter of claim 1 then on file was not novel and the subject-matter of the other claims then on file did not involve an inventive step.
- II. In response to the summons to attend oral proceedings before the Board, the appellant filed a main request and an auxiliary request with the letter dated 1 April 1999.
- III. Claim 1 of the main request and the auxiliary request are identical and worded as follows:
- "A magnetic recording medium comprising a non-magnetic substrate and a magnetic layer formed thereon which contains a chromium oxide ferromagnetic powder comprising chromium oxide of the formula CrO_x , wherein x is not smaller than 1.74 and not larger than 1.91 obtainable by a gradual conversion of CrO_2 to a non-magnetic material comprising Cr_2O_3 (Cr(III)-oxide) by heating at a temperature not lower than 410°C , wherein the chromium oxide ferromagnetic powder has saturation magnetization of 40 to 75 emu/g."
- IV. Many prior art documents were cited in the opposition and appeal proceedings, but the decisive arguments were based on the following documents:
- D1: Chamberland: "The Chemical and Physical Properties of CrO_2 and Tetravalent Chromium Oxide Derivatives" in CRC CRITICAL REVIEWS in SOLID STATE and MATERIAL SCIENCES, Vol. 7, No. 1, November 1997, pages 1 to 31.

D2: Bhatnagar et al: "The Magnetic Susceptibilities of Metallic Oxides, with Special Reference to those of Chromium and Manganese" in Journal of the Chemical Society, pages 1433 to 1441 (1939).

D6: US-A-3 529 930.

V. Oral proceedings were held on 4 May 1999.

VI. The appellant argued that no document cited by the respondent disclosed a magnetic recording medium intended for solving the problem specified in the description of the patent specification (page 3, lines 16 to 18), namely to provide a magnetic recording medium having good overwriting properties and improved stability so that its magnetic properties were less deteriorated after long time storage. The inventors had discovered that these properties might be obtained when CrO_2 was heated at a temperature not lower than 410°C , to produce chromium oxide of the formula CrO_x , wherein x is not smaller than 1.74 and not larger than 1.91, until the saturation magnetization of the so obtained oxide ferromagnetic powder had a value of 40 to 75 emu/g. A reasonable decrease of the saturation magnetization was acceptable because it was compensated by an improvement of the overwriting properties and a reduction of the deterioration of magnetic properties after long term storage.

It was agreed that D1 generally described the existence of Cr_5O_9 , and referred to the study of the magnetic properties of this material reported on in document D2. However, as could be seen from the graph on page 1438 of D2 showing the changes in the value of x in CrO_x as the temperature was raised, the chromium oxide composition changed sharply from $\text{CrO}_{2.4}$ to $\text{CrO}_{1.2}$ at around 360°C (from points C to D on the graph)

indicating that the method of D2 was not suitable for producing a stable chromium dioxide. As described in the last seven lines on page 1437 of D2, chromium oxide was paramagnetic between points B and C on the graph but suddenly became faintly ferromagnetic at point D. This meant that CrO_x was not ferromagnetic when x was larger than 1.8 and only faintly ferromagnetic when x was 1.8. D2 described that the ferromagnetism became stronger as the second (Cr_5O_9) non-stoichiometric range was traversed and that at point F the normal paramagnetic susceptibility of chromic oxide was registered, but this document did not disclose the maximum level of ferromagnetism. Since the chromium oxide was paramagnetic at point F, it was to be assumed that the ferromagnetism between points D and F was not high enough for magnetic recording media, the provision of which however was a chief aim of the present invention. Consequently, a skilled person learned from D1 only that chromium oxides of various compositions, including Cr_5O_9 , were available and that CrO_2 was used in magnetic recording media. There was no hint in D1 or D2 that the use of chromium oxide CrO_x with x in the range 1.74 to 1.91 might have improved the magnetic stability and the overwriting properties of a recording medium, so that it was not obvious to try to manufacture magnetic recording media using sub-stoichiometric chromium oxides according to D2 while maintaining the good magnetic properties of CrO_2 .

The method described in document D6 increased the saturation magnetization, unlike the present invention. Consequently, it would not have been easy to know which level of saturation magnetization or which kind of chromium oxide could provide magnetic recording media having good long term storage stability while maintaining the overwriting properties of CrO_2 . The Board had indicated it thought a skilled person was able to control the decrease of magnetization due to

the increase in proportion of Cr_2O_3 . However, none of the prior art documents disclosed or suggested the selection of a particular range of chromium oxide composition which was stable and could suppress the deterioration of saturation magnetization over a long time. Thus, there was absolutely no incentive to go that way. In response to critical comments on table 3 of the patent specification made by the respondent concerning the values of the residual magnetic flux density after storage for four weeks, it should be noted that the invention aimed at improving the stability of this flux density over a long time and not at increasing its absolute value.

- VII. The respondent argued that, using the figures in table 3 of the contested patent, calculations of the values of the residual magnetic flux densities after storage of samples 3, 4, 5 and 10 prepared according to the invention showed that an improvement in the residual magnetic flux densities after storage in comparison to that of some of the untreated samples could not be convincingly established.

The relatively horizontal region between points D and E of the graph on page 1438 of D2 showed that relatively stable ferromagnetic oxides existed in the range $\text{CrO}_{1.9-1.7}$, when the temperature varied from 400 to 500°C. This marked structural stability led to a reduction of the deterioration of magnetic properties after long term storage.

It was known from D1 (last line of table 2 on page 18) that the Curie temperature could be raised, and consequently magnetic stability improved, by substituting Cr(III) ions for some of the Cr(IV) ions of CrO_2 in the system $\text{CrO}_2\text{-Cr}_2\text{O}_3$, and, for the person skilled in the art, the corresponding concomitant decrease of the magnetization saturation ($\Delta\mu_s < 0$) shown

in this table indicated that a deterioration of the overwriting properties was not to be expected. Therefore it was obvious to attempt to replace at least part of the chromium dioxide CrO_2 by a ferromagnetic sub-stoichiometric chromium oxide having a composition around $\text{CrO}_{1.8}$ for producing a magnetic recording medium.

VIII. The Appellant requested that the decision under appeal be set aside and that the patent be maintained in amended form according to:

- the main request: claims 1 to 6 and pages 3 and 4 of the description as filed with the letter dated 1 April 1999; pages 2 and 5 to 7 and Figures 1 and 2 of the patent specification; or
- the auxiliary request: claims 1 to 3 and pages 3 and 4 of the description as filed with the letter dated 1 April 1999; pages 2 and 5 to 7 and Figures 1 and 2 of the patent specification.

IX. The respondent requested that the appeal be dismissed.

Reasons for the Decision

1. The appeal is admissible.
2. In the present case it is appropriate to go straight to the question of whether the subject-matter of claim 1 of the appellant's main and auxiliary requests involves an inventive step, since if this question is decided against the appellant there is no need to consider any other point.

3. According to the description of the patent specification as granted the main problem to be solved is the providing of a magnetic recording medium having good overwriting properties and improved stability so that its magnetic properties are less deteriorated after long term storage (see page 3, lines 16 to 18).

4. This problem is solved by a magnetic recording medium according to claim 1 which specifies that the magnetic layer of the recording medium contains a chromium oxide ferromagnetic powder comprising chromium oxide of the formula CrO_x , where $1.74 \leq x \leq 1.91$, obtainable by a gradual conversion of CrO_2 to a non-magnetic material comprising Cr_2O_3 , but specifies no details of this gradual conversion other than that it is by heating at a temperature not lower than 410°C and, by implication, that it produces chromium oxide ferromagnetic powder having saturation magnetization of 40 to 75 emu/g.

5. D1 discloses that the primary use of CrO_2 is in magnetic tape applications (page 9, right hand column, end of first paragraph) and that crystals of CrO_2 (rutile structure) are decomposed into Cr_2O_3 (corundum structure) by a topotactic reaction without shattering the original crystals on heating in air at temperatures "around 415°C " or "at 435°C "; see page 9, right-hand column, third and fourth paragraphs and page 28, left-hand column). These temperatures are above the lower limit of 410°C specified in claim 1. D1 cites on page 2, right-hand column, third paragraph, earlier works on the thermal decomposition of CrO_3 leading, inter alia, to the phase: Cr_5O_9 ($\text{CrO}_{1.7}$ to 1.9), and mentions that, according to the document reference 16, which is in fact document D2, the Cr_5O_9 phase was characterized as the magnetic intermediate.

6. Document D2 mentions that dry methods reported to yield CrO_2 gave compounds which appear to be non-stoichiometric, belonging to the oxide range $\text{CrO}_{1.9-1.7}$ (see page 1436, lines 30 and 31) and **that oxides of the Cr_5O_9 , non-stoichiometric range are normally ferromagnetic** (page 1436, lines 43 to 45; emphasis added by the Board). The graph on page 1438 of D2 shows that such sub-stoichiometric oxides CrO_x with x ranging between about 1.7 and 1.8 are obtained between 400 and 500°C. D2 further discloses (see page 1437, last three lines to page 1438, line 4) that ferromagnetism becomes stronger as the second non-stoichiometric range (that is, the Cr_5O_9 , range) is traversed and that the chief interest of these results "lies in the absence of any significant change in magnetic properties during non-stoichiometric decomposition, even although the oxide composition varied considerably." The person skilled in the art would immediately understand from this that, since Cr_5O_9 ($x=1.8$) is ferromagnetic and is located exactly in the middle of the range $x = 1.7$ to 1.9, other non-stoichiometric chromium oxides in this range can be expected to have relatively stable ferromagnetic properties.
7. D1 reports (page 13, left-hand column, first paragraph) that "pure fine-grained CrO_2 had room temperature saturation magnetization values (at infinite field) of 77 to 85 emu/g" whereas "large-grained CrO_2 possessed values of 98 to 108 emu/g". Claim 1 does not indicate the purity or the grain size of the parent CrO_2 to be used, so it is not possible to assess objectively whether the values 40 to 75 emu/g of the saturation magnetization specified in the claim could be indicative of a particular or surprising effect produced by this process or not. Rather, since the magnetic recording medium containing CrO_x according to claim 1 also contains non-ferromagnetic material comprising Cr_2O_3 obtained by gradual thermal conversion

of parent ferromagnetic CrO_2 , it seems to be self-evident that the final magnetic recording medium would be less ferromagnetic than the parent CrO_2 and would consequently exhibit a saturation magnetisation value lower than that of the parent CrO_2 .

8. It seems therefore obvious to the skilled person that any predetermined value of saturation magnetisation σ_s lower than that of the untreated CrO_2 powder can be obtained by properly adjusting the proportion of non-magnetic material comprising Cr_2O_3 formed during the heating process of gradually converting CrO_2 into non-magnetic material. The skilled person would also be guided by the necessity of arriving at values of σ_s which are not too weak, so that the produced ferromagnetic powder is still suitable for preparing a ferromagnetic medium for magnetic recording. According to D6, claim 1, values of σ_s above 70 emu/g are obtained after heating CrO_2 at about 150°C to 450°C under a pressure of 0.2 to 3000 atmospheres in an oxidizing environment. Such a value of σ_s falls within the range of 45 to 75 emu/g specified in claim 1 of the patent opposed.
9. The process by which the ferromagnetic powder specified in claim 1 may be obtained is not distinguishable from the prior art thermal treatments above 410°C indicated in D1 or D2 and therefore must be assumed to produce the same effects and products. Claim 1 specifies no details of the process which could cause it to produce different results.
10. The Board concludes that the subject-matter of claim 1 of the main and auxiliary requests does not involve an inventive step having regard to documents D1 and D2 as they would be understood by the person skilled in the art. Therefore the patent cannot be maintained with this claim.

11. Since neither of the appellant's requests can be allowed, it follows that the appeal must be dismissed.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar:



M. Kiehl

The Chairman:



W. J. L. Wheeler

