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(54) Hard magnetic material of a rare earth metal, iron and carbon.

(57) A hard magnetic material having the substantially stoichiometric composition $Nd_2Fe_{14}C$. It can be obtained by casting and subsequently recrystallization annealing at a temperature between 840 and 890° C. A part of Fe may be substituted by Co. In this case the magnetic material is obtained by casting and subsequently recrystallization annealing at a temperature of approximately 850° C.

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"Hard magnetic material of a rare earth metal, iron and carbon."

The invention relates to a hard magnetic material comprising neodymium, iron and carbon.

A known material of this type is $Nd_2Fe_{14}B$ having the tetragonal crystal structure. It is known that in this compound substitution of B by C leads to an increased anisotropy (see, for example, Journal de Physique Colloque C6, supplément au no. 9, T 46, Sept. 1985, page C6-305/308: "Magnetic Anisotropy of Carbon Doped $Nd_2Fe_{14}B$" by Bolzoni, Leccabue, Pareti and Sanchez). In this article it is stated on page 306 that it was impossible to obtain the tetragonal phase when borium was completely substituted by carbon in the compound $Nd_2Fe_{14}B$. This is also stated in an article in Solid State Communications Vol. 64, No. 5 p. 639-644 (1987). In this article "Synthesis and Magnetic Properties of Ternary Carbides $R_2Fe_{14}C$" by Gueramiom, Bezinge, Yvon and Muller it is stated on page 640 that the tetragonal structure in, amongst others, R = neodymium is not found by means of recrystallization annealing.

It is an object of the invention to provide a hard magnetic material having a high crystal anisotropy, which comprises only carbon instead of boron and also a relatively high content of neodymium.

It was found that this object could be achieved by means of a material which has the tetragonal crystal structure and which consists of a recrystallization annealed cast material having a composition which does not or hardly deviate from the stoichiometric composition $Nd_2Fe_{14}C$. In this composition Fe may be substituted by Co up to a maximum of 6 at.% calculated on the overall composition. If more than 6 at.% of Fe is substituted by Co the quantity of hard magnetic material having the tetragonal crystal structure, which can be obtained in the annealing treatment decreases significantly.

The hard magnetic material in accordance with the invention can be obtained as follows. The starting materials neodymium, iron, (possibly) cobalt and carbon are melted together in a substantially stoichiometric ratio, preferably, under an inert gas atmosphere such as argon. The melt is cast in a mould. The material has the Nd_2Fe_{17} structure and is not hard magnetic; the carbon is dissolved in the lattice. It is assumed that the carbon in the lattice could substitute one or more iron atoms. The structure is rhombohedral. If desired, the casting obtained can be homogenization annealed at a temperature of 900° or higher. If the material only comprises neodymium, iron and carbon it is annealed at a temperature between 840 and 890° C so that recrystallization takes place. It has been found that recrystallization takes place only within this temperature range, thereby forming the tetragonal $Nd_2Fe_{14}C$ phase. Surprisingly it has been found that on weighing out the starting materials it is undesirable to clearly deviate from the stoichiometric composition as is required in the case of the corresponding boron compounds. A small positive deviation of no more than 20% in the quantity of neodymium and/or carbon relative to the stoichiometric composition and of 15% in the quantity of iron, however, has been found permissible. The tetragonal phase is not formed outside the indicated temperature range (above 890° C) or, if said phase is formed, predominantly, a second phase having the Nd_2Fe_{17} structure remains in addition to the tetragonal phase (below 840° C). If annealing is carried out in the indicated temperature range the tetragonal phase is predominantly formed. A substantially single-phase material having optimum magnetic properties can be obtained if annealing is carried out at temperatures between 850 and 880° C, preferably at 870° C. If the material also comprises Co, the annealing process in which recrystallization takes place is preferably carried out at a temperature of approximately 850° C. It has been found that cobalt considerably increases the Curie temperature of the hard magnetic material, which may be desirable for certain applications. In the manufacture of the Co-containing hard magnetic materials in accordance with the invention, it has been found that on weighing out the individual elements a small deviation from the stoichiometric composition is not necessary, but it is permissible. A positive deviation of 20% in the quantity of neodymium and/or carbon as well as a positive deviation of 15% in the quantity of iron and/or cobalt is permissible. The experiments leading to the invention were also carried out using praseodymium instead of neodymium; it was found impossible to obtain a substantially single-phase material having the tetragonal crystal structure.

Example 1:

As has been described above, castings were manufactured having the overall composition $Nd_2Fe_{14}C$. After casting the material had a Nd_2Fe_{17} structure (rhombohedral). Without previous homogenization annealing the castings obtained were directly recrystallization annealed for 500 hours at 870° C. The material was single phase and had a tetragonal crystal structure. The lattice constants were measured. It was found that: A = 8.814 Å, C = 12.015 Å. The material in

accordance with the invention has a Curie temperature of 535 K. The saturation magnetization at 20° C = 130 EMU per gram. Single-phase materials having the tetragonal crystal structure were obtained in the same manner using materials having the overall composition $Nd_{12.1}Fe_{81.8}C_6$ and $Nd_{11.8}Fe_{81.1}C_{7.1}$.

Magnets can be obtained by directly casting the material in the desired shape. After casting and recrystallization annealing the material can be powdered and subsequently sintered to the desired shape in a magnetic field or be mixed with synthetic resin and pressed into the desired shape.
In the said materials, part of the iron may be substituted by other 3d metals and/or aluminium, gallium, silicon etc. Part of the neodymium can be substituted by one or more rare earth metals.

Example 2:

A casting was manufactured, the overall composition of which corresponded to the formula $Nd_2Fe_{13}CoC$. Subsequently, this casting was directly subjected to a recrystallization annealing process at 850° C for 150 hours without being previously homogeneization annealed. The material thus obtained is substantially single phase and has a tetragonal crystal structure. The material has a Curie temperature of 620 K.

Claims

1. A hard magnetic material comprising a rare earth metal, iron and carbon, characterized in that the material consists of a recrystallization annealed cast material having a tetragonal crystal structure and a composition which does not or hardly deviate from the stoichiometric composition $Nd_2Fe_{14}C$.

2. A hard magnetic material as claimed in Claim 1, characterized in that a maximum of 6 at.% of the Fe present in the hard magnetic material, calculated on the overall composition, has been substituted by Co.

3. A hard magnetic material as claimed in Claim 1, characterized in that as regards neodymium and carbon the composition's positive deviation from the stoichiometric composition does not exceed 20%, and as regards iron the positive deviation does not exceed 15%.

4. A hard magnetic material as claimed in Claim 3, characterized in that as regards Nd and C the composition's positive deviation from the stoichiometric composition does not exceed 20%, and as regards iron and cobalt the positive deviation does not exceed 15%.

5. A hard magnetic material as claimed in Claim 3, characterized in that the lattice constants amount to A = 8.814 Å and C = 12.015 Å.

6. A method of manufacturing a hard magnetic material comprising neodymium, iron and carbon, characterized in that the starting materials neodymium, ion and carbon are melted together in a quantity which deviates little from the stoichiometric ratio $Nd_2Fe_{14}C$, and cast in a mould, after which the casting is recrystallization annealed at a temperature between 840 and 890° C.

7. A method as claimed in Claim 6, characterized in that the material is annealed at a temperature between 850 and 880° C.

8. A method of manufacturing a hard magnetic material comprising neodymium, iron, cobalt and carbon, characterized in that the starting materials neodymium, iron, cobalt and carbon are melted together in a quantity which deviates little from the stoichiometric ratio $Nd_2Fe_{14-x}Co_xC$ (wherein $x < 1.02$) and cast in a mould, after which the casting is recrystallization annealed at a temperature of approximately 850° C.

9. Magnetic castings comprising a hard magnetic material as claimed in anyone of the Claims 1 upto and including 5, and/or magnetic castings obtained by means of a method as claimed in anyone of the Claims 6 up to and including 8.

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EUROPEAN SEARCH REPORT

Application Number

EP 88 20 2784

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.4)
A	JOURNAL OF APPLIED PHYSICS, PROCEEDINGS OF THE THIRTY-FIRST ANNUAL CONFERENCE ON MAGNETISM AND MAGNETIC MATERIALS, vol. 61, no. 8, part 11A, 15th Aptil 1987, pages 3574-3577, American Institute of Physics, New York, US; N.C. LIU et al.: "High intrinsic coercivities in iron-rare earth-carbon-boron alloys through the carbide or boro-carbide Fe14R2X(X=BxC1-x) * Page 3574, right-hand column, paragraph 1; page 3575, left-hand column, last paragraph - page 3576, right-hand column, last paragraph * ---	1	H 01 F 1/04
D,A	SOLID STATE COMMUNICATIONS, vol. 64, no. 5, 1987, pages 639-644, Pergamon Journals Ltd, London, GB; M. GUERAMIAN et al.: "Synthesis and magnetic properties of ternary carbides R2Fe14C(R = Pr,Sm,Gd,Tb,Dy,Ho,Er,Tm,Lu) with Nd2Fe14B structure type" * Page 640, left-hand column, last paragraph - right-hand column, table 1 * ---	1	TECHNICAL FIELDS SEARCHED (Int. Cl.4) H 01 F
A	MATERIALS LETTERS, vol. 4, nos. 8-9, August 1986, pages 377-380, Elsevier Science Publishers, N.V., Amsterdam, NL; N.C. LIU et al.: "High coercivity permanent magnet materials based on iron-rare-earth-carbon alloys" * Page 377, right-hand column * --- -/-	1	

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
THE HAGUE	07-03-1989	DECANNIERE L.J.

CATEGORY OF CITED DOCUMENTS

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DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.4)
A	PATENT ABSTRACTS OF JAPAN, vol. 4, no. 109 (E-20)[591], 6th August 1980, page 92 E 20; & JP-A-55 67 110 (SUWA SEIKOSHA K.K.) 21-05-1980 * Abstract *		

TECHNICAL FIELDS SEARCHED (Int. Cl.4)

The present search report has been drawn up for all claims

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THE HAGUE	07-03-1989	DECANNIERE L.J.

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