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EUROPEAN PATENT APPLICATION

21 Application number: **89200693.3**

51 Int. Cl.⁴: **H01F 1/04**

22 Date of filing: **20.03.89**

30 Priority: **24.03.88 NL 8800739**

43 Date of publication of application:
27.09.89 Bulletin 89/39

84 Designated Contracting States:
AT CH DE FR GB IT LI NL

71 Applicant: **N.V. Philips' Gloeilampenfabrieken**
Groenewoudseweg 1
NL-5621 BA Eindhoven(NL)

72 Inventor: **De Mooij, Dirk Bastiaan**
c/o INT. OCTROOIBUREAU B.V.
Prof.Holstlaan 6
NL-5656 AA Eindhoven(NL)
Inventor: **Buschow, Kurt Heinz Jürgen**
c/o INT. OCTROOIBUREAU B.V.
Prof.Holstlaan 6
NL-5656 AA Eindhoven(NL)

74 Representative: **Weening, Cornelis et al**
INTERNATIONAAL OCTROOIBUREAU B.V.
Prof. Holstlaan 6
NL-5656 AA Eindhoven(NL)

54 **Hard magnetic material.**

57 A hard magnetic material having the composition $RE_2Fe_{14-x}Mn_xC$, wherein RE is at least one element selected from the group formed by Nd, Pr, Ce and La, and wherein $0.2 \leq x \leq 2$.

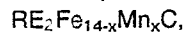
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Hard magnetic material

The invention relates to a hard magnetic material comprising a rare earth metal, iron and carbon. The invention also relates to a magnet on the basis of hard magnetic material.

A hard magnetic material of the type described in the opening paragraph is known from, *inter alia*, J. Appl. Phys. 61 3574-3576 (1987). The authors of the article classify the compound $\text{Nd}_2\text{Fe}_{14}\text{C}$ described therein as not stable. Consequently, this material cannot be used as a starting material for a permanent magnet.

It is an object of the invention to provide a hard magnetic material which comprises a rare earth metal (RE), iron and carbon, and which is stable. This object is achieved in accordance with the invention by a hard magnetic material which is further characterized in that the material contains a hard magnetic phase the composition of which corresponds to the formula



wherein RE is at least one element selected from the group formed by Nd (Neodymium), Pr (Praseodymium), Ce (Cerium) and La (Lanthanum), and wherein $0.2 \leq x \leq 2$. It has been found that the value of x cannot be selected randomly. The materials in which x is smaller than 0.2 were found to be insufficiently stable. The Curie temperature of the materials in which x is larger than 2 is about room temperature or below room temperature, which is problematic for the magnetic application of these materials. In principle, the Curie temperature can be raised by substituting a part of the iron present in the material with cobalt. The absence of B in the compounds according to the invention is an advantage because as a consequence thereof no toxic, volatile boron compounds can be formed during the preparations of said compounds.

In experiments it has further been established that the hard magnetic phase of the compounds according to the invention has a tetragonal crystal structure of the ThMn_{12} type, which is also the case with the known $\text{Nd}_2\text{Fe}_{14}\text{B}$. As is shown in the Table below, the Curie temperature (T_c) of the compounds according to the invention is between 300 and 550 K. The Curie temperature of compounds according to the invention having an identical rare earth metal is lower as the Mn content is higher.

Table

Compound	Curie temperature (K)
$\text{Nd}_2\text{Fe}_{13.7}\text{Mn}_{0.3}\text{C}$	505
$\text{Nd}_2\text{Fe}_{13.5}\text{Mn}_{0.5}\text{C}$	492
$\text{Nd}_2\text{Fe}_{13}\text{Mn}\text{C}$	452
$\text{Nd}_2\text{Fe}_{12.5}\text{Mn}_{1.5}\text{C}$	400
$\text{Nd}_2\text{Fe}_{12}\text{Mn}_2\text{C}$	355
$\text{Pr}_2\text{Fe}_{13.7}\text{Mn}_{0.3}\text{C}$	481
$\text{Pr}_2\text{Fe}_{13.5}\text{Mn}_{0.5}\text{C}$	460
$\text{Pr}_2\text{Fe}_{13}\text{Mn}\text{C}$	412
$\text{Pr}_2\text{Fe}_{12}\text{Mn}_2\text{C}$	306

The hard magnetic materials according to the invention can be obtained in a customary way by melting together suitable starting materials in ratios corresponding to the compositional formula, after which they are subjected to an annealing treatment in a temperature range from 800°C - 950°C in a protective gas or a vacuum for two days; during which treatment recrystallization occurs.

It is noted that in the experiments leading to the present invention it has become clear to Applicants that the manufacture of a stable material having a hard magnetic phase, the composition of which corresponds to the formula $\text{Nd}_2\text{Fe}_{14}\text{C}$, can be achieved in principle. This was attained by subjecting a casting of the compositional formula $\text{Nd}_2\text{Fe}_{14}\text{C}$ to a prolonged annealing treatment within a narrow temperature range, preferably between 850 and 880°C , during which treatment recrystallization occurs. Applicants have reason to believe that in comparison with the known method the preparation of the material according to the invention requires a shorter annealing treatment for recrystallization in order to obtain the desired tetragonal structure. Moreover, in the case of the materials according to the invention, the annealing

treatment can take place in a wider temperature range. On the other hand, this stable material can also be prepared by melt spinning a composition with formula $\text{Nd}_2\text{Fe}_{14}\text{C}$, followed by an annealing treatment of the ribbon formed, during which treatment recrystallization occurs. Both known methods have the disadvantage that they are less suitable from an economical point of view.

5 The hard magnetic properties of the compounds in accordance with the invention are comparable with those of materials on the basis of $\text{Nd}_2\text{Fe}_{14}\text{B}$. For example, the magnetization measured at $\text{Nd}_2\text{Fe}_{13.7}\text{Mn}_{0.3}\text{C}$ powder at room temperature is approximately $100 \text{ Am}^2/\text{kg}$. This value is comparable with the saturation magnetization which is measured at $\text{Nd}_2\text{Fe}_{14}\text{B}$ powder at room temperature. It is noted that this value cannot be regarded as the saturation magnetization, since the material under consideration is magnetically
10 very anisotropic.

After the materials obtained have been pulverized, magnets in the form of shaped bodies can be manufactured from said materials in the usual manner.

15 Claims

1. A hard magnetic material comprising a rare earth metal, iron and carbon, characterized in that the material contains a hard magnetic phase the composition of which corresponds to the formula
 $\text{RE}_2\text{Fe}_{14-x}\text{Mn}_x\text{C}$

20 wherein RE is at least one element selected from the group formed by Nd (Neodymium), Pr (Praseodymium), Ce (Cerium) and La (Lanthanum), and wherein $0.2 \leq x \leq 2$.

2. A hard magnetic material as claimed in Claim 1, characterized in that a part of the iron present in the material is replaced by cobalt.

3. A permanent magnet on the basis of a hard magnetic material as claimed in Claim 1.

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EP 89 20 0693

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)
D,A	JOURNAL OF APPLIED PHYSICS, vol. 61, no. 8, 15th April 1987, pages 3574-3576, American Institute of Physics; N.C. LIU et al.: "High intrinsic coercivities in iron-rare earth-carbon-boron alloys through the carbide or boro-carbide Fe ₁₄ R ₂ X(X=BxC _{1-x})" ---		H 01 F 1/04
A	MATERIALS LETTERS, vol. 4, nos. 8,9, August 1986, pages 377-380, Elsevier Science Publishers B.V., Amsterdam, NL; N.C. LIU et al.: "High coercivity permanent magnet materials based on iron-rare-earth-carbon alloys" ---		
A	JOURNAL OF APPLIED PHYSICS, vol. 52, no. 3, part II, March 1981, pages 2049-2051, American Institute of Physics, New York, US; K. HARDMAN et al.: "Magnetic structures of Y ₆ (Fe _{1-x} Mnx) ₂₃ compounds" -----		
			TECHNICAL FIELDS SEARCHED (Int. Cl.4)
			H 01 F
The present search report has been drawn up for all claims			
Place of search THE HAGUE		Date of completion of the search 15-06-1989	Examiner DECANNIERE L.J.
CATEGORY OF CITED DOCUMENTS X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document			