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R. Coehoorn, D. de Mooij, J. Duchateau, K. Buschow. NOVEL PERMANENT MAGNETIC MATERIALS MADE BY RAPID QUENCHING. Journal de Physique Colloques, 1988, 49 (C8), pp.C8-669-C8-670. 10.1051/jphyscol:19888304 . jpa-00228477

HAL Id: jpa-00228477
https://hal.science/jpa-00228477

Submitted on 4 Feb 2008

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JOURNAL DE PHYSIQUE
Colloque C8, Supplément au n° 12, Tome 49, décembre 1988

NOVEL PERMANENT MAGNETIC MATERIALS MADE BY RAPID QUENCHING

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Abstract. – Two types of permanent magnetic materials were obtained by annealing melt spun flakes: (i) materials consisting mainly of Fe_3B with $Nd_2Fe_{14}B$ as a secondary phase, and (ii) materials based on $Nd_2Fe_{14}C$. The methods of preparation and the resulting magnetic properties are presented.

1. Introduction

Melt spinning was employed widely in the last decade to prepare amorphous alloys. However, it is also possible, by a variation of the wheel speed, to obtain partially crystallized alloys, characterized by a high concentration of nuclei and small crystallites. A modest heat treatment of the latter alloys then leads to a microstructure consisting of much smaller grains than would have been obtained by normal casting procedures. In addition to the microstructural aspect melt spinning offers the possibility to prepare metastable alloys.

We have used the two above mentioned aspects to prepare two novel types of materials suitable for permanent magnet applications:

1) magnets containing mainly the metastable compound Fe_3B, with a small amount of $Nd_2Fe_{14}B$, prepared by crystallizing Nd-Fe-B melt spun ribbons. The Nd content of these materials is 3-5 atomic percent;

2) permanent magnet materials containing the intermetallic compound $Nd_2Fe_{14}C$ as the main phase. This phase, which is difficult to prepare by annealing castings, can be obtained easily by a short anneal treatment of melt spun flakes.

2. Fe_3B-based magnets

A promising method in the search for novel materials for permanent magnets is the preparation of metastable intermetallic compounds by crystallizing amorphous melt spun flakes at relatively low temperatures. Using this method, Buschow *et al.* [1] found in the Nd-Fe-B system two new metastable compounds: cubic $Nd_2Fe_{23}B_3$ ($T_c = 655$ K) and hexagonal $NdFe_{12}B_6$($T_c = 230$ K). Due to their low anisotropy and Curie temperature, respectively, these compounds were unfortunately not suitable for permanent magnet applications. However, in the same study they also found a compositional area in which Fe_3B (the tetragonal modification), $Nd_2Fe_{14}B$ and some α-Fe were the crystallization products.

In this paper we report the magnetic properties of this type of materials. Figure 1 shows the low-field part of the hysteresis curve of flakes with the composition $Nd_4Fe_{80}B_{20}$, isothermally annealed at 670 °C during 30 minutes. The material is magnetically isotropic, with at room temperature a saturation magnetization $\mu_0 M_{sat} = 1.6$ T, remanence $\mu_0 M_r = 1.2$ T and a coercive field $\mu_0 H_c = 0.3$ T. An analysis of the material from the intensities of the peaks in a ^{57}Fe – Mössbauer spectrum shows that, expressed in the number of Fe-atoms, it consists of 73 % Fe_3B, 15 % Nd_2Fe_4B and 12 % α-Fe. The saturation magnetization is determined mainly by the saturation of Fe_3B and $Nd_2Fe_{14}B$, which are both 1.6 T at room temperature. Fe_3B has a high Curie temperature ($T_c = 785$ K), and a relatively weak anisotropy, which is uniaxial [2]. The coercivity is clearly due to the presence of some hard-magnetic $Nd_2Fe_{14}B$. At first, in Nd-free binary Fe_3B materials or in materials in which Gd, Y or Lu was used instead of Nd, the coercive field is very low. Secondly, the coercive field in $Nd_4Fe_{80}B_{20}$ decreases linearly with temperature to zero at the Curie temperature of $Nd_2Fe_{14}B$.

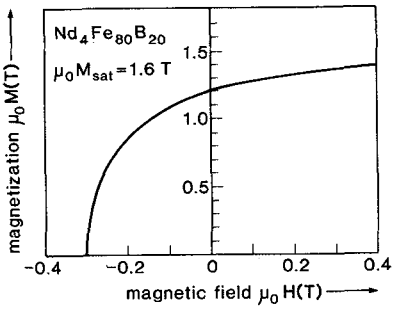

Fig. 1. – Low-field part of the hysteresis curve of crystallized $Nd_4Fe_{80}B_{20}$.

The most remarkable property is the ratio of the remanence over the saturation magnetization, which may become as high as 0.75. This is much higher than 0.5, which would be expected for an assembly of non-interacting paricles with uniaxial anisotropy. The origin of the high M_r / M_{sat} ratio is a subject of current investigations. Transmission electron microscopy shows that the material consists typically of 300 Å Fe_3B grains, with 100 Å $Nd_2Fe_{14}B$ grains in between them. Owing to the large remanence, the $(BH)_{max}$

Article published online by EDP Sciences and available at http://dx.doi.org/10.1051/jphyscol:19888304

energy product is quite high: 93 kJ/m^3 (11.7 MGOe), making the material a suitable candidate for application in inexpensive bonded magnets.

3. $Nd_2Fe_{14}C$-based magnets

In the search for new intermetallic compounds for permanent magnets attempts to prepare $Nd_2Fe_{14}C$ have been unsuccessful for a long time. By standard casting and annealing techniques $R_2Fe_{14}C$ compounds could be prepared for R = Gd, Dy and Er (heavy rare earths) [3, 4], and also the preparation of mixed compounds with some Dy substituted for Nd or some B substituted for C was reported [5]. Recently it was shown that for the light rare earth elements the $R_2Fe_{14}C$ phase is only stable at relatively low temperatures [6]. At a transition temperature T_t it decomposes, forming mainly the high temperature $R_2Fe_{17}C_x$ phase, in which some carbon is dissolved interstitially. For the heavy rare earth elements T_t is approximately 1 100 °C, but for Nd it is only 890 °C. Annealing of castings during several weeks in the narrow temperature interval below 890 °C, but above approximately 820 °C, indeed leeds to the $Nd_2Fe_{14}C$ phase. The lower temperature limit is determined by the kinetics of the transformation, which is too sluggish below 820 °C. The long annealing time severely hampers the manufacturing of sintered $Nd_2Fe_{14}C$ magnets.

We have used the melt spinning technique and a subsequent anneal treatment to overcome this problem. Due to the high degree of disorder in the melt spun flakes, the transformation kinetics is much quicker than in normally cast alloys. Figure 2a shows the X-ray diagram of an as-spun sample with the composition $Nd_{13.5}Fe_{79.6}C_{6.9}$. The peaks can be indexed according to the high temperature $Nd_2Fe_{17}C_x$-phase. Their width indicates that the grains are very small. The broad background feature is due to the presence of an amorphous phase. Upon heating the flakes in the differential scanning calorimeter with a rate of 20 K/min, two peaks due to exothermic transformations were observed. First, at 595 °C, the amorphous phase crystallizes (see Fig. 2b). At 695 °C, the $Nd_2Fe_{17}C_x$ phase transforms into $Nd_2Fe_{14}C$ (Fig. 2c).

The hysteresis curve of a sample of similar composition, annealed for 3 minutes at 720 °C, was measured in a 15 T magnet of the High Field Laboratory of the University of Nijmegen. In figure 3 we show the low field part. The material is magnetically isotropic with a saturation magnetization of 1.40 T. The remanence is 0.72 T and the coercive field $\mu_0 H_c$ is 1.0 T. The small dip in the curve around $\mu_0 H \simeq -0.2$ T is probably due to the presence of some α-Fe as a second phase, which is soft magnetic. The coercive field decreases linearly with temperature to zero at 265 °C, the Curie temperature of $Nd_2Fe_{14}C$. Results of investigations of the compositional dependence of the magnetic properties and the phase diagram will be published elsewhere [7].

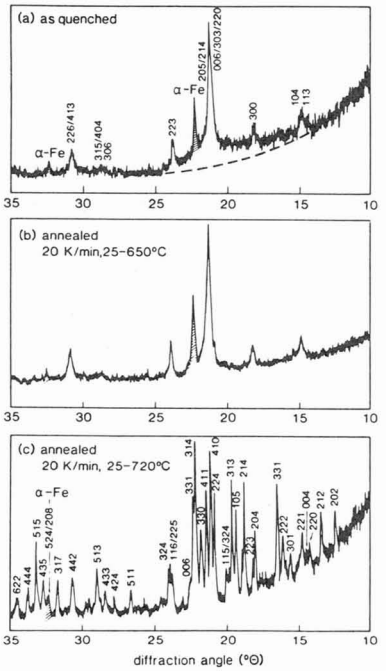

Fig. 2. – Cu-K_α X-ray diagrams of as-quenched $Nd_{13.5}Fe_{79.6}C_{6.9}$ flakes (a), and flakes heated in the differential scanning calorimeter up to 650 °C (b), and up to 720 °C (c).

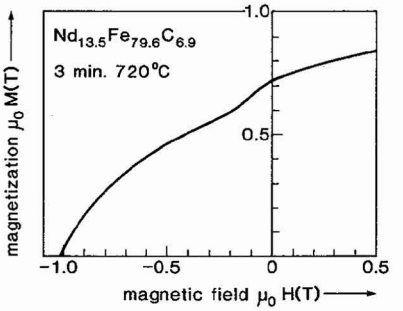

Fig. 3. – Low field part of the hysteresis curve of annealed $Nd_{13.5}Fe_{79.6}C_{6.9}$ flakes.

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