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## LETTER TO THE EDITOR

## Hard magnetic behaviours of composite materials of the iron nanoparticles and HDDR NdDy-FeCo-B powders

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Abstract. Hard magnetic behaviours of the composite materials  $(NdDy-FeCo-B)_{1-x}Fe_x$ prepared by direct mixing of iron nanoparticles with HDDR NdDy-FeCo-B powders have been investigated. With increasing the content of iron nanoparticles in composite materials, the remanence first increases, undergoes a peak and then decreases, whereas the coercivity decreases monotonically. The squareness of the demagnetization curves decreases with increasing content of iron nanoparticles. Meanwhile, a high degree of reversibility in the minor loops along the demagnetization branch in the fields below coercivity has also been observed.

Recently, two-phase nanocrystalline materials consisting of a hard magnetic rare-earthbased intermetallic and an Fe-based soft magnetic phase have attracted much attention due to potential application as permanent magnets with large energy products [1-7]. In such composite permanent materials, the hard magnetic grains give a large coercivity, while the soft magnetic grains, which are sufficiently small and strongly exchange-coupled with hard magnetic grains, induce a high magnetization. Coehoorn et al [1] discovered the effect of remanence enhancement in melt-spun NdFeB, consisting of 85% soft magnetic phases (73% Fe<sub>3</sub>B and 12%  $\alpha$ -Fe) and only 15% hard magnetic phase Nd<sub>2</sub>Fe<sub>14</sub>B. Ding et al [2] reported maximum energy products of more than 20 MG Oe in mechanically alloyed isotropic Sm<sub>7</sub>Fe<sub>93</sub> nitride powders where a significant remanence enhancement was found due to the exchange interaction between a hard magnetic phase  $Sm_2Fe_{17}N_x$  and a soft magnetic phase  $\alpha$ -Fe in powders. Manaf et al [3] also reported a similar effect in  $Nd_2Fe_{14}B + \alpha$ -Fe melt-spun ribbons. The effect of remanence enhancement found in both melt-spun and mechanically alloyed materials is based on the principle that the hard and soft magnetic phases involved emerge from a common metastable matrix phase, such as the glass-like state, from which the very fine grains may be obtained after subsequent heat treatment, and strong magnetic exchange-coupling between soft and hard phases occurs. In the present work, we report on the hard magnetic behaviours of the composite magnets prepared by direct mixing of the iron nanoparticles (about 20-50 nm) and HDDR NdDy-FeCo-B powders followed by ball milling for five minutes, and finally bonded with epoxy resin with the same shape of cylinder. The remanence and coercivity are determined from demagnetization curves without the correction of the demagnetization factors, which are measured by a pulsed high magnetic field. The minor loops along the demagnetization branch for the composite magnet are measured by a vibrating sample magnetometer (VSM). The remanence enhancement has also been found in the materials investigated. Our result implies that one may obtain permanent magnets with high remanences by the direct mixing of nanostructured soft and hard magnetic phases.

Figure 1 shows the weight composition x dependence of the remanence at room temperature for isotropic composite magnets  $(NdDy-FeCo-B)_{1-x}Fe_x$  prepared by direct mixing iron nanoparticles and HDDR NdDy-FeCo-B powders and ball milling for five minutes and then bonding with the epoxy resin. The remanence increases from 61.88 emu  $g^{-1}$  at x = 0 to 68.9 emu  $g^{-1}$  at x = 0.30. With a further increase of the Fe component in composite materials, there is a decrease in remanence. The remanence of the composite material at x = 0.70 of (NdDy-FeCo-B)<sub>1-x</sub>Fe<sub>x</sub> is only 37.37 emu g<sup>-1</sup>. Such remanence enhancement in the composite magnets may come from the exchange interactions of interparticles between soft and hard magnetic phases. Our experimental results indicate that the effect of the remanence enhancement can occur not only in materials through the technique where the soft and hard phases emerge from a common glass-like state by heat treatment, but also in materials obtained through the technique where the soft and hard magnetic phases involved are produced previously and independently. However, the extent of the remanence enhancement occurring in two such kinds of material may be different. This difference may mainly come from the distances between soft and hard magnetic particles. From the point of view of crystallographic coherence and strong magnetic exchange-coupling, it seems that the extent of the remanence enhancement is larger when soft and hard phases emerge from a common metastable matrix phase, as pointed out by Kneller and Hawig [4]. Here, we also wish to point out that by using the high-pressure technique, the extent of the remanence enhancement in the composite magnet prepared by direct mixing of double-nanostructured soft-hard magnetic phases may also be improved.



Figure 1. The weight composition x dependence of the remanence for isotropic composite magnets (NdDy-FeCo-B)<sub>1-x</sub>Fe<sub>x</sub>.

Figure 2. The weight composition x dependence of the coercivity for isotropic composite magnets (NdDy-FeCo-B)<sub>1-x</sub>Fe<sub>x</sub>.

The weight composition x dependence of the coercivity at room temperature for the composite magnets  $(NdDy-FeCo-B)_{1-x}Fe_x$  is shown in figure 2. It is evident that with an increase in the content of iron nanoparticles, the coercivity decreases monotonically. The coercivity is 1.13 T for a magnet with x = 0.0 and 0.92 T for a magnet with x = 0.30. Exchange interactions between the hard and soft magnetic phases preserve a relatively high coercivity for x < 0.3. With a further increasing content of iron nanoparticles in composite magnets, the exchange hardening becomes weak and the coercivity decreases sharply. The coercivity is 0.38 T for bonded composite magnet  $(NdDy-FeCo-B)_{1-x}Fe_x$ , x = 0.70.

The demagnetization curves for the different bonded composite magnets (NdDy-FeCo- $B_{1-x}Fe_x$  which are measured in a pulsed high magnetic field after application of the

magnetizing field of 5 T, are shown in figure 3. It is clear that, with increasing content of iron nanoparticles in composite magnets, the squareness of the demagnetization curve decreases sharply, which is most evident for the composite x = 0.7 in figure 3. Meanwhile, the minor loops along the demagnetization branch for the composite magnet (NdDy-FeCo-B)<sub>1-x</sub>Fe<sub>x</sub> with x = 0.30 have also been measured by a VSM at room temperature. Results are shown in figure 4. A high degree of reversibility in the minor loops along the demagnetization branch in the fields below the coercivity has also been observed.



Figure 3. The demagnetization curves for the different composite magnets  $(NdDy-FeCo-B)_{1-x}Fe_x$ .



Figure 4. Minor loops along the demagnetization branch for composite magnet  $(NdDy-FeCo-B)_{1-x}Fe_x$  with x = 0.30.

In conclusion, the hard magnetic behaviours in the composite magnets (NdDy-FeCo-B)<sub>1-x</sub>Fe<sub>x</sub> prepared by direct mixing of iron nanoparticles with HDDR NdDy-FeCo-B powders have been investigated. With increasing content of iron nanoparticles in the composite magnets, the remanence first increases, undergoes a peak and then decreases, whereas the coercivity decreases monotonically. The squareness of the demagnetization curves decreases with increasing content of iron nanoparticles in composite magnets. Meanwhile, a high degree of the reversibility in the minor loops along the demagnetization branch in the fields below the coercivity has also been observed.

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