Bulk SmCo$_5$/x-Fe nanocomposite permanent magnets fabricated by mould-free Joule-heating compaction

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Bulk SmCo$_5$/x-Fe nanocomposite magnets have been prepared using a Joule-heating compaction technique. Nearly fully dense bulk magnets are obtained by compacting the milled powders under a pressure of 2 GPa at temperatures above 400 °C. Structural analysis shows that the grain size of both the SmCo$_5$ and the x-Fe phases is in the range of 10 to 15 nm when the compaction temperature is lower than 500 °C, which ensures effective interphase exchange coupling. A further increase in compaction temperature leads to significant grain growth and deterioration of magnetic properties. A maximum energy product of about 18.5 MGOe was obtained in the bulk SmCo$_5$/x-Fe nanocomposite magnets, which is 90% higher than that of the single-phase counterpart prepared under the same conditions. © 2011 American Institute of Physics. [doi:10.1063/1.3563098]

I. INTRODUCTION

Nanocomposites consisting of magnetically hard and magnetically soft phases are being extensively investigated due to their potentially superhigh maximum energy products \([BH]_{\text{max}}\). The high energy products originate from the interphase exchange coupling between the neighboring hard and soft magnetic phases.\(^{1-3}\) The prerequisite for effective exchange coupling is a small grain size of the soft magnetic phase, with the critical dimension estimated to be on the order of 10 nm.\(^4\) The experimental nanocomposite materials are typically in the form of low dimensional powder particles or thin films.\(^5-9\) For application purposes, the fabrication of bulk nanocomposite magnets is essential. However, there are major obstacles to producing bulk nanocomposite magnets. Conventional sintering and hot-pressing methods, which are used to produce single-phase microcrystalline permanent magnets, are ineffective in producing bulk nanocomposite magnets, because those techniques require extensive heat treatments, which lead to excessive grain growth. Recent attempts to fabricate bulk nanostructured magnets by using unconventional and expensive compaction techniques, including spark plasma sintering (SPS),\(^{10,11}\) dynamic compaction,\(^12\) and warm compaction,\(^13\) have achieved grain size control in the nanocomposite systems. In this paper, we report our recent work in an alternative methodology for fabricating bulk nanocomposite magnets by using a Joule-heating compaction technique.

II. EXPERIMENT

The raw materials of the nanocomposites were prepared by high energy ball milling the hard magnetic phase SmCo$_5$ particles and the soft magnetic phase x-Fe particles. The weight ratio of powder to ball is around 1:30, and the milling time is 4 h. The as-milled powders contain an amorphous SmCo matrix and a bcc structured soft magnetic phase. The as-milled powders were sealed in a container with a pressure of 2 GPa using a modified QUICKpress apparatus. For comparison, the single-phase SmCo was also prepared by the same process. The schematic view of the Joule-heating compaction set-up is shown in Fig. 1. By changing the applied current through the samples, the heating of the sample is adjustable from room temperature to 650 °C. The density of the compacts was measured by the Archimedes method and by measuring the mass and the volume of regular shaped bulks. The Vickers hardness was measured using a LECO Microindentation Hardness Tester LM247AT. The morphology and crystalline structure were characterized by scanning electron microscopy, transmission electron microscopy (TEM), energy-filtered TEM (EFTEM), and x-ray diffraction (XRD) using Cu K$_\alpha$ radiation. Magnetic properties were measured in a superconducting quantum interference device magnetometer with a maximum applied field of 70 kOe.

III. RESULTS AND DISCUSSION

The Joule-heating compaction (JHC) in this work was performed on an in-house built apparatus. The fixed ac current is directly passed through the loading rams as well as the powder compacts, as shown in Fig. 1(a). The generated Joule heat, i.e., resistance heating from the sample, internally heats the sample. This method is different from conventional hot pressing, in which the heat is provided by external heating elements, such as graphite furnaces [as shown in Fig. 1(b)]. Our facility is similar to spark plasma sintering, also known as pulsed electric current sintering.\(^{10,11}\) However, there are several differences between JHC and SPS: (1) SPS adopts a pulsed dc current,
whereas the JHC in our work uses a stable ac current; and (2) SPS normally uses graphite punches to push the sample, whereas JHC uses a tool steel piston. The tool steel provides higher pressure (up to 2 GPa in this work) than graphite. In addition, the steel has lower resistance than graphite, and thus most of the heat is generated from the sample itself in the JHC technique. This facility has the ability to heat samples at a rate of around 300 °C/min. The fast heating benefits the grain size and nanostructure controlling in the compaction processes for the preparation of magnetic hard/soft nanocomposite magnets, in which the grain size of the magnetically soft phase is critically important. We will demonstrate the experimental results based on the SmCo<sub>5</sub>/α-Fe nanocomposite system in the following paragraphs.

Figure 2 shows the dependence of the density of the bulk samples on the compaction temperature, \( T_{cp} \). One can see that the samples compacted at room temperature have a density of only about 7.4 g/cm<sup>3</sup>, which is about 88% of the theoretical value (8.4 g/cm<sup>3</sup>). The density increases monotonically with increasing \( T_{cp} \) and reaches saturation when \( T_{cp} \) is above 450 to 500 °C. This is attributed to the fact that both SmCo<sub>5</sub> and Fe become more compressible upon heating to high temperatures. The interphase diffusion at high temperatures may also help the consolidation, as reported in our previous work.\(^{13}\) Figure 2 also gives the dependence of the Vickers hardness on \( T_{cp} \) with a load of 500 gs. The hardness increases fast with \( T_{cp} \) up to 500 °C, and then the increase slows down. This also indicates that a nearly full density is obtained when \( T_{cp} \) is above 500 °C.

Figure 3(a) shows the XRD patterns of the bulk SmCo<sub>5</sub> + 20 wt.% α-Fe magnets compacted from amorphous powders at different temperatures. Only broadened peaks for the bcc α-Fe phase were observed when \( T_{cp} \) was lower than 400 °C. It is obvious that the SmCo matrix is amorphous. The crystalline SmCo<sub>5</sub> phase starts to form when \( T_{cp} \) is higher than 500 °C because the crystallization temperature of amorphous SmCo is about 470 °C, according to differential scanning calorimetry measurements. Compaction at 540 °C leads to the formation of a well-developed SmCo<sub>5</sub> phase; however, further increases of \( T_{cp} \) result in sharper peaks of both the SmCo<sub>5</sub> and the α-Fe phases, indicating significant grain growth. A quantitative analysis was performed using the Williamson–Hall method\(^{13,14}\) [Fig. 3(b)]. The measured α-Fe grain size is about 6 nm for the bulk compacted at room temperature. With increasing \( T_{cp} \), the grain size of the α-Fe phase remained constant to ~400 °C and started to increase with \( T_{cp} \) above 450 °C. Fortunately, the grain growth was not excessive, and the grain size is less than 15 nm if \( T_{cp} \) is kept under 550 °C, while significant grain growth is observed when \( T_{cp} \) is higher than 550 °C. It should be noted that the Sm–Co phase was amorphous when \( T_{cp} \) was lower than 470 °C, so there were no data for grain size below that temperature. For comparison, the grain size of single-phase SmCo magnets prepared under same conditions is also given in Fig. 3(b).

TEM micrographs allow for direct observation of the morphology of the nanostructured composites. Figures 4(a) and 4(b) show the morphology and Fe element distribution of the bulk sample compacted from amorphous powders at room temperature. A higher brightness in the EFTEM image corresponds to a higher Fe concentration in the Fe distribution map. One can see that the Fe grain size is as small as 5 nm and the grain distribution is homogenous. It should be noted that codiffusion took place between the Sm–Co and Fe phases, so, actually, the magnetically soft phase contains Co, similar to the bulk nanocomposite magnet prepared by warm compaction.\(^{15,16}\) As \( T_{cp} \) increases to 540 °C, the grain size of

![FIG. 2. (Color online) The dependence of density and Vicker hardness on \( T_{cp} \) of the bulk magnets.](image2)

![FIG. 3. (Color online) (a) XRD patterns of the bulk magnets compacted at different temperatures. (b) The dependence of the grain size of both the SmCo<sub>5</sub> and the α-Fe phases on \( T_{cp} \) of the bulk magnets.](image3)
the α-Fe phase increases to 8 to 15 nm, as shown in Figs. 4(c) and 4(d); this is still within the effective exchange-coupling length. The magnetically soft phase is also well distributed in the Sm–Co matrix. However, when the compaction temperature was increased to 640 °C, the overall grain size significantly increased to 20 to 25 nm, as shown in the TEM image of Fig. 4(e). The TEM analysis confirms the XRD results quite well. For comparison, Fig. 4(f) shows the TEM image of the SmCo5/Fe magnets compacted by conventional hot compaction at 640 °C for 10 min under a pressure of 2 GPa. The grain size (30 to 40 nm) of the conventionally compacted magnets is significantly larger than that of the JHC compacted magnets (20 to 25 nm). The smaller grain size of the JHC compacted sample may be due to the fast and internal heating and cooling.

Figure 5(a) shows the demagnetization curves of the bulk magnets compacted at different temperatures. Similar demagnetization curves with low coercivity were observed for the samples compacted at 20 and 400 °C, because $T_{cp}$ is lower than the crystallization temperature. The crystallization process starts in the 500 °C compacted samples, while a high enough coercivity is obtained only when $T_{cp}$ is higher than 540 °C. The squareness of the demagnetization curves of the 540 °C compacted samples is attributed to the fine grain size below 15 nm, which is close to the effective exchange-coupling length, i.e., twice the domain wall width. A kink was observed in the 640 °C compacted magnets, most likely due to the grain growth and insufficient interphase exchange coupling. Figure 5(b) shows the dependence of the coercivity ($H_c$) and the energy product ($BH_{max}$) on $T_{cp}$ of the bulk nanocomposite magnets. The ($BH_{max}$) of single-phase magnets is also given for comparison. One can see that $H_c$ is only about 1 kOe when $T_{cp}$ is lower than 450 °C, and thus ($BH_{max}$) is low. The coercivity reaches a maximum when $T_{cp}$ is about 540 °C. The maximum ($BH_{max}$) of about 18.5 MGOe—which is 90% higher than that of SmCo5 single-phase nanocrystalline magnets—was also obtained in the 540 °C compacted samples. A further increase of $T_{cp}$ leads to a decrease in ($BH_{max}$) due to the grain growth.

IV. CONCLUSIONS

In summary, a Joule-heating compaction technique has been developed to produce fully dense bulk SmCo5/α-Fe nanocomposite magnets under high pressure at moderate temperatures. The grain size of the magnetic soft phase can be controlled under 15 nm, which ensures effective exchange coupling between the hard and the soft phases. A maximum energy product of 18.5 MGOe has been obtained, which is 90% higher than that of the single-phase counterpart. It was also observed that the grain size of the Joule-heating compacted bulks is smaller than that of the conventional hot pressed bulks prepared under similar conditions.

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