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EFFECT OF SOFT PHASE ON MAGNETIC PROPERTIES OF BULK Sm–Co/α–Fe NANOCOMPOSITE MAGNETS (POSTPRINT)

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14. ABSTRACT
Bulk Sm–Co/*α*–Fe nanocomposite magnets were fabricated by high energy ball milling and subsequent quick hot-pressing. Three different parameters were investigated which are soft phase content, type, and distribution. The effect of Fe soft phase content on the magnetic properties of the final bulk samples was examined. Increasing the Fe content significantly increased the saturation magnetization but at the cost of reduced coercivity. The optimum Fe addition for the highest maximum energy product was determined to be 15 wt% under the present processing conditions. Besides pure Fe a Fe_{49} Co_{49} V_2 powder was also studied as a soft phase addition. The results indicated that FeCoV powder was more readily alloyed with the Sm–Co phase during the milling procedure. For the final bulk magnets, nanoscale Fe particles existed in Sm–Co matrix with pure Fe addition, but with Fe_{49} Co_{49} V_2 addition the soft phase particles were undetectable. Two methods for adding soft phase Fe into hard phase $SmCo_5$ were utilized. The investigation showed that milling Fe and $SmCo_5$ together resulted in a more uniformly distributed mixture of nanoscale soft phase particles, improved squareness of the demagnetization curves of bulk nanocomposite magnets, and higher $(BH)_{max}$ as compared to blending Fe with $SmCo_5$.

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Bulk magnets, coupling, nanocomposite, Sm–Co/*α*–Fe

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Effect of Soft Phase on Magnetic Properties of Bulk Sm–Co/α–Fe Nanocomposite Magnets

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Bulk Sm–Co/α–Fe nanocomposite magnets were fabricated by high energy ball milling and subsequent quick hot-pressing. Three different parameters were investigated which are soft phase content, type, and distribution. The effect of Fe soft phase content on the magnetic properties of the final bulk samples was examined. Increasing the Fe content significantly increased the saturation magnetization but at the cost of reduced coercivity. The optimum Fe addition for the highest maximum energy product was determined to be 15 wt% under the present processing conditions. Besides pure Fe a $Fe_{49}Co_{49}V_2$ powder was also studied as a soft phase addition. The results indicated that FeCoV powder was more readily alloyed with the Sm–Co phase during the milling procedure. For the final bulk magnets, nanoscale Fe particles existed in Sm–Co matrix with pure Fe addition, but with $Fe_{49}Co_{49}V_2$ addition the soft phase particles were undetectable. Two methods for adding soft phase Fe into hard phase $SmCo_5$ were utilized. The investigation showed that milling Fe and $SmCo_5$ together resulted in a more uniformly distributed mixture of nanoscale soft phase particles, improved squareness of the demagnetization curves of bulk nanocomposite magnets, and higher $(BH)_{max}$ as compared to blending Fe with $SmCo_5$.

***Index Terms*—Bulk magnets, coupling, nanocomposite, Sm–Co/α–Fe.**

I. Introduction

MAGNETICALLY hard/soft nanocomposite magnets consist of a hard component with high magnetocrystalline anisotropy and a soft component with high saturation magnetization. In order to take advantage of the outstanding specific magnetic characteristics of each component and to obtain high energy product nanocomposite magnets, the hard and soft phases should be coupled through intergranular magnetic exchange interactions [1], [2]. In theory, this magnetic exchange coupling requires the grain size of the soft phase to be twice the domain wall width of the hard phase. The resulting gain in the saturation magnetization of the composites should depend linearly upon the amount of soft phase. The distribution of hard and soft phases also affects the exchange coupling behavior [2], [3]. The magnetic properties of nanocomposite magnets derive not only from the intrinsic properties of the magnetic phases (hard and soft), but also from the dimensions, content, and distribution of the soft phase. To date, most composite systems have been based on hard phase components such as Nd(Pr)-Fe-B, Sm–Co and soft phase components such as Fe and Fe-Co. Bulk composite magnets have been prepared using compaction techniques such as hot pressing/deformation, dynamic shock compaction, spark plasma sintering, and warm compaction [4][5]–[9]. In our previous study [10], bulk Sm–Co/α–Fe nanocomposite magnets were fabricated by hot pressing of the composite powders prepared by high energy ball milling $SmCo_5$ and Fe powders. The effect of milling time on the magnetic properties of the bulk magnets was studied. In this paper, we further investigated the effect of soft phase content, type and distribution on magnetic properties.

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II. Experiment

As-cast $SmCo_5$ powder with a typical size less than 250 μm was used as the hard magnetic material precursor. Microscale α–Fe and $Fe_{49}Co_{49}V_2$ (FeCoV) powders with particle sizes of 1–3 μm and < 10 μm, respectively, were the soft magnetic material precursors. The $SmCo_5$ powders were combined with 10, 15, and 20 wt% of Fe or FeCoV powders in a stainless steel jar with hardened steel ball media. The powder to ball weight ratio was 1:10. The mixtures were sealed under argon atmosphere and milled using a SPEX 8000 Mixer/Mill for 8 h. A small amount of powder was pulled out every 2 h for XRD analysis. The milled powders were hot pressed at the temperature of 650 °C under a pressure of 400 MPa in vacuum, producing rod-shaped bulk samples 12 mm in length and 8 mm in diameter.

The aforementioned method for adding soft phase Fe into hard phase $SmCo_5$ is the milling process. Another method, described below, is the blending process. Fe powder (1–3 μm) was blended with amorphous $SmCo_5$ powder which was fabricated by high energy ball milling for 8 h. After compaction of the two composite powders at 650 °C, the bulk composite magnets were respectively called the milled sample and the blended sample.

Magnetic properties of the bulk samples at room temperature were measured by a closed-loop hysteresisgraph (model HG-700, KJS Associates, Inc.). The crystal structure, phase fractions, and microstructure were characterized by X-ray diffraction (XRD) with Cu$K\alpha$ radiation and scanning electron microscopy (SEM).

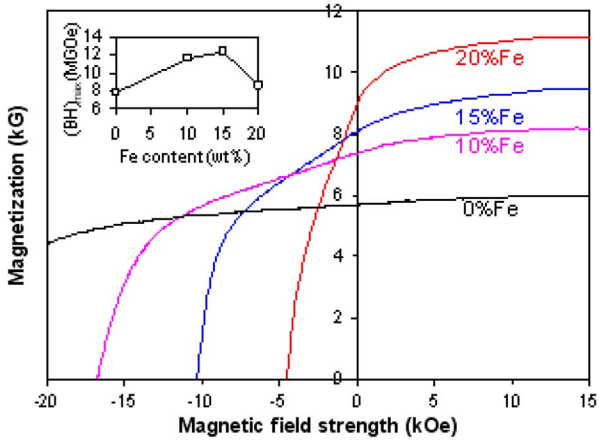

Fig. 1. Demagnetization curves of bulk Sm–Co/α–Fe magnets with different amount of Fe addition and maximum energy product versus Fe content (inset).

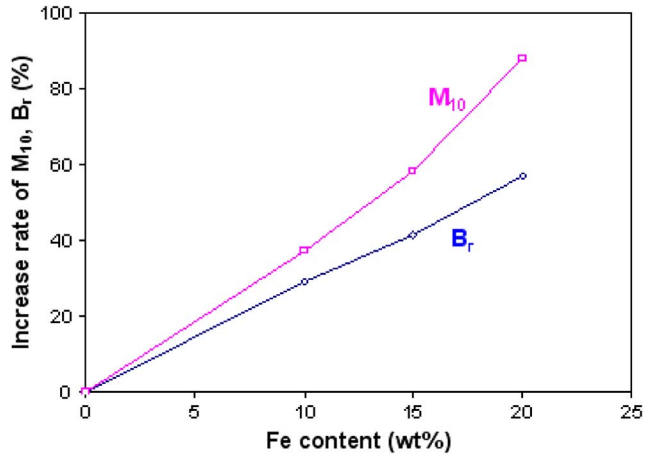

Fig. 2. Dependence on Fe content of the rate of increase of the magnetization at 10 kOe and remanence for the Sm–Co/α–Fe magnets.

III. Results and Discussion

The effect of Fe content on the magnetic properties of Sm–Co/α–Fe magnets was examined. Fig. 1 shows demagnetization curves of the nanocomposite magnets with different amounts of Fe soft phase. As the Fe content increased, the magnetization at the first quadrant significantly increased and the coercivity (H_{ci}) decreased. The opposite trends in the magnetization and the coercivity led to a peak in $(BH)_{max}$ value at 15 wt% Fe addition, as shown in the inset of Fig. 1. For the sample with 20 wt% Fe addition, M_{10} (magnetization at 10 kOe) increased by 88% and H_{ci} decreased by $\sim$82% in comparison to the values of single phase $SmCo_5$ (the sample without the Fe addition), which resulted in the lower $(BH)_{max}$ of 8.64 MGOe. The optimal soft phase Fe addition was determined to be 15 wt% which results in remanence B_r of 8.1 kG, coercivity H_{ci} of 10.3 kOe, and $(BH)_{max}$ of 12.3 MGOe. It is noted that the remanence (B_r) was not enhanced as much as M_{10}. Fig. 2 shows the increase rate of M_{10} and B_r versus Fe addition, which were calculated by $[M_{10}(composite) - M_{10}(SmCo_5)]/M_{10}(SmCo_5)$ and $[B_r(composite) - B_r(SmCo_5)]/B_r(SmCo_5)$, respectively. The increase rate of remanence is lower than that of magnetization at 10 kOe and the difference between B_r and M_{10} becomes larger at higher Fe content, which indicates a poorer exchange coupling behavior.

$Fe_{49}Co_{49}V_2$ as a soft phase was used to prepare hard/soft nanocomposite magnets since it has a higher saturation magnetization. Fig. 3 shows the demagnetization curves of bulk samples with FeCoV and Fe addition. The saturation magnetization of FeCoV (24.5 kG) is higher than that of pure Fe (21.6 kG). However, the magnetization at the first quadrant of the sample with FeCoV addition is lower than that with Fe addition for the same amount of soft phase. The microstructure of the samples was observed by backscattered electron (BSE) imaging in SEM, and the corresponding images are shown in the insets of Fig. 3. For the $SmCo_5 + Fe$ sample the Fe particles (dark grey) are uniformly distributed in the Sm–Co matrix (light gray). However, it was hard to find any FeCoV particles within the entirety of the $SmCo_5 + FeCoV$ sample. The possibility that FeCoV particles do not exist in a single soft phase was confirmed by the XRD results shown in Fig. 4. For the bulk sample with Fe addition, the diffraction peaks indicated $TbCu_7$ type Sm–Co 1:7 and Fe phases exist. For the bulk sample with FeCoV addition, all of the diffraction peaks were determined to be Sm–Co 1:7 phase, and there was no soft phase peak, which matches with the SEM results. The soft phase peak was not even present for the as-milled powder [Fig. 4(b)], which indicates that the FeCoV soft phase did not exist in the milled powder before compaction and it should be mechanically alloyed into the hard phase after 8 h milling. Fig. 5 shows XRD patterns of as-milled powders with different milling time. The soft phase diffraction peak was present up to only 4 h of milling. The results indicate that FeCoV powder is more readily mechanically alloyed with the Sm–Co phase during the milling procedure. Although the $Fe_{49}Co_{49}V_2$ soft phase has a higher saturation magnetization, it is not suitable as a soft phase in the present process.

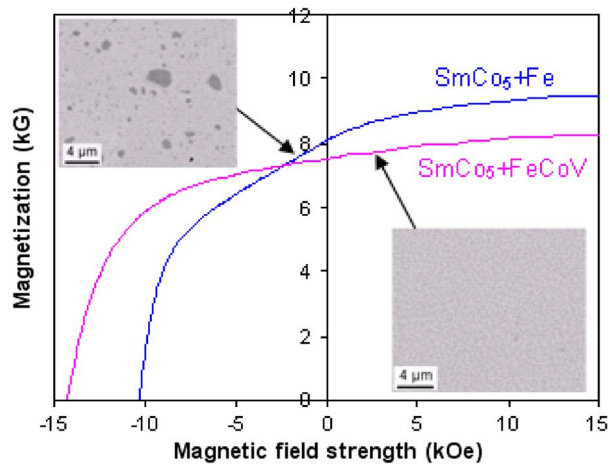

Fig. 3. Demagnetization curves for bulk Sm–Co/α–Fe magnets pressed at 650 °C after milling for 8 h with starting composition of $SmCo_5 + 15$ wt% soft phase (Fe and FeCoV); BSE images of the bulk samples (insets).

To study the effect of soft phase distribution on the magnetic properties of bulk Sm–Co/α–Fe composite magnets, the milled sample and the blended sample were prepared. Fig. 6 presents the microstructure of the two bulk samples. For the milled sample, Fe soft phase (black) distributed separately and

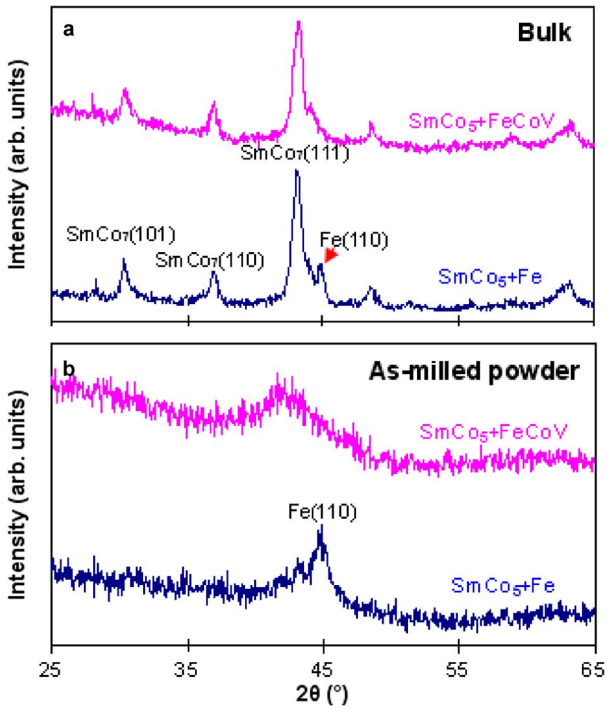

Fig. 4. XRD patterns for (a) bulk samples and (b) as-milled powders with the starting composition of $SmCo_5 + 15$ wt% soft phase (Fe and FeCoV) after milling for 8 h.

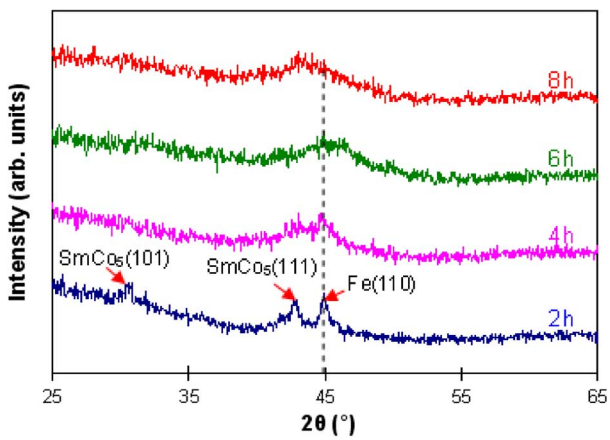

Fig. 5. XRD patterns for as-milled powders with the starting composition of $SmCo_5 + 15$ wt% FeCoV after milling for 2, 4, 6, and 8 h.

uniformly in the Sm–Co matrix [Fig. 6(a)]. Most of the Fe particles existed with particle size of a few hundred nanometers. There were several larger particles that were deformed but not broken into small pieces with particle sizes of a few micrometers. For the blended sample, there were some regions where Fe particles aggregated into clusters of around 10 μm [Fig. 6(b)]. However, in most other regions the Fe phase could not be observed. Thus, the Fe soft phase in the blended sample was distributed inhomogeneously within the hard phase matrix.

Fig. 7 shows demagnetization curves of the two samples. For the blended sample the magnetization dramatically decreased around zero field, which is characteristic of a soft phase. Although the blended Sm–Co/Fe sample has relatively high magnetization at the first quadrant, its remanence B_r dropped to

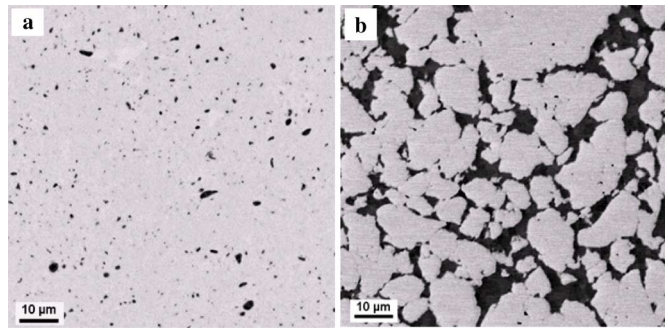

Fig. 6. BSE images for bulk Sm–Co/α–Fe composite magnets pressed at 650°C: (a) milled sample of $SmCo_5 + 15$ wt% Fe for milling of 8 h; (b) blended sample of amorphous $SmCo_5$ with 15 wt% Fe.

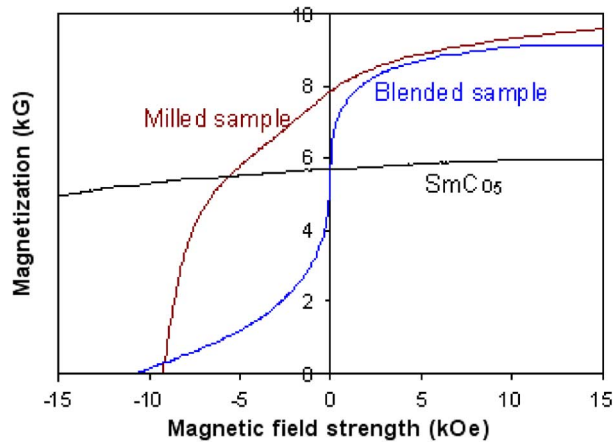

Fig. 7. Demagnetization curves for $SmCo_5$ magnet and Sm–Co/α–Fe composite magnets prepared from blended and milled powder.

the same value as the single phase $SmCo_5$ magnet. The ratio of B_r/M_{10} was only 0.61 and the curve showed a deep downward shoulder in the second quadrant. The results indicated that there was little magnetic coupling between the hard and soft phases due to the aggregation and uneven distribution of the soft phase. For the milled sample, the shape of the curve was greatly improved and a higher remanence value was obtained ($B_r/M_{10} = 0.84$). The milled composite magnet exhibited better magnetic coupling between the hard and soft phases owing to a uniform nanoscale distribution of the soft phase.

XRD patterns for the two bulk samples are shown in Fig. 8. The blended sample consisted of Sm–Co 1:5 and Fe phases; the milled sample consisted of Sm–Co 1:7 and Fe phases. It is also noted the intensity ratio of soft phase peak (110) to hard phase peak (111) for the blended sample is higher than that of the milled sample. These results are caused by interdiffusion between Fe and Co in the composite. The mechanical alloying and thermal diffusion took place during milling procedure and pressing procedure, respectively [10], [11]. For the blended sample there is no diffusion or mechanical alloying during the mixing process (as opposed to the milled sample). For this sample, thermal diffusion might occur in the pressing procedure. Interdiffusion should be weak due to the lower Sm–Co/Fe interface for microscale Fe particles. For the milled sample, diffusion took place during both the milling and pressing procedures, with a stronger tendency of thermal diffusion during

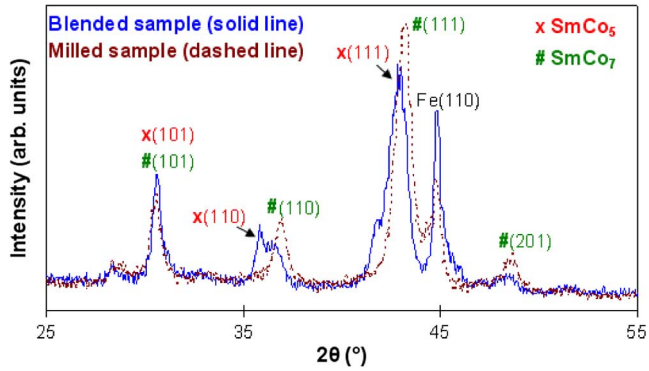

Fig. 8. XRD patterns for bulk Sm–Co/α–Fe composite magnets prepared from blended powder and milled powder.

the pressing procedure due to larger interface area between soft phase and hard phase for nanoscale Fe particles. However, the interdiffusion under the present process parameters have not destroyed the magnetic properties, it seems that 1:7 phase appearance and FeCo formation are gainful for magnetization enhancement [10], [12]. With a large amount of interdiffusion there will be more soft phase loss, which will result in a reduced saturation magnetization. By controlling the milling and pressing process and optimizing the parameters, it is possible to control the interdiffusion between the phases to obtain the nanocomposite magnets with the best properties.

IV. Conclusions

In summary, the soft phase content, distribution, and type were investigated for bulk Sm–Co/α–Fe magnets prepared by high energy ball milling and subsequent hot-pressing. Soft phase Fe content significantly affected on saturation magnetization, remanence, and coercivity of the composite magnets. The energy product obtained the maximum value with 15 wt% Fe addition. Fe distribution mainly affected the shape of demagnetization curve and therefore affected the energy product. The milled composite magnet showed a more uniform distribution of nanoscale soft phase, improved squareness of the demagnetization curves, and higher $(\mathrm{BH})_{\max}$ as compared to blended composite magnet. $Fe_{49}Co_{49}V_2$ powder was readily mechanically alloyed with Sm–Co in the milling procedure and is not suitable as a soft phase using the milling procedure.

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