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## The effect of grain size on magnetic properties in $\text{Sm}_2\text{Fe}_{17}\text{N}_x$

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The microstructures of melt spun  $\text{Sm}_{11}\text{Fe}_{89}$  and  $(\text{Sm}_{11}\text{Fe}_{89})_{94}\text{Ti}_3\text{C}_3$  have been investigated under different solidification conditions. The magnetic properties of the nitrided materials have been related to the observed microstructures. Melt spinning of the  $\text{Sm}_{11}\text{Fe}_{89}$  alloys resulted in grain sizes at or above the single domain limit. The addition of Ti and C resulted in an order of magnitude refinement in the microstructural scale. The magnetic properties of the samples with grain sizes below the single domain limit decreased with decreasing grain size due to increased intergranular exchange coupling. © 1998 American Institute of Physics. [S0021-8979(98)08610-1]

There has been much interest in the interstitial compound  $\text{Sm}_2\text{Fe}_{17}\text{N}_x$  because of its superior magnetic properties.<sup>1,2</sup> The base  $\text{Sm}_2\text{Fe}_{17}$  compound is rhombohedral of the  $\text{Th}_2\text{Zn}_{17}$ -type structure. This structure can be derived from the  $\text{CaCu}_5$  structure through the ordered substitution of a pair of Fe “dumbbell” atoms for one third of the Sm atoms.<sup>3</sup> The  $\text{Sm}_2\text{Fe}_{17}$  structure is closely related to the  $\text{TbCu}_7$ -type  $\text{SmFe}_7$  structure. In this case, substitution of the Fe dumbbells for Sm occurs randomly, resulting in the  $\text{SmFe}_7$  structure being a “disordered” derivative of the  $\text{Sm}_2\text{Fe}_{17}$  structure.

The binary  $\text{Sm}_2\text{Fe}_{17}$  compound, which is subsequently nitrided to form a hard magnetic compound, has been successfully produced by melt spinning rapid solidification.<sup>4–6</sup> The alloy composition strongly influences the phase formation and the structural nature of the  $\text{Sm}_2\text{Fe}_{17}$  compound. For instance, at high Sm content (above 17 at. %) an amorphous structure formed.<sup>4</sup> At lower Sm content, a mixture of the  $\text{SmFe}_7$  and  $\text{SmFe}_2$  phases was observed.<sup>5,6</sup> Close to the  $\text{Sm}_2\text{Fe}_{17}$  stoichiometry, the amount of Fe dumbbell ordering was observed to depend on the Sm content, as revealed by x-ray diffraction analysis.<sup>5</sup> Hyperstoichiometric alloys tended to form the more ordered  $\text{Sm}_2\text{Fe}_{17}$  structure. The ordering decreased as the Sm content decreased, and hypostoichiometric alloys displayed a tendency toward the disordered  $\text{SmFe}_7$  structure. A transition to the  $\text{ThMn}_{12}$ -type tetragonal structure was observed as the Sm content was further decreased.

While the structural state of rapidly solidified Sm–Fe alloys has been investigated, the microstructural evolution and the effect of the microstructural scale on the magnetic properties of  $\text{Sm}_2\text{Fe}_{17}\text{N}_x$  have not. In this communication, we report the grain size dependence of the magnetic properties as well as on grain refinement during rapid solidification through alloying additions.

Samples with nominal compositions of  $\text{Sm}_{11}\text{Fe}_{89}$  and  $(\text{Sm}_{11}\text{Fe}_{89})_{94}\text{Ti}_3\text{C}_3$  were arc melted from high purity elemental constituents. The arc melted ingots were then melt spun at tangential velocities ranging from 15 to 40 m/s utilizing a copper wheel. Samples were characterized by transmission electron microscopy utilizing a JEOL 2000FXII transmission electron microscope. The magnetic properties of nitrided samples were measured with a PARC vibrating sample magnetometer, which has a maximum field of 2.3 T, after pulse magnetization at 6.5 T. Samples were nitrided at 550 °C for 4 h in flowing  $\text{N}_2$ . The nitrogenation was confirmed by x-ray diffraction, which revealed peak shifts characteristic of the presence of interstitial nitrogen in the  $\text{Sm}_2\text{Fe}_{17}$  structure.

Selected area electron diffraction patterns of rapidly solidified  $\text{Sm}_{11}\text{Fe}_{89}$  were indexed to the  $\text{Sm}_2\text{Fe}_{17}$  structure. However, reflections arising from the Fe dumbbell ordering were relatively weak, indicating only partial ordering on the dumbbell sites (Fig. 1). This partial ordering was characteristic of all of the  $\text{Sm}_{11}\text{Fe}_{89}$  alloys regardless of wheel speed.

The scale of the microstructure of the binary  $\text{Sm}_{11}\text{Fe}_{89}$  alloys was proportional to the solidification rate (i.e., wheel velocity). At 15 m/s, the grain size was approximately 800 nm, while at 40 m/s the grain size was approximately 330 nm (Fig. 2). A highly mottled contrast was observed within the grains of the sample melt spun at 15 m/s, which was due to the extensive antiphase domain structure resulting from the disordered Fe dumbbell configuration. Annealing experiments at 800 °C eliminated this domain structure, increased the amount of ordering, and resulted in stacking faults along the *c* axis. The stacking faults in this case are essentially antiphase domains, as the ordering of the Fe–Fe dumbbells along the *c* axis is occasionally out of order.

The compound addition of Ti and C to the base  $\text{Sm}_{11}\text{Fe}_{89}$  alloy resulted in a refinement of the microstructure by approximately an order of magnitude compared to the binary alloys. Evaluation of the electron diffraction ring patterns revealed the presence of the  $\text{SmFe}_7$  structure. The ordering

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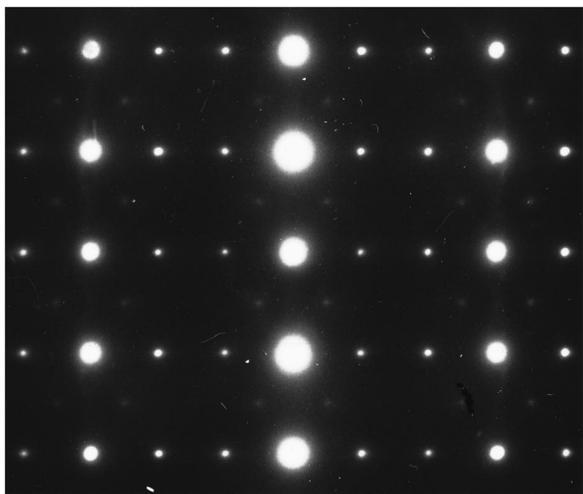


FIG. 1. Selected area electron diffraction pattern of the [1010] zone axis of  $\text{Sm}_{11}\text{Fe}_{89}$  melt spun at 30 m/s. The weak superlattice reflections indicate partial ordering of the Fe dumbbells.

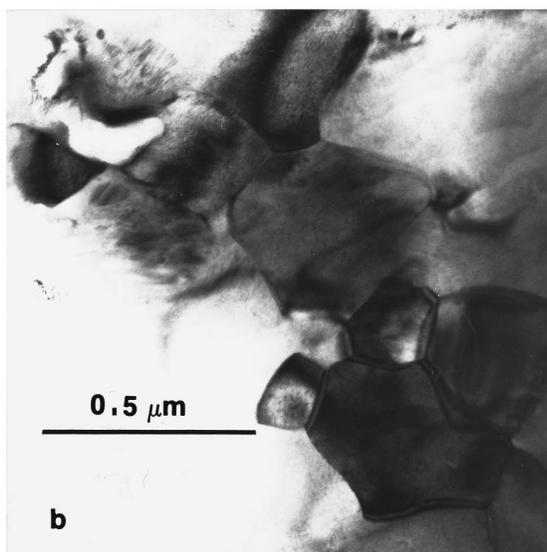
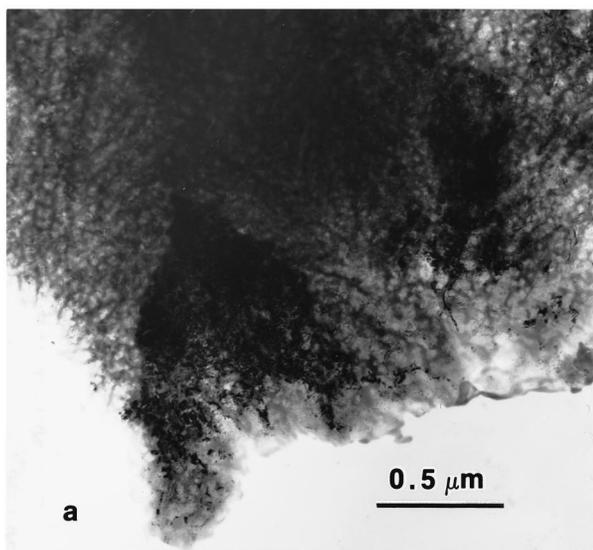


FIG. 2. Bright field TEM images of  $\text{Sm}_{11}\text{Fe}_{89}$  melt spun at (a) 15 m/s and (b) 40 m/s.

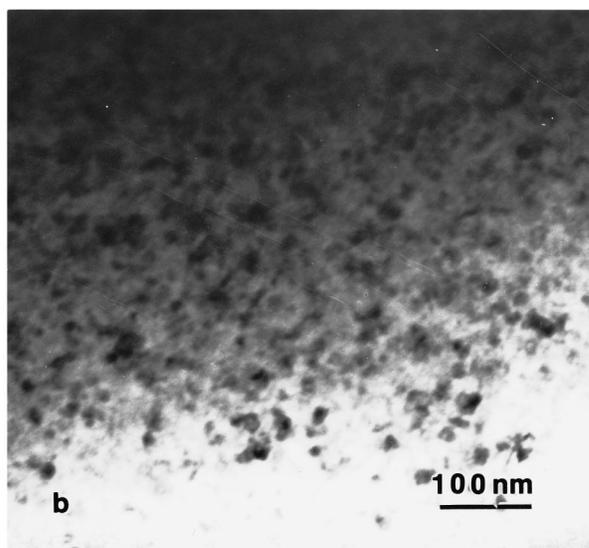
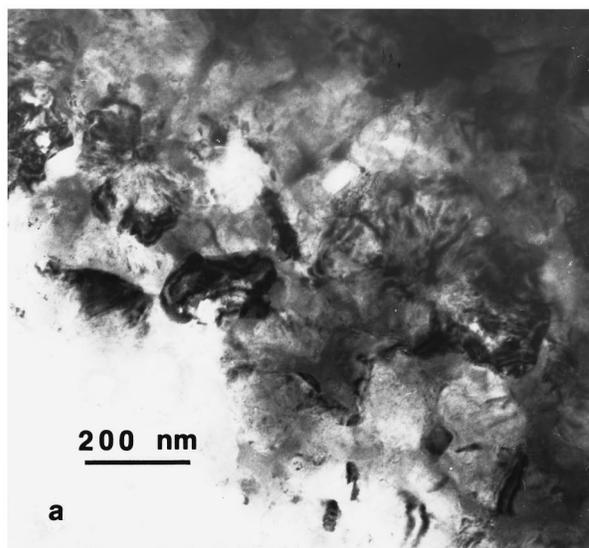


FIG. 3. Bright field TEM image of  $(\text{Sm}_{11}\text{Fe}_{89})_{94}\text{Ti}_3\text{C}_3$  melt spun at (a) 20 m/s and (b) 40 m/s.

was more difficult to characterize, as the grain size was smaller than the selected area aperture, making single crystal alignment and diffraction difficult. The grain size varied from approximately 150 nm for  $(\text{Sm}_{11}\text{Fe}_{89})_{94}\text{Ti}_3\text{C}_3$  melt spun at 20 m/s to 25 nm for this alloy melt spun at 40 m/s (Fig. 3). The grain morphology also became more equiaxed than in the binary Sm–Fe alloy. Compound additions of Ti and C also affected the solidification behavior in Nd–Fe–B alloys.<sup>7</sup>

The magnetic properties were measured on samples after nitriding at 550 °C for 4 h in flowing  $\text{N}_2$ . X-ray diffraction was used to confirm the formation of the  $\text{Sm}_2\text{Fe}_{17}\text{N}_x$  compound. The nitrided  $\text{Sm}_{11}\text{Fe}_{89}$  alloys melt spun at 15, 20, and 30 m/s displayed coercivities of approximately 1.4 kOe and remanent ratios  $M_1/M_s$  of approximately 0.3. The nitrided  $\text{Sm}_{11}\text{Fe}_{89}$  melt spun at 40 m/s showed a higher coercivity of 4 kOe and a remanent ratio of 0.5. For the nitrided  $(\text{Sm}_{11}\text{Fe}_{89})_{94}\text{Ti}_3\text{C}_3$  samples, the maximum magnetic properties were observed in the sample melt spun at 20 m/s. At

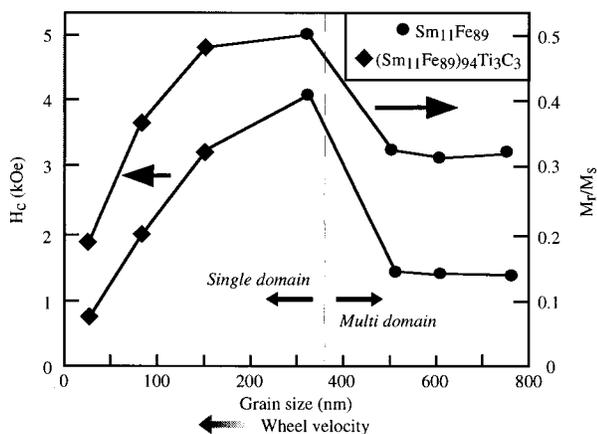


FIG. 4. The coercivity and remanent ratio of nitrated  $\text{Sm}_{11}\text{Fe}_{89}$  and  $(\text{Sm}_{11}\text{Fe}_{89})_{94}\text{Ti}_3\text{C}_3$  displayed a strong dependence on grain size.

higher wheel speeds, the coercivities and remanent ratios decreased. This decrease in magnetic properties was attributed to a grain size effect and not due to changes in the intrinsic magnetic properties of the alloyed samples. The shapes of the hysteresis loops also became more wasp-waisted as the grain size decreased. It should also be mentioned that the coercivities observed here are significantly lower than previously observed for melt spun  $\text{Sm}_2\text{Fe}_{17}\text{N}_x$  alloys,<sup>5,6</sup> possibly due to the presence of  $\alpha$ -Fe. The presence of  $\alpha$ -Fe is difficult to avoid since the  $\text{SmFe}_2$  phase decomposes to  $\text{SmN}$  and  $\alpha$ -Fe in a nitrogen-bearing atmosphere,<sup>8</sup> and partial decomposition of the  $\text{Sm}_2\text{Fe}_{17}$  occurs during nitrogenation.

Figure 4 illustrates the strong dependence of magnetic properties on the grain size. In  $\text{Sm}_2\text{Fe}_{17}\text{N}_x$  the single domain limit is approximately 360 nm.<sup>9</sup> Above this grain size, the magnetic properties drop off sharply as the grains are in a

multidomain state with minimal resistance to domain wall movement. Below the single domain grain size, the coercivity rapidly decreases with decreasing grain size. The loss of coercivity can be attributed to increased exchange interactions as the grain size decreases.

The grain size of binary  $\text{Sm}_{11}\text{Fe}_{89}$  alloys was dependent on the solidification rate. However, grain sizes of samples melt spun at tangential velocities of 30 m/s and less were larger than the single domain limit. Alloying additions of Ti and C resulted in a refinement of the grain size below the single domain limit. The magnetic properties of the  $\text{Sm}_2\text{Fe}_{17}\text{N}_x$  compound were strongly dependent on grain size both above and below the single domain limit. The dependence below the single domain limit was related to increased exchange interactions between grains.

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