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Structural and magnetic properties of mechanically milled SmCo$_5$:C

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Mechanically milling SmCo$_5$ powder significantly increases coercivity and remanence ratio by introducing defects; however, these defects can be removed by room-temperature aging, with a resultant decrease in coercivity. A series of $(SmCo_5)_x$:C$_{1-x}$ ($0.15 \leq x \leq 1$) samples has been fabricated to investigate the effect of C on oxidation protection and magnetic properties. SmCo$_5$ was premilled for 1 h, then added to C powder and milled for times ranging from 15 min to 7 h. X-ray diffraction indicates the presence of crystalline graphite and SmCo$_5$ for milling times ≤ 6 h and also shows the presence of fcc Co for milling times >7 h. The magnetic properties are very weakly dependent on milling time after the C addition, which is attributed to the lack of further grain refinement. The saturation magnetization scales linearly with the wt % of SmCo$_5$. Remanence ratios are approximately 0.7 and independent of volume fraction. The maximum coercivity of 16.5 kOe is comparable to the maximum obtained by milling SmCo$_5$ without C. Samples exposed to air for times up to two months show no decrease in coercivity or remanence ratio for $x \leq 0.70$. The addition of C has no detrimental effect on the magnetic properties obtained by milling, except the expected reduction of M_s. © *2000 American Institute of Physics.* [S0021-8979(00)24508-8]

I. INTRODUCTION

Mechanical milling is useful for producing nonequilibrium magnetic structures[1-3] that cannot be produced by other techniques. Mechanical milling significantly increases the coercivity (H_c) and remanence ratio (M_r/M_s) of SmCo$_5$ by introducing point defects.[4,5] Room-temperature aging removes many of these defects, decreasing H_c by up to a factor of 2. Other studies have shown that encapsulation of magnetic nanoparticles in carbon can enhance oxidation resistance and increase nanostructure stability.[6-8] This article details the effect of carbon on the magnetic properties and oxidation resistance of $(SmCo_5)_x$:C$_{1-x}$ with $0.15 \leq x \leq 1$.

II. EXPERIMENTAL DETAILS

Mechanical milling was carried out using a tungsten-carbide vial and balls in an argon-filled glovebox to prevent oxidation. A 3:1 ball-to-powder mass ratio was used. Powders were milled for 15 min followed by a cooling period of 15 min. All times reported here are the times during which the powder was actively milled.

SmCo$_5$ powder (-100 mesh) was milled for one hour before graphite powder (-325 mesh) was added to produce $(SmCo_5)_x$:C$_{1-x}$, with $0.15 \leq x \leq 1$. A small amount of sample from each concentration was aged in air for times ranging from ten days to two months for oxidation studies. The remainder of the powder was kept in the argon atmosphere. The diffracting crystallite size (DCS) is determined from x-ray diffraction measurements using the Scherrer equation.[9]

Samples for measurement in a SQUID magnetometer were placed in paraffin-filled polyethylene bags and sealed. The samples not aged in air were prepared and sealed in an argon atmosphere. The paraffin was melted to immobilize the particles and prevent whole-particle motion.

III. RESULTS AND DISCUSSION

A. Dependence of magnetic properties on milling time

Figure 1 shows the x-ray diffraction patterns for the $x = 0.15$ sample for milling times from 0.25 to 7 h. This sample is representative of samples with volume fractions up to 0.70. Samples milled for ≤ 6 h indicate a mixture of SmCo$_5$ and graphite. Samples milled for ≥ 7 h show SmCo$_5$, graphite, and additional peaks due to fcc Co. The broad
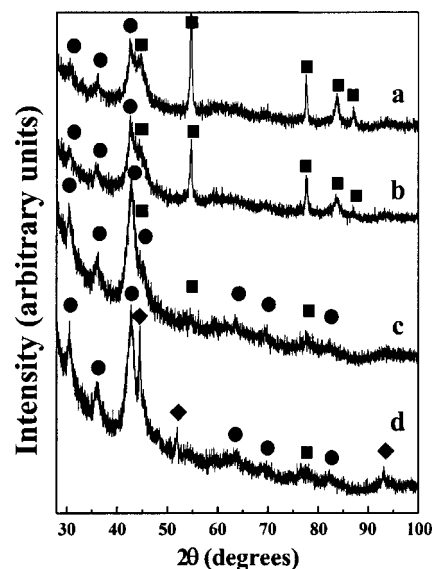

FIG. 1. X-ray diffraction patterns of $(SmCo_5)_{0.15}$:C$_{0.85}$ milled for (a) 0.25 h, (b) 1 h, (c) 6 h, and (d) 7 h. Diffraction peaks corresponding to graphite are denoted by squares, SmCo$_5$ by filled circles, and fcc Co by diamonds.

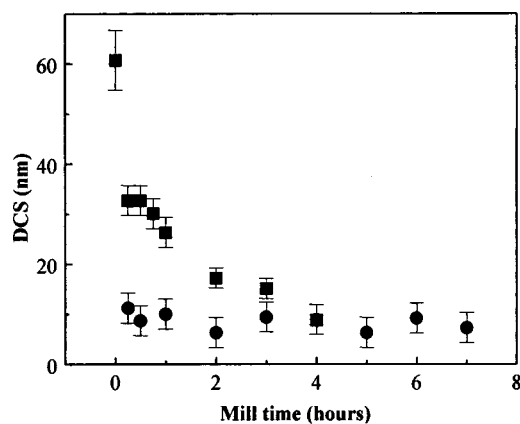[a)]Electronic mail: diandra2@unl.edu

FIG. 2. Diffracting crystallite sizes (DCSs) of graphite (squares) and SmCo$_5$ (circles) for $(SmCo_5)_{0.15}:C_{0.85}$ as a function of milling time.

FIG. 3. Linear relationship between the saturation magnetization and wt % SmCo$_5$ in the $(SmCo_5)_x:C_{1-x}$ samples.

SmCo$_5$ peaks are indicative of the relatively small DCS (~9 nm), which does not change significantly with milling time after the premill, indicating that the carbon powder prevents additional grain refinement. The graphite, which was not premilled, shows a sharp decrease in DCS from 61 ± 6 to 33 ± 3 nm after the first 15 min of milling. The DCS decreases to 9 ± 1 nm after 4 h of milling, as shown in Fig. 2. The graphite peaks for milling times >4 h are broad and weak, making it difficult to obtain accurate DCSs.

The magnetic properties do not change significantly after the first 15 min of milling, which is consistent with the observed constancy of the DCS. The average saturation magnetization (M_s) is 26 ± 3 emu/g, where all magnetization measurements are given in emu per gram of $(SmCo_5)_x:C_{1-x}$ sample. The average remanence ratio (M_r/M_s) is 0.73 ±0.02, and the average coercivity (H_c) is 12.7 ± 0.3 kOe.

B. Dependence of magnetic properties on SmCo$_5$ volume fraction

The structural properties do not change significantly as a function of SmCo$_5$ volume fraction. After 1 h of milling, the SmCo$_5$ DCS is approximately the same (9.2 ± 0.4 nm) over the entire range of volume fractions. Changes in magnetic properties therefore are not due to changes in the DCS. SmCo$_5$ is present in all patterns and graphite peaks are also observed for $x \leqslant 0.30$.

Figure 3 shows the dependence of M_s on wt % SmCo$_5$. The linear relationship between M_s and wt % SmCo$_5$ is described by $M_s = 0.64$ wt %. Extrapolation to 100 wt % indicates $M_s = 64.3$ emu/g, which is lower than expected for bulk SmCo$_5$; however, this decrease in M_s is typical of milled SmCo$_5$.[4]

Figure 4 shows H_c and M_r/M_s as functions of the volume fraction, x. M_r/M_s is relatively independent of volume fraction, with an average value of 0.72. All values of H_c are higher than the nonmilled SmCo$_5$ starting powder. Since no grain refinement occurs after the addition of carbon, the enhanced coercivity relative to the unmilled powder is attributed to the one-hour premill of the SmCo$_5$ prior to the addition of carbon. H_c increases as x increases, reaching a broad maximum of 16.5 kOe, which is comparable to the maxi-
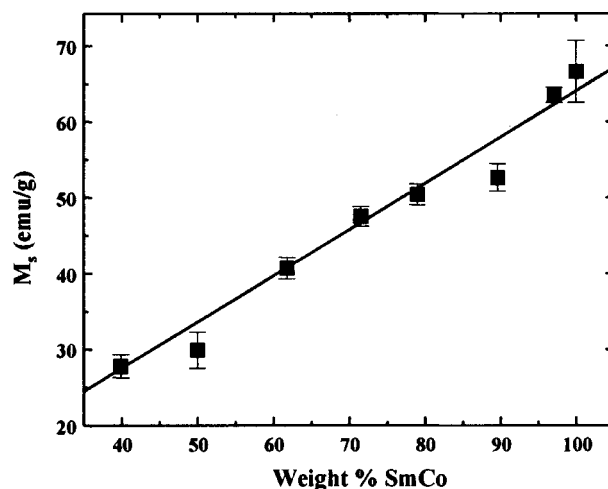
mum obtained by milling SmCo$_5$ only.[4] H_c decreases slightly for $x \geqslant 0.5$. The dependence of H_c on x is not due to a grain size effect, as the DCS is relatively constant for all volume fractions at this milling time. One possible explanation for this behavior is that, as x increases, the distance between SmCo$_5$ grains decreases, increasing the intergrain exchange.

C. Oxidation resistance

Figure 5 compares the room-temperature hysteresis loops of the protected (''nonoxidized'') and air-aged (''oxidized'') $(SmCo_5)_{0.90}:C_{0.10}$ systems. The hysteresis loop of the oxidized sample is not significantly different from the nonoxidized sample. Similar behavior is seen for other volume fractions. The open triangles in Fig. 6 show that the
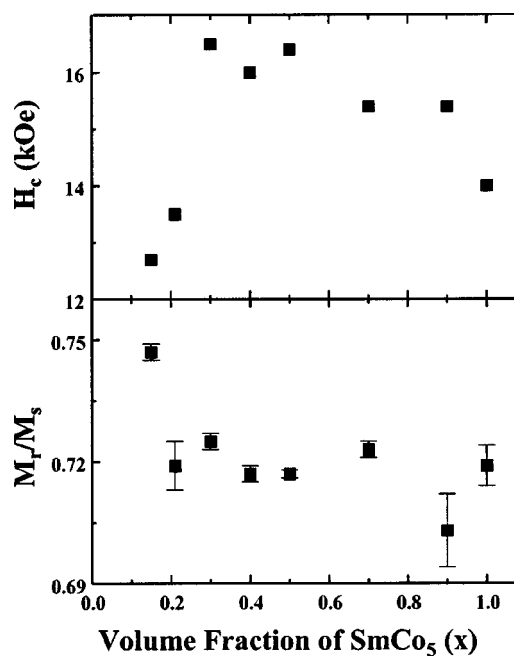

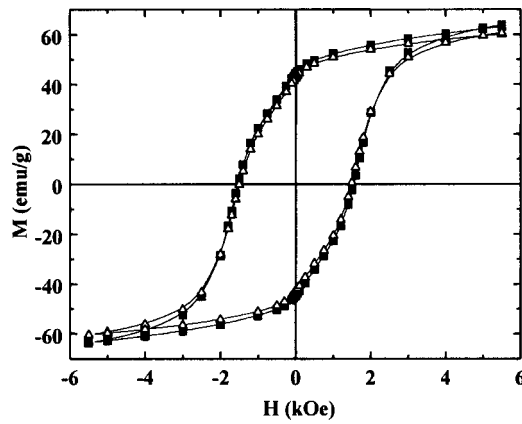FIG. 4. Coercivity and remanence ratio of $(SmCo_5)_x:C_{1-x}$ samples as functions of x.

FIG. 5. Representative room-temperature hysteresis loop for the $x=0.90$ sample before oxidation (squares) and after oxidation (open triangles).

coercivities and remanence ratios of the oxidized samples for all volume fractions are similar to those of the nonoxidized samples.

The saturation magnetizations of oxidized samples with $x \leq 0.70$ are higher, sometimes by 20%, than the correspond-
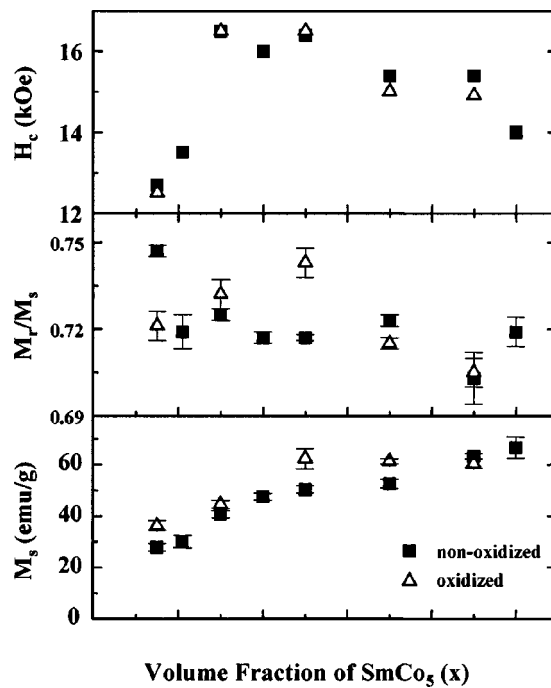

FIG. 6. Coercivity, remanence ratio, and saturation magnetization of $(SmCo_5)_x:C_{1-x}$ samples as functions of x for both nonoxidized (squares) and oxidized (open triangles) samples. In the top graph, the error bars are approximately the same size as the symbols.

ing nonoxidized samples. The increase in M_s is consistent with observed increases after comparable periods of room-temperature aging;[5] however, the increase in M_s in aged, milled $SmCo_5$ was accompanied by significant decreases in H_c and M_r/M_s, which are not observed in these samples. Only the sample with $x=0.90$ shows a decrease in M_s after exposure to air, possibly indicating that a threshold amount of carbon is necessary for increased oxidation resistance.

IV. CONCLUSIONS

The enhanced magnetic properties resulting from mechanically milling $SmCo_5$ can be retained and the oxidation resistance increased by the addition of carbon. Carbon prevents additional grain refinement for all volume fractions up to 1 h, and for all times up to 6 h for $x \leq 0.70$. M_s scales linearly with wt % $SmCo_5$, with the extrapolated $x=1$ value consistent with that obtained from mechanically milled $SmCo_5$. M_r/M_s is independent of volume fraction, while H_c displays a broad peak with a maximum value of 16.5 kOe. The coercivity and remanence ratio do not change significantly for any concentration. The saturation magnetization of oxidized samples with $x \leq 0.70$ are comparable to or higher than the corresponding nonoxidized samples. The lack of a decrease in saturation magnetization suggests that carbon protects the sample from oxidation. The slight increase in M_s may be due to aging effects. The sample with $x=0.90$ departs from this trend, suggesting that there is a threshold amount of C necessary to provide oxidation resistance. Further studies, including transmission electron microscopy to elucidate the nanostructure as a function of volume fraction, are necessary to understand the role of carbon in this system.

ACKNOWLEDGMENTS

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