Advanced Materials '93, I / B: Magnetic, Fullerene, Dielectric, Ferroelectric, Diamond and Related Materials, edited by M. Homma et al.
Trans. Mat. Res. Soc. Jpn., Volume 14B

MAGNETIZATION PROCESSES IN HIGH COERCIVITY $SmCo_5$ AND $Sm_2Fe_{17}N_x$ PREPARED BY MECHANICAL ALLOYING

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A study of the magnetic behaviour of nanocrystalline $SmCo_5$ prepared by mechanical alloying has been carried out. The measurements show that exchange interaction between grains may play an important role in the irreversible magnetization process. The exchange coupling between soft and hard phases of α-Fe and $Sm_2Fe_{17}N_x$ has also been studied by magnetization process measurements and viscosity tests.

1. Introduction

Coercivities exceeding 35 kOe and 55 kOe have been attained in mechanically alloyed $Sm_2Fe_{17}N_x$ [1] and $SmCo_5$ [2], respectively. Recently, remanence enhancement was observed in mechanically alloyed α-Fe - $Sm_2Fe_{17}N_x$ powders [3]. These studies have shown that mechanical alloying is a very useful method for preparation of permanent magnet materials.

In this paper, we report magnetization measurements of nanocrystalline $Sm_2Fe_{17}N_x$ and $SmCo_5$ prepared by mechanical alloying and also discuss results of magnetic viscosity tests.

2. Experimental

Isotropic $SmCo_5$ specimens were pressed into cylinders, and annealed at 800 °C after mechanical alloying elemental powders with a starting composition of $Sm_{19}Co_{81}$ [2].

Single phase $Sm_2Fe_{17}N_x$ and two phase α-Fe - $Sm_2Fe_{17}N_x$ samples were also prepared by mechanical alloying and heat treatment. Using the starting compositions of $Sm_{13}Fe_{87}$ and $Sm_{14}Fe_{86}$, the powders consisted of the single Sm_2Fe_{17} phase after vacuum heat treatment at temperatures of 650-800 °C. The $Sm_2Fe_{17}N_x$ phase was formed after nitriding at 400 °C [1]. Lower Sm concentrations lead to formation of a mixture of α-Fe and $Sm_2Fe_{17}N_x$ after heat treatment [3]. Two remanence enhanced specimens with starting compositions of Sm_7Fe_{93} and $Sm_{10}Fe_{90}$, respectively, were annealed at 600 °C for 1 hour before nitriding.

Magnetic measurements were performed at room temperature using a Vibrating Sample Magnetometer (VSM3001, Oxford Instrument Company) with a maximum applied field of 120 kOe. Measurements of the remanence $M_r(H)$ and the coercivity $H_c(H)$ were taken from minor loops with increasing field H, using samples which were thermally demagnetized before the measurement. The demagnetization remanence $M_d(H)$ [5,6] was measured after demagnetization at -H, after the sample had been saturated at a positive field of 120 kOe. Values of magnetic susceptibilities χ, viscosity coefficient S and parameter Λ were measured from viscosity tests [4].

3. Results and Discussion

3.1. $SmCo_5$

The coercive force H_c and the remanence M_r are plotted in Fig. 1 as function of the applied field H. H_c and M_r initially remain small and increase rapidly in the field range of 20-60 kOe. The saturation of H_c and M_r requires fields of 80-100 kOe, which are much higher than the maximum coercive force of 57 kOe for this sample. Such properties are typical of nanocrystalline materials with grain sizes less than the single domain particle size [2].

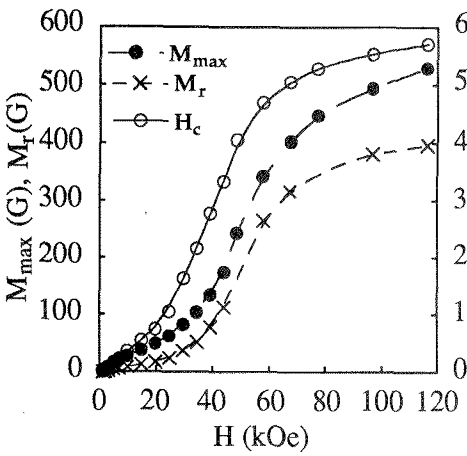

Fig. 1: Coercive force H_c, maximum magnetization M_{max} and remanence M_r as function of the field H for $SmCo_5$.

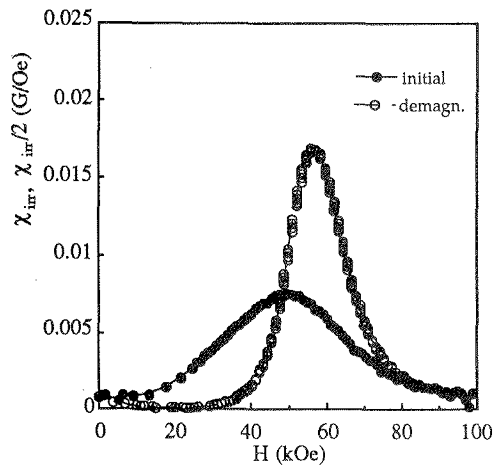

Fig. 2: The irreversible susceptibility χ_{irr} measured at the initial curve and $\chi_{irr}/2$ taken at the demagnetization curve for $SmCo_5$.

The irreversible susceptibility χ_{irr} is given as function of field H in Fig. 2. The curve of χ_{irr} for initial magnetization and the curve of $\chi_{irr}/2$ for demagnetization should be identical, if the magnetization process is same for initial magnetization and magnetization reversal. The measurements of χ_{irr} showed that the magnetization process is different for the initial magnetization and the demagnetisation. The measured irreversible susceptibility χ_{irr} for the initial magnetization has a much broader shape than that for the demagnetization, and also the position of maximum shifts to lower fields. This behaviour can be explained by the existence of strong exchange interactions between grains [5].

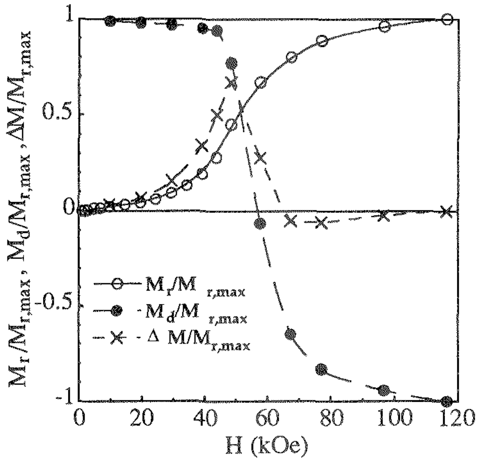

Fig. 3: Relative remanence $M_r/M_{r,max}$, relative demagnetization remanence $M_d/M_{r,max}$ and the relative difference $\Delta M_d/M_{r,max}$ between the measured M_d value and that calculated Md value from the Wohlfarth relationship ($M_d(H) = M_{r,max} - 2 \cdot M_r(H)$).

In order to study the interaction, the demagnetization remanence M_d was measured as function of the remanence according to the Wohlfarth relationship [6] of $M_d(H) = M_{r,max} - M_r(H)$, which is expected to be satisfied for the system of non-interacting single domain particles. Deviation of M_d from the Wohlfarth relationship has been interpreted as resulting from presence of interaction fields [5].

The relative deviation of M_d is plotted in Fig. 3. Significant deviation from the Wohlfarth relationship starts at about 20 kOe and increases with the magnetic field H. The maximum is in the field range of about 50 kOe, which is close to the maximum coercive field of 57 kOe. After reaching maximum, ΔM decreases quickly to zero at fields of about 65 kOe and takes small negative values at higher fields. Such behaviour is similar to that reported by Mayo et al. [5] and is probably due to strong exchange interaction of grains [2,5].

Viscosity tests provided the viscosity coefficient S from the relation of $M(t) = M_0 + S \ln (t + t_0)$ [4]. The measurements of S showed similar behaviour as found for χ_{irr}, in that S had much lower values and exhibited a much broader curve for the initial magnetisation than for demagnetization [2]. The viscosity parameter Λ increased almost linearly with the field H during the initial magnetisation and was not strongly dependent on the field at the demagnetization curve [2]. The values of Λ of 170-180 Oe in the field range of 50-60 kOe correspond to activation volumes of 0.2-0.4 x 10^{-18} cm^3.

3.2. $Sm_2Fe_{17}N_x$ and α-Fe - $Sm_2Fe_{17}N_x$

For samples consisting of nearly single $Sm_2Fe_{17}N_x$ phase, the coercive force H_c and the remanence M_r had similar curves as plotted in Fig. 1. The rapid increase of H_c and M_r started at fields of about 20 kOe and high fields of $H \geq 60$ kOe were required for saturation [1], while the coercivity was estimated to be 30-40 kOe.

However, in contrast to the measurements in $SmCo_5$ (Fig. 2), no significant difference between χ_{irr} for the initial curve and $\chi_{irr}/2$ for the demagnetization curve was evident (Fig. 4). In samples annealed at 700 °C, the measured demagnetization remanence M_d agreed well with the Wohlfarth relationship. A small deviation (Fig. 5) was found in specimens annealed at lower temperatures (about 650 °C), where the powders had a smaller grain size. The results indicate that the exchange interaction is reduced by increasing grain size, since the proportion of the grain boundary area decreases.

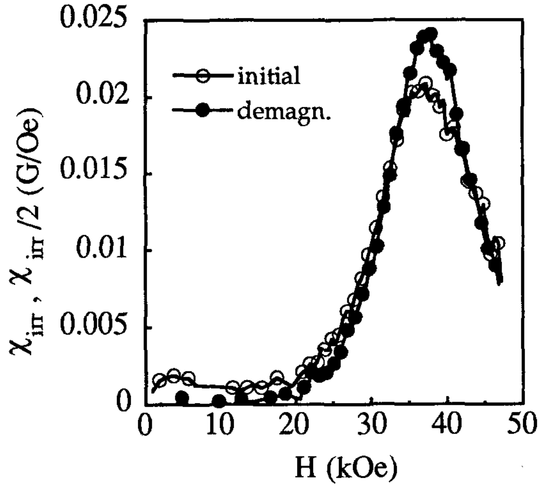

Fig. 4: The irreversible susceptibility χ_{irr} measured at the initial curve and $\chi_{irr}/2$ taken at the demagnetization curve for $Sm_{14}Fe_{86}$-nitride.

For the remanence enhanced α-Fe - $Sm_2Fe_{17}N_x$ materials, the exchange interaction results in a negative deviation of M_d (Fig. 5). The deviation starts at fields of 7-8 kOe, where the irreversible magnetization process in the grains of the hard magnetic $Sm_2Fe_{17}N_x$ phase takes place.

The magnetic viscosity parameter Λ showed nearly a linear relationship with the magnetic field H for samples annealed at higher temperatures of $T_a \geq 700$ °C, when samples consisted of nearly the single $Sm_2Fe_{17}N_x$ phase [1]. An interesting fact is that the curve of Λ for samples annealed at lower temperatures of $T_a \leq 650$ °C is similar to that for the α-Fe - $Sm_2Fe_{17}N_x$ specimens

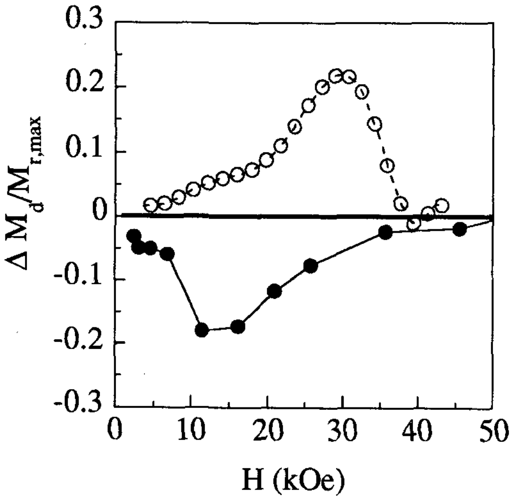

Fig. 5: The normalised deviation of the demagnetization remanence $\Delta M_d/M_{r,max}$ for $Sm_{13}Fe_{87}$-nitrde annealed at 650 °C (o) and Sm_7Fe_{93}-nitrde annealed at 600 °C (•) as function of the magnetic field H.

with remanence enhancement, in that Λ was not significantly dependent on the magnetic field H in the field range of H ≥ 10 kOe (Fig. 6).

The viscosity measurements carried out on the remanence enhanced α-Fe - $Sm_2Fe_{17}N_x$ specimens showed that at low values of H, where rotation and reversal of the soft phase is expected to occur, the viscosity parameter, Λ, was about 20-40 Oe, nearly independent of the volume fraction of α-Fe. In this region a small peak is evident on the χ_{irr} curve in Fig. 4, which is assumed to be associated with the

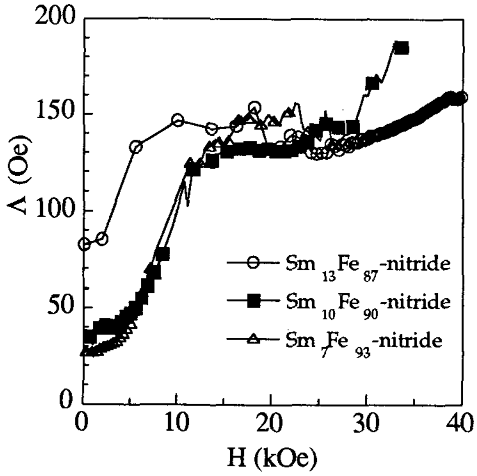

Fig 6: The viscosity parameter Λ as function of the magnetic field H at the demagnetization field H.

irreversible reversal of a small number of α-Fe grains. At higher fields values of Λ of ~130 Oe were measured (Fig. 6), associated with the irreversible magnetisation process of the hard magnetic $Sm_2Fe_{17}N_x$ phase.

References

1. J. Ding, R. Street and P. G. McCormick, 7th Int. Symp. on Magn. Anisotropy and Coercivity in Rare-Earth Trans. Metal Alloys, CANBERRA, July 1992, p.130.
2. J. Ding, R. Street, P. G. McCormick and P. Smith, to be published.
3. J. Ding, R. Street and P. G. McCormick, to be published.
4. Y. Estrin, P. G. McCormick and R. Street, J. Phys.: Cond. Met. 1 (1989) 4845.
5. P. I. Mayo, K. O'Grady, P. E. Kelly, J. Cambridge, I. L. Sanders, T. Yogi and R. W. Chantrell, J. Appl. Phys. 68 (1991) 4733.
6. E. P. Wohlfarth, J. Appl. Phys. 29 (1958) 595.