Magnetostrictions of Sm_2Fe_{17} and $Sm_2Fe_{17}N_3$

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The magnetostrictions of $Sm_2Fe_{17}N_3$ and Sm_2Fe_{17} were investigated using strain gauge rotating-sample methods. In a magnetic field of 8 T, the magnetostriction λ_t ($\lambda_t = \lambda_{//} - \lambda_\perp$) of Sm_2Fe_{17} is about -155×10^{-6} at 300 K. It becomes positive after nitrogenation [$Sm_2Fe_{17}N_3$, λ_t is about $+44 \times 10^{-6}$ at 300 K ($H = 8$ T)] due to the crystal field effect and the exchange striction. It was noticed that inverse magnetostriction effect can be used to increase the magnetic alignment and remanence of the bonded magnets. As a result, transverse die-pressing ($H \perp P$) was very effective in increasing the degree of magnetic alignment of $Sm_2Fe_{17}N_3$ powders, yielding remanence higher than that obtained by axial die-pressing (H//P). By increasing the degree of magnetic alignment of powders, the maximum energy product $(BH)_{max}$ of $Sm_2Fe_{17}N_3$ bonded magnet was improved by 15%.

Index Terms—**Magnetic alignment, magnetostriction, permanent magnet, $Sm_2Fe_{17}N_3$.**

I. Introduction

ONE OF the important developments in the field of hard magnetic materials is the discovery that the introduction of interstitial atoms has a significant effect on the improvement of magnetic properties in some rare earth iron intermetallic compounds [1], [2]. By introducing nitrogen atoms into the Sm_2Fe_{17} lattice, the intrinsic magnetic properties of Sm_2Fe_{17} compound are drastically changed, resulting in an increase in Curie temperature T_c, saturation magnetization M_s, and a change of the magnetocrystalline anisotropy from easy-plane to strong easy-axis, making this compound favorable for permanent magnet applications [1]–[3]. Recently, $Sm_2Fe_{17}N_3$ powders and bonded magnets have been produced [4]–[8], and powders with a high maximum energy product 41 MGOe were reported [9].

The magnetostriction is an important intrinsic parameter in determining magnetic properties of materials. When a stress is applied to a ferromagnetic sample, it may induce a magnetoelastic anisotropy through coupling with the intrinsic magnetostriction constants. This will in turn change parameters of the magnetization curve such as coercive field and magnetic susceptibility. It was found that the magnetostrictions λ_t of the R_2Fe_{17} (R = Y, Pr, Sm, Tb, Dy, Ho, Er, Tm) compounds were small (less than 200×10^{-6}) and negative at room temperature, except that of Pr_2Fe_{17} [10]. The effects of nitrogenation and hydrogenation on the magnetostrictions were studied in R_2Fe_{17} $(N, H)_x$ (R = Y, Pr, Nd, Tb, Dy, Ho, Er) [11], [12]. The results indicated that the nitrogenation could reduce the magnetostriction or change the sign of the magnetostriction. The magnetostriction λ_t changed sign from positive to negative for R = Tb, Dy, Ho (Stevens factor $\alpha_J < 0$) after nitrogenation or hydrogenation, but from negative to positive for R = Er ($\alpha_J > 0$) after hydrogenation; the sign of λ_t cannot be accurately determined for $Er_2Fe_{17}N_x$ because of the small value of it. However, the effect of the nitrogenation on the magnetostriction of $Sm_2Fe_{17}N_3$ compound has not been reported.

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In this paper, we have investigated the magnetostrictions of $Sm_2Fe_{17}N_3$ and its parent alloy with strain gauge rotating-sample methods. The magnetic properties of Sm-Fe-N bonded magnets produced by different pressing methods were also reported. We propose that the improvement of the magnetic alignment of $Sm_2Fe_{17}N_3$ powders with transverse die-pressing (TDP) method is closely associated with the sign of the magnetostriction.

II. Experimental Method

The Sm_2Fe_{17} alloy was produced by strip-casting technique using a wheel speed of 1–5 m/s. The alloy was then annealed in an argon atmosphere at 1050°C for one week. X-ray powder diffraction analyses indicated that the sample was in single phase with a Th_2Zn_{17}-type structure after heat treatment. A Sm_2Fe_{17} plate with a dimension of $10 \times 8 \times 0.5$ mm^3 was selected for the magnetostriction measurement.

The Sm_2Fe_{17} alloy was crushed into powders with the size of about 20–50 μm, nitrogenated at temperatures of 450–500° for 20 h in N_2 atmosphere under 0.1 MPa pressure, and subsequently ball-milled to fine powders. The magnetic properties of the $Sm_2Fe_{17}N_3$ powders are $M_r = 142$ emu/g, ${}_iH_c = 1.21$ T, and $(BH)_{max} = 39$ MGOe. For magnetostriction measurement, the powders of $Sm_2Fe_{17}N_3$ were bonded using epoxy resin with a volume fraction of ~15% to ensure the isotropic characteristic of the bonded sample. The measurement results were modified according to the reported method [13]. The effective magnetostriction ratio λ^* is

$$\lambda^* \cong \frac{f}{1 - 1.25\sigma_2}(1.1538 - 1.2628\sigma_2 - 0.2122 f^{2/3}) \quad (1)$$

$$\lambda = \lambda(f)/\lambda^* \quad (2)$$

where f is the filling volume fraction of magnetostrictive component, σ_2 is the Poisson ratio of nonmagnetostriction medium ($\sigma_2 = 0.4$ for epoxy resin), $\lambda(f)$ is the measured magnetostriction for the composite materials including nonmagnetic medium(epoxy resin), and λ is the magnetostriction for the pure magnetic material. Thus, we have $\lambda^* \cong 0.18$ for $Sm_2Fe_{17}N_3$ sample. As a comparison, a magnetically aligned $Sm_2Fe_{17}N_3$ sample was also prepared with the same amount

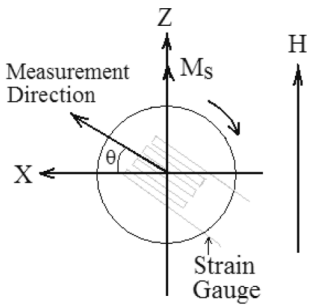

Fig. 1. Illustration of the rotating-sample measurement method.

of epoxy resin in a pulse magnetic field of 4 T. The dimension of bonded sample was $10 \times 8 \times 1$ mm^3.

The magnetostriction measurements were carried out using a Wheatstone bridge. The strain gauges were bonded to the surface of the test samples. To minimize the magnetoresistance and thermal expansion effects on the strain gauge, a compensation strain gauge in the bridge circuit was glued on a copper plate and was placed together with the measured samples. The magnetostrictions as a function of angle were measured using the horizontal sample rotator option of Physical Property Measurement System (PPMS, Quantum Design Inc.), which can turn the sample from 0° to 360° in the magnetic field. The sample plane is always perpendicular to the rotating axis. Three wires from the measurement strain gauge and compensation strain gauge were connected to the strain bridge. A magnetic field of up to 8 T was applied during the measurements.

Fig. 1 illustrates the rotating-sample measurement method. The starting angle of the rotator ($\theta = 0$) corresponds to the $\lambda_\perp$ direction. The strain bridge was tuned to the balance state in a magnetic field of 8 T before rotating measurements, i.e., the reading data on the bridge correspond to $(\lambda(\theta) - \lambda_\perp)$. Here, we refer it as the rotating-sample method. We also measured the magnetostriction $(\lambda_{//} - \lambda_\perp)$ as a function of the applied magnetic field H. In this paper, we refer to this method as the conventional method. The data acquisition and processing were performed by a computer using the IEEE-488 interface bus and the LabVIEW software.

For the isotropic polycrystalline magnetic materials, the value of $\lambda(\theta) = (3/2)\bar{\lambda}(\cos^2\varphi - (1/3))$, which leads to $\bar{\lambda} = 2/3^*(\lambda_{//} - \lambda_\perp)$, where φ is the angle between the magnetization and the measurement direction (note: $\varphi = 90° - \theta$ since the starting angle of the rotator $(\theta = 0)$ corresponds to the $\lambda_\perp$ direction). The $\bar{\lambda}$ is the saturation magnetostriction for polycrystalline materials measured along the magnetic field direction.

In order to study the relationship of magnetostriction and the magnetic alignment of permanent magnets, the bonded $Sm_2Fe_{17}N_3$ magnets with 5 wt% of epoxy resin were produced in a magnetic field of 2.0 T with TDP and axial die-pressing (ADP) methods.

III. Results and Discussion

First, in order to test whether the magnetostriction $[\lambda(\theta) - \lambda_\perp]$ can be accurately measured using the rotating-sample method, the magnetostriction of a polycrystalline Ni plate-like sample was measured as a function of the measured angle θ (see Fig. 2). We obtained that $(\lambda_{//} - \lambda_\perp) = -45 \times 10^{-6}$, $\bar{\lambda} = 2/3^*(\lambda_{//} - \lambda_\perp) = -30 \times 10^{-6}$, which are in a good agreement with the standard value of Ni($\bar{\lambda} = -33 \times 10^{-6}$). Thus, we believe that the rotating-sample method is accurate enough to investigate the magnetostriction of magnetic materials with a similar magnitude of magnetostriction.

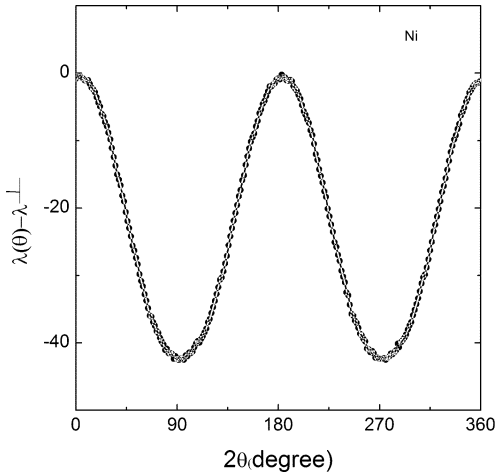

Fig. 2. Magnetostriction $[\lambda(\theta) - \lambda_\perp]$ of Ni at 300 K as a function of measurement angle θ (H = 2 kOe).

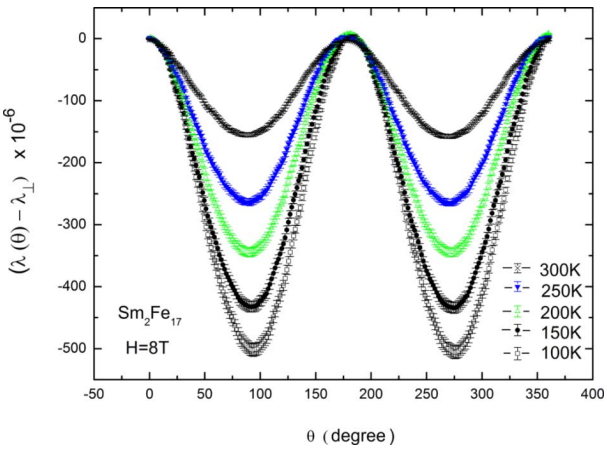

Fig. 3. Magnetostrictions $(\lambda(\theta) - \lambda_\perp)$ of Sm_2Fe_{17} as a function of the angle θ in a field of 8 T at different temperatures.

The magnetostriction $(\lambda(\theta) - \lambda_\perp)$ of Sm_2Fe_{17} as a function of measurement angle θ was measured using the rotating-sample method in a magnetic field of 8 T at different temperatures. The $(\lambda(\theta = 90°) - \lambda_\perp)$ corresponds to $(\lambda_{//} - \lambda_\perp)$. As shown in Fig. 3, the $(\lambda_{//} - \lambda_\perp)$ of Sm_2Fe_{17} are -500, -430, -344, -264, and -155×10^{-6} at 100 K, 150 K, 200 K, 250 K, and 300 K, respectively. The absolute values of the magnetostrictions decrease with the increasing temperature.

Fig. 4 shows the $(\lambda(\theta) - \lambda_\perp)$ versus θ curves of Sm_2Fe_{17} at 300 K in different magnetic fields (H = 3 T, 5 T, 8 T). No saturation was observed in a field of up to 8 T. It was found that the magnetostrictions show strong dependence on the applied magnetic field. For example, the value of $(\lambda_{//} - \lambda_\perp)$ is about -120 ppm at 300 K with a magnetic field of 3 T, which corresponds to a value of $\bar{\lambda} = -85$ ppm. The value $(\lambda_{//} - \lambda_\perp)$ at 3 T is closed to that of Sm_2Fe_{17} (-98 ppm) measured at a magnetic field of 2.5 T (see [10]). We have also measured the magnetostriction $(\lambda_{//} - \lambda_\perp)$ as a function of applied field H using the

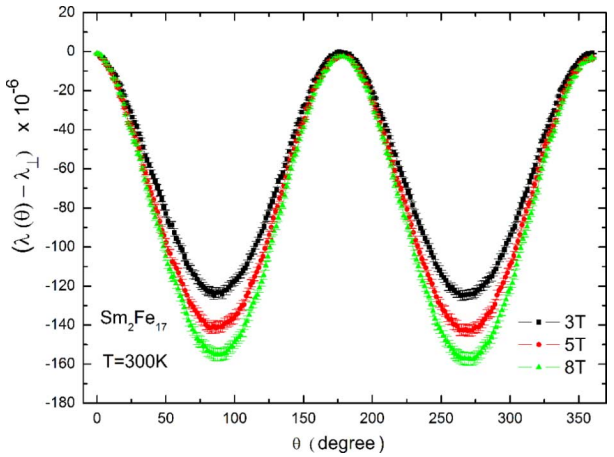

Fig. 4. Magnetostrictions $(\lambda(\theta)-\lambda_\perp)$ of Sm_2Fe_{17} as a function of the angle θ (H = 3, 5 and 8 T) at 300 K.

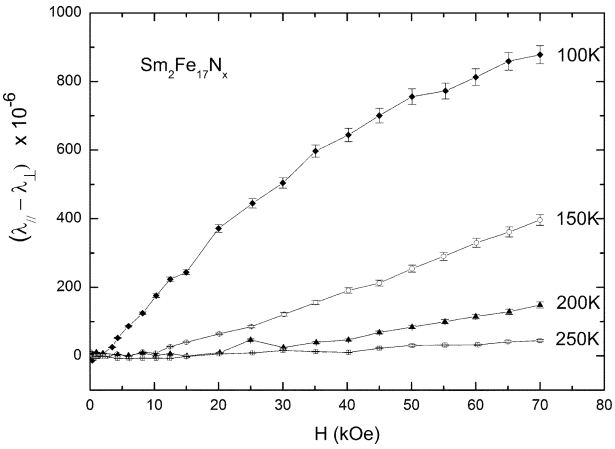

Fig. 5. Magnetostriction $(\lambda_{//}-\lambda_\perp)$ of $Sm_2Fe_{17}N_3$ as a function of applied magnetic field H at different temperatures.

conventional method (results not shown here). The results are in good agreement with those measured by the rotating-sample method. These aspects will be discussed later in the case of $Sm_2Fe_{17}N_3$ that exhibits a small value of magnetostriction.

The magnetostriction of Sm_2Fe_{17} can be assumed to result from both rare-earth Sm and Fe sublattices. In order to estimate the contribution of Sm sublattice to the total magnetostriction, we compared the magnetostriction of Sm_2Fe_{17} to that of Y_2Fe_{17} ($(\lambda_{//}-\lambda_\perp)=-126\times10^{-6}$ at room temperature) [11], where the magnetostriction should mainly arise from the Fe sublattice. It is clear that the magnetostriction of Sm_2Fe_{17} at room temperature mainly results from the Fe sublattice, and only a small portion comes from Sm sublattice due to the weak exchange interactions among rare-earth ions at the temperatures close to the Curie temperature ($T_c = 395$ K for Sm_2Fe_{17}). As the temperature decreases, the $(\lambda_{//}-\lambda_\perp)$ of Sm_2Fe_{17} reaches -500×10^{-6} at 100 K (the $(\lambda_{//}-\lambda_\perp)$ of Y_2Fe_{17} is -31×10^{-6} at 80 K [11]). This indicates that the contribution from Sm sublattice has a negative sign and the absolute value of magnetostriction increases dramatically when the temperature decreases.

The curves of the magnetostrictions $(\lambda_{//}-\lambda_\perp)$ versus applied field H of $Sm_2Fe_{17}N_3$ at various temperatures were measured using the conventional method (Fig. 5). The magnetostrictions $(\lambda_{//}-\lambda_\perp)$ of $Sm_2Fe_{17}N_3$ in a magnetic

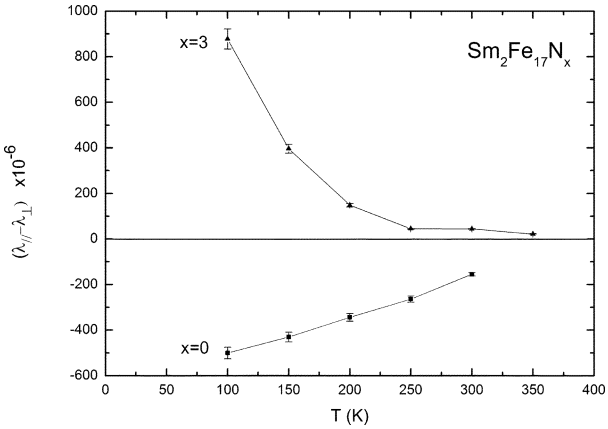

Fig. 6. Magnetostrictions $(\lambda_{//}-\lambda_\perp)$ of $Sm_2Fe_{17}N_x$ (x = 0, 3) at different temperatures.

field of 7 T are $+878\times10^{-6}$, $+396\times10^{-6}$, and $+148\times10^{-6}$ at 100 K, 150 K, and 200 K, respectively. Above 200 K, the magnetostriction of $Sm_2Fe_{17}N_3$ is too small to be measured accurately, and the sign of the magnetostriction becomes uncertain. The difficulty of completely demagnetizing the sample is especially true for permanent magnetic materials, causing large uncertainty in the measurements of these materials with small values of magnetostriction.

The rotating-sample method is more convenient for the measurements of permanent magnetic materials. This is because the measurements are performed in a constant magnetic field and temperature, thus the demagnetization is not required. The $(\lambda(\theta)-\lambda_\perp)$ versus θ curves of $Sm_2Fe_{17}N_3$ at 300 K and 350 K are measured by the rotating-sample method. The obtained $(\lambda_{//}-\lambda_\perp)$ of $Sm_2Fe_{17}N_3$ are $+44\times10^{-6}$ at 300 K and $+22\times10^{-6}$ at 350 K (H = 8 T). Since the anisotropy field μ_0H_a of $Sm_2Fe_{17}N_x$ (between 11 and 26 T at 300 K [2]) is much higher than the applied field of 8 T, the magnetostriction of $Sm_2Fe_{17}N_3$ could not be saturated in our measurements. Furthermore, the anisotropy field of $Sm_2Fe_{17}N_x$ increases with the decreasing temperature [2], thus no cosine function is displayed from the curve of $(\lambda(\theta)-\lambda_\perp)$ versus θ at low temperature. It is noted that the signal-to-noise ratio (5:1) is high for these measurements due to the small values of the magnetostrictions (less than 10^{-5} before modification).

As a comparison, a magnetically aligned $Sm_2Fe_{17}N_3$ sample was also prepared and measured. Compared to the results obtained from unaligned samples, the data from the aligned one cannot be modified with the formula (1, 2) due to the fact that the formula is based on the assumption of the isotropic property of samples. However, the peak–peak value is close to the $\bar{\lambda}$ measured on the isotropic sample, which confirms that the magnetostriction of $Sm_2Fe_{17}N_3$ is positive and the saturation value should be higher.

To understand the effect of nitrogenation on the magnetostriction, the $(\lambda_{//}-\lambda_\perp)$ of the $Sm_2Fe_{17}N_x$ (x = 0, 3) as a function of temperature was plotted (Fig. 6). Different from the compound without nitrogenation (Sm_2Fe_{17}), it has a negative sign; the compound with nitrogenation ($Sm_2Fe_{17}N_3$) has positive contribution. It has been found that the nitrogen atom partially occupies the interstitial 9e sites of Th_2Zn_{17}-type rhombohedral structure (e.g., $Sm_2Fe_{17}N_3$) [14]. Specifically, each Sm^{+3} is surrounded by six iron atoms in the basal plane

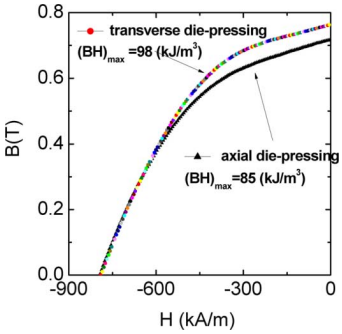

Fig. 7. Demagnetization curves of Sm-Fe-N bonded magnets produced with different aligning processes (the measurement direction is along the magnetic aligned direction).

before nitrogenation. After nitrogenation, three interstitial nitrogen atoms are added in the basal plane around each Sm^{+3} ion. These electronegative interstitial nitrogen atoms not only lead to a lattice expansion, but also a modification of the ambient electric field gradient. For example, the principal component of electric field gradient V_{zz} at the nucleus of Sm changes from 10.2 to 33.9 (10^{21} V/m^3) after nitrogenation [2]. As a result, nitrogenation changes the crystal field and the exchange striction of Sm_2Fe_{17} and subsequently changes the magnetostriction of $Sm_2Fe_{17}N_x$ from negative $(x = 0)$ to positive $(x = 3)$.

Taking the inverse magnetostriction effect or the magnetomechanical effect into consideration, the magnetoelastic energy of a polycrystalline material is given by the following equation [15]:

$$E_{me} = -\frac{3}{2}\lambda_s \sigma \cos^2 \varphi \tag{3}$$

where σ is the applied stress, λ_s is the magnetostriction, and φ is the angle between "easy" direction of the magnetization and the stress σ direction. For the bonded anisotropic magnets, the compressive stress σ is negative $(\sigma < 0)$ during the magnetic alignment process. Therefore, if $\lambda < 0$, the energy will be minimal when the magnetization (or applied magnetic field) is in parallel to the stress direction axis (i.e., $\varphi = 0$); if $\lambda > 0$, the energy will be minimal when the magnetization (or applied magnetic field) is in perpendicular to the stress direction axis (i.e., $\varphi = 90°$). In other words, a compressive stress σ will help to align the easy axis of single domain or persu-single domain particles along the direction of the applied magnetic field H, where $H//\sigma$ for $\lambda < 0$ materials, or $H \perp \sigma$ for $\lambda > 0$ materials.

To test this hypothesis, $Sm_2Fe_{17}N_3$ bonded magnets were produced with different pressing processes, i.e., the TDP and ADP methods. Fig. 7 is the demagnetization curves of $Sm_2Fe_{17}N_3$ bonded magnets that underwent different pressing processes (the measurement is along the magnetic aligned direction). It is found that the remanence B_r of TDP sample increases by ~0.460 T, and the $(BH)_{max}$ increases by 13 kJ/m^3 (1.57 MGOe) (about 14.6%) as compared to that of the ADP sample. This is due to the significant improvement in the alignment of magnetic powders. This effect also can be used to explain the result of Saito *et al.* [16], where a compression shearing method can lead to the enhancement of remanence for the Sm-Fe-N magnets due to applied shear stress. We have observed a similar effect in the $Nd_2Fe_{14}B$ and Sr-ferrites bonded magnets.

IV. Conclusion

The magnetostriction of Sm_2Fe_{17} is negative. Upon nitrogenation, $Sm_2Fe_{17}N_x$ shows a positive magnetostriction due to the change of the crystal field and exchange magnetic striction. It is found that TDP can improve the magnetic alignment for the $\lambda > 0$ materials, whereas ADP can improve the alignment for the $\lambda < 0$ materials. Thus, based on the positive magnetosriction of $Sm_2Fe_{17}N_3$ magnets, significant improvements in the magnetic alignment and maximum energy product $(BH)_{max}$ have been made using TDP method.

Acknowledgment

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