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Dynamic magnetoelastic properties of epoxy-bonded Sm$_{0.88}$Nd$_{0.12}$Fe$_{1.93}$ pseudo-1–3 negative magnetostrictive particulate composite

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Pseudo-1–3 negative magnetostrictive particulate composite is prepared by embedding and aligning light rare earth (Sm and Nd)-based negative magnetostrictive Sm$_{0.88}$Nd$_{0.12}$Fe$_{1.93}$ particles with randomly distributed sizes of 10–180 μm in a passive epoxy matrix using a particle volume fraction of 0.5. The dynamic magnetoelastic properties of the composite are investigated as a function of both magnetic bias field and frequency under a constant magnetic drive field. The dynamic relative permeability ($\mu_{r33}$) exhibits a flat frequency response with no observable dispersion at all bias field levels, except for the fundamental shape resonance range of 40–50 kHz. The free ($\mu_{r33}^0$) and clamped ($\mu_{r33}^c$) relative permeabilities attain their maximum values at low bias field levels of $\leq$10 kA/m because of the relatively easy 180° domain-wall motion. The elastic modulus at constant magnetic field strength ($E_D^0$) and at constant magnetic flux density ($E_D^0$) show a maximum negative-ΔE effect, accompanying a maximum dynamic strain coefficient ($d_{33}$) of $\sim$2 nm/A, at about 100 kA/m due to the maximum motion of non-180° domain walls. © 2012 American Institute of Physics. [doi:10.1063/1.3679045]

Pseudobinary RR’Fe$_2$ Laves alloys (R, R’ ≡ rare earths, where R ≠ R’) have received considerable research interest over the past decade due to their potentially high magnetoelastic properties and technologically important applications in sonar and actuation devices.⁴ Among various kinds of RR’Fe$_2$ Laves alloys available today, Tb$_0.3$Dy$_0.7$Fe$_{1.92}$ (Terfenol-D) and Sm$_{0.88}$Dy$_{0.12}$Fe$_{1.93}$ (Samfenol-D) have been recognized as key positive and negative magnetostrictive alloys, respectively, because of their giant positive and negative magnetostrictions with low magnetocrystalline anisotropy, and material cost can be improved.⁴–⁹

While Terfenol-D, Samfenol-D, and their composites are useful, the inclusion of certain amounts of scanty and expensive heavy rare earths Tb and Dy in these alloys has called for alternatives which are constituted by relatively abundant and cost-effective light rare earths in more recent years. On the side of the positive magnetostrictive alloys, several good light rare earth-substituted candidates have been synthesized by substituting light rare earth Pr or Nd for Tb and/or Dy in the Terfenol-D composition.¹⁰–¹² On the side of the negative magnetostrictive alloys, few reports have been made even though Sm$_{0.88}$Nd$_{0.12}$Fe$_{1.93}$ alloy has showed promise at room temperature.¹³

We have succeeded in synthesizing Sm$_{1-x}$Nd$_x$Fe$_{1.55}$ (0 ≤ x ≤ 0.56) Laves alloys, preparing epoxy-bonded Sm$_{1-x}$Nd$_x$Fe$_{1.55}$ (0 ≤ x ≤ 0.56) pseudo-1–3 composites, and reporting their quasi-static magnetoelastic properties.¹⁴,¹⁵ In this paper, we further prepare epoxy-bonded Sm$_{0.88}$Nd$_{0.12}$Fe$_{1.93}$ pseudo-1–3 composite with 0.5 particle volume fraction and study its dynamic magnetoelastic properties as a function of both magnetic bias field and frequency under a constant magnetic drive field in order to obtain an improved physical insight into such an emerging magnetostrictive material system for enabling device applications.

Polycrystalline Sm$_{0.88}$Nd$_{0.12}$Fe$_{1.93}$ alloy was synthesized by an arc-melting process in a high-purity argon atmosphere.¹⁴ The constituent metals used in the arc-melting process were 99.9% pure Sm, 99.9% pure Nd, and 99.8% pure Fe. An extra 3 wt. % Sm was added to compensate the volatilization of Sm for its high vapor pressure during the arc-melting process. The ingot was sealed in a quartz tube for homogenization at 600 °C for 7 days in a high-purity argon atmosphere. The homogenized ingot was ground into particles with randomly distributed sizes of 10–180 μm. X-ray diffractionometry with CuKα radiation was applied to analyze the phase structure of the particles at room temperature in a Rigaku D/max-2500pc diffractometer equipped with a graphite monochromator. The x-ray diffraction results confirmed the presence of a single cubic Laves phase in the as-prepared alloy.

To fabricate an epoxy-bonded Sm$_{0.88}$Nd$_{0.12}$Fe$_{1.93}$ pseudo-1–3 composite, Sm$_{0.88}$Nd$_{0.12}$Fe$_{1.93}$ particles and Araldite LY5210/HY2954 epoxy were homogeneously mixed in a paper cup using a particle volume fraction of 0.5. The mixed...
slurry was transferred into a bronze mold with a rectangular cavity of 6 × 6 mm² cross-section and 20 mm length and degassed under vacuum for 30 min to eliminate air bubbles. The mold was sealed and placed between a pair of Nd–Fe–B permanent magnets to experience a uniform magnetic field of about 180 kA/m along the length direction of the mold. This caused the enclosed particles to align with the magnetic flux lines, producing particulate chains similar to pseudo-fiber composites or, in general, a pseudo-1–3 particulate composite. With the particles being lengthwise-aligned in the mold, the entire mold-magnet assembly was placed in a temperature-controlled chamber at 80 °C for 10 h to ensure a proper cure of the epoxy for achieving a certainly high degree of cross-linking as well as to impart an average axial residual compressive stress of about 3 MPa to the particulate chains through the thermal shrinkage of the epoxy in the composite. This built-in residual compressive stress was shown to be effective in creating a preferred non-180° domain state in the as-prepared composite although no external stress was used. The effect is similar to the case of applying an external prestress to assert an initial non-180° domain state in monolithic Terfenol-D. After demolding, the particle volume fraction of the composite was confirmed to be 0.5 based on Archimedes’ principle and rule-of-mixture formulation for density.

An in-house automated magnetostrictive measurement system was used to measure the dynamic magnetoelastic properties of the composite in the length direction at room temperature and with zero external stress bias. The composite to be measured was placed in the middle of a pair of Helmholtz coils, and the whole sample-coil assembly was situated in the pole gap of a C-shaped, water-cooled electromagnet (Myltem coils, and the whole sample-coil assembly was situated in the soloidal H meter (F.W. Bell 5080), respectively. By sweeping a sinusoidal H₃ of amplitude 80 A/m over a prescribed frequency (f) range of 1–70 kHz at a rate of 26 step/s, the corresponding magnetic flux density (B₃) and dynamic strain (S₃) at discrete frequency intervals of 25 Hz/step were measured under various H_Bias of 10–240 kA/m. H₁ and H_Bias were monitored in situ by a pick-up coil and a Hall probe connected to a Gaussmeter (F.W. Bell 5080), respectively. B₃ and S₃ were acquired using a search coil wrapped around the composite and a strain gauge attached to the center of the composite, respectively. All qualities were gathered using a data acquisition unit (Nation Instruments BNC-2110 and NI-PCI6132) under the control of a computer with a Labview program. The f dependence of dynamic relative permeability (μ_r33) was determined, together with the H_Bias dependence of elastic modules at constant magnetic field strength (E'₃), elastic module at constant magnetic flux density (E''₃), and dynamic strain coefficient (d₃3). Details can be found elsewhere.  

Figure 1 shows the f dependence of μ_r33 of the Sm₀.88Nd₀.12Fe₁.93 composite at various H_Bias. It is seen that μ_r33 is essentially flat over the measured f range of 1–70 kHz with no observable frequency dispersion effect for all H_Bias levels of 10–240 kA/m, apart from the fundamental longitudinal-mode resonance-induced variations at about 45 kHz. The observation indicates that the eddy-current losses are insignificant in the composite for f up to 70 kHz. This suggests potential applications of the composite at high frequencies.

Figure 2 plots the dependence of μ_r33 and μ_l33 on H_Bias for the Sm₀.88Nd₀.12Fe₁.93 composite. μ_r33 and μ_l33 denote the relative permeability at constant stress (i.e., free relative permeability) and that at constant strain (i.e., clamped relative permeability), respectively. The values of μ_r33 and μ_l33 are extracted from the measured μ₃3 spectrum in Fig. 1 with the corresponding μ₃3 values at 1 and 70 kHz, respectively. It is found that the two permeabilities attain their highest values at low H_Bias levels of ≤10 kA/m and their values decrease with an increase in H_Bias. In fact, the permeabilities are primarily determined by domain-wall motion at the beginning of the technical magnetization process. Accordingly, they are maximized at rather low H_Bias levels of ≤10 kA/m due to the relatively easy motion of 180° domain walls. As H_Bias is increased beyond 10 kA/m, the reduced motion of 180° domain walls competes with the increased motion of non-180° domain walls, giving rise to an obvious decrease in their values.

Figure 3 illustrates the variation of E'₃ and E''₃ on H_Bias for the Sm₀.88Nd₀.12Fe₁.93 composite. It is clear that both moduli decrease initially with increasing H_Bias from 10 to 100 kA/m, leading to a negative-ΔE effect with a maximization near H_Bias = 100 kA/m. At elevated H_Bias levels of 100–240 kA/m, both moduli increase with increasing H_Bias. Due to the fact that residual compressive stress has been preset in our
composite through thermal curing of the epoxy phase during composite fabrication, this residual compressive stress essentially exerts on the embedded Sm_{0.88}Nd_{0.12}Fe_{1.93} particles to create a preferred non-180° domain-wall state in the as-prepared composite.\(^5,6,16\) Since non-180° domain-wall motion changes both magnetization (\(M_3\)) and strain (\(S_3\)) while 180° domain-wall motion changes only \(M_3\),\(^1,5,6,17\) the initial decrease in moduli with increasing \(H_{\text{Bias}}\) in Fig. 3 is mainly attributed to non-180° domain-wall motion. As \(H_{\text{Bias}}\) is increased to about 100 kA/m where maximum negative-\(\Delta E\) effect occurs, the compliance associated with the increased deformation contribution from this non-180° domain-wall motion reaches a maximum, resulting in a minimum in stiffness. Above 100 kA/m, the moduli increase with increasing \(H_{\text{Bias}}\) because constraining of non-180° domain-wall motion by \(H_{\text{Bias}}\) tends to stiffen the composite.

Figure 4 shows \(d_{33}\) at 1 kHz as a function of \(H_{\text{Bias}}\) for the Sm_{0.88}Nd_{0.12}Fe_{1.93} composite. It is noted that \(d_{33}\) carries a negative sign, indicating the presence of a negative magnetostrictive effect (i.e., a magnetic field-induced negative strain) in the composite. The value of \(d_{33}\) increases initially up to the maximum of about \(-2\) nm/A near 100 kA/m and then decreases with increasing \(H_{\text{Bias}}\). This is a result of increasing and maximizing \(S_3\) contribution from the non-180° domain-wall motion and then domain saturation with limited \(S_3\) contribution from the motion of the available non-180° domain walls. In particular, the presence of maximum \(d_{33}\) near \(H_{\text{Bias}} = 100\) kA/m is confirmed by the quasistatic magnetostriction–magnetic field (\(\lambda-H\)) curve measured at a low frequency of 0.05 Hz and with zero external stress bias in the inset of Fig. 4. It is obvious that the composite is biased in the center of the “burst region” of the \(\lambda-H\) curve where non-180° domain-wall motion is maximum.\(^1,5,6,17\) The results agree with the minimization of the two moduli and the maximization of negative-\(\Delta E\) near 100 kA/m (Fig. 3). It is noted that the \(d_{33}\) values reported in Fig. 4 are not necessarily the largest available values of our Sm_{0.88}Nd_{0.12}Fe_{1.93} composite because the \(d_{33}\) and \(\lambda\) measurements were done under a mechanically free condition. The use of an additional mechanical stress would preset more initial non-180° domains and thus may produce a larger \(S_3\) output for a given \(H_3\) value, i.e., a larger \(d_{33}\).\(^1,4,16\)

We have fabricated epoxy-bonded Sm_{0.88}Nd_{0.12}Fe_{1.93} pseudo-1–3 negative magnetostrictive composite with 0.5 particle volume fraction and investigated its dynamic magnetoelastic properties as a function of both \(H_{\text{Bias}}\) and \(f\) under an \(H_3\). It has been found that the composite generally possesses insignificant eddy-current losses and its magnetoelastic properties depend greatly on \(H_{\text{Bias}}\). The magnetoelastic properties of the composite at low \(H_{\text{Bias}}\) levels of \(\leq 10\) kA/m are mainly influenced by the relatively easy 180° domain-wall motion. The maximization of negative-\(\Delta E\) effect and \(d_{33}\) near \(H_{\text{Bias}} = 100\) kA/m is a result of maximizing non-180° domain-wall motion. The present study provides an improved physical insight into such an emerging light rare earth-based negative magnetostrictive material system for device applications.

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