Journal of Alloys and Compounds 509 (2011) 4954–4957

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: www.elsevier.com/locate/jallcom

Magnetomechanical properties of epoxy-bonded $Sm_{1-x}Nd_xFe_{1.55}$ ($0 \le x \le 0.56$) pseudo-1–3 magnetostrictive particulate composites

Fang Yang[a,b], Chung Ming Leung[a], Siu Wing Or[a,*], Wei Liu[b], Xiangke Lv[b], Zhidong Zhang[b]

[a] Department of Electrical Engineering, The Hong Kong Polytechnic University, Hung Hom, Kowloon, Hong Kong
[b] Shenyang National Laboratory for Materials Science, Institute of Metal Research and International Centre for Materials Physics, Chinese Academy of Sciences, Shenyang 110016, PR China

ARTICLE INFO

Article history:
Received 23 November 2010
Received in revised form 22 January 2011
Accepted 27 January 2011
Available online 2 February 2011

Keywords:
Light rare earths
Magnetocrystalline anisotropy
Magnetomechanical properties
Negative magnetostriction
Magnetostrictive composites
$Sm_{1-x}Nd_xFe_{1.55}$ alloys

ABSTRACT

Pseudo-1–3 magnetostrictive particulate composites consisting of light rare earth (Sm and Nd)-based magnetostrictive $Sm_{1-x}Nd_xFe_{1.55}$ particles with the Nd content x of 0–0.56 and randomly distributed sizes of 10–180 μm embedded and aligned in a passive epoxy matrix are fabricated using the particulate volume fraction of 0.5. The quasistatic magnetomechanical properties of the composites are investigated and the results are compared with their monolithic alloys for various x. The composites exhibit similar qualitative trends in properties with the alloys for all x. The $Sm_{0.92}Nd_{0.08}Fe_{1.55}$ composite shows a large unsaturated magnetostriction λ of −530 ppm at 500 kA/m and a high piezomagnetic coefficient d_{33} of −2.0 nm/A at 100 kA/m as a result of the magnetocrystalline anisotropy compensation between Sm^{3+} and Nd^{3+} ions in the $Sm_{0.92}Nd_{0.08}Fe_{1.55}$ alloy.

1. Introduction

Pseudobinary $RR'Fe_2$ Laves alloys (R, R′≡rare earths, where R ≠ R′) have received much attention in recent years for magnetostrictive transducer and actuator applications because of their potentially high magnetostrictions and low magnetocrystalline anisotropy at room temperature [1,2]. To reduce eddy-current losses and mechanical brittleness intrinsic in the alloys, special attention has been paid to develop epoxy-bonded magnetostrictive particulate composites [3–8]. In fact, composites with pseudo-1–3 configuration (i.e., aligning magnetostrictive particles in a passive polymer matrix) generally demonstrate higher magnetomechanical properties than those with 0–3 configuration (i.e., dispersing particles in the matrix) due to their anisotropic nature [6,7]. Moreover, there exists an optimal particulate volume fraction of 0.5 in the pseudo-1–3 composites by taking the tradeoff between the maximization of magnetomechanical properties and the minimization of material cost into consideration [8].

Among the available $RR'Fe_2$ Laves alloys, $Tb_{0.3}Dy_{0.7}Fe_{1.92}$ (Terfenol-D) and $Sm_{0.88}Dy_{0.12}Fe_{1.93}$ (Samfenol-D) have been widely investigated and shown to possess giant positive and negative magnetostrictions, respectively [9,10]. Since the main functional components of Terfenol-D and Samfenol-D involve scanty and expensive heavy rare earths Tb and Dy, it will be a great benefit to applications if these heavy rare earths in the ordinary Terfenol-D and Samfenol-D compositions can be substituted by abundant and cost-effective light rare earths and maintain competitive properties. To date, literature reports have been mainly on light rare earth-substituted positive magnetostrictive alloy systems. Accordingly, several good candidates have been prepared by substituting Pr or Nd for Tb and/or Dy in Terfenol-D. These include: $(Tb_{0.3}Dy_{0.7})_{1-x}Pr_xFe_{1.55}$ ($0 \le x \le 0.4$) [11], $Tb_{0.2}Dy_{0.8-x}Pr_x(Fe_{0.9}B_{0.1})_{1.93}$ ($0 \le x \le 0.4$) [12], $Tb_xNd_{1-x}Fe_{1.9}$ ($0 \le x \le 0.8$) [13], etc. For the equally important negative magnetostrictive alloy systems, reports are relatively limited.

Recently, we have synthesized light rare earth-based $Sm_{1-x}Nd_xFe_{1.55}$ ($0 \le x \le 0.56$) Laves alloys showing high negative magnetostrictions [14]. Nd is considered as a good candidate for partial substitution in stable $SmFe_2$ because $NdFe_2$ has opposite anisotropy and a small positive magnetostriction [15–17]. In this work, we aim to extend our work on $Sm_{1-x}Nd_xFe_{1.55}$ ($0 \le x \le 0.56$) Laves alloys to fabricate epoxy-bonded $Sm_{1-x}Nd_xFe_{1.55}$ ($0 \le x \le 0.56$) pseudo-1–3 magnetostrictive particulate composites and investigate their quasistatic magnetomechanical properties at a frequency of 0.05 Hz. Comparison of the measured properties between the composites and the alloys is made on the same function of Nd content x in order to obtain a relatively complete picture on the $Sm_{1-x}Nd_xFe_{1.55}$ ($0 \le x \le 0.56$) system.

* Corresponding author. Tel.: +852 34003345; fax: +852 23301544.
E-mail address: eeswor@polyu.edu.hk (S.W. Or).

0925-8388/$ – see front matter © 2011 Elsevier B.V. All rights reserved.
doi:10.1016/j.jallcom.2011.01.138

2. Experimental details

Polycrystalline $Sm_{1-x}Nd_xFe_{1.55}$ ($0 \le x \le 0.56$) alloys with 10 different Nd contents x of 0, 0.08, 0.12, 0.16, 0.20, 0.24, 0.32, 0.40, 0.48, and 0.56 were prepared by arc-melting the constituent metals in a high-purity argon atmosphere [14]. The constituent metals were commercially acquired and their purities were 99.9% for Sm and Nd and 99.8% for Fe. As the vapor pressure of Sm was rather high during arc-melting, an extra 3 wt% Sm was adopted to compensate the volatilization of Sm during the arc-melting process. The ingots were sealed in a quartz tube and homogenized at 873 K for 7 days in a high-purity argon atmosphere. The homogenized ingots were ground into particles with randomly distributed sizes. Two different sieves were used to eliminate any particles smaller than 10 μm and larger than 180 μm, giving a preferred particulate size range of 10–180 μm. X-ray diffractrometry with CuKα radiation was applied to the homogenized particles at room temperature in a Riguku D/max-2500pc diffractrometer equipped with a graphite monochromator. The X-ray diffraction (XRD) analysis indicated that the alloys with the Nd content x varying from 0 to 0.48 contain mainly cubic Laves phase, with minor rare-earth phases. For the alloys having an elevated x of 0.56, the cubic Laves phase coexisted with a small amount of (Sm, Nd)Fe_3 because $NdFe_2$ can only be formed at high pressures. In fact, it was reported that the lattice parameter a increases with increasing x, while the Curie temperature T_c trends to the opposite way [14].

To fabricate the epoxy-bonded $Sm_{1-x}Nd_xFe_{1.55}$ ($0 \le x \le 0.56$) pseudo-1–3 composites, predetermined quantities of $Sm_{1-x}Nd_xFe_{1.55}$ ($0 \le x \le 0.56$) particles ground from the homogenized ingots were homogeneously mixed with Araldite LY5210/HY2954 epoxy in plastic molds with a rectangular cavity of dimensions 7 mm × 7 mm × 28 mm. The mixed slurries were degassed under vacuum for 30 min to eliminate air bubbles. The molds were then sealed to prevent particles from migrating out once they were placed between a pair of Nd–Fe–B permanent magnets. These magnets produced a uniform dc magnetic field of about 150 kA/m along the longitudinal direction of the molds, causing the enclosed particles to align with the magnetic flux lines and producing particulate chains similar to pseudo-fiber composites or, in general, pseudo-1–3 particulate composites [4,8]. After the particles had been lengthwise-aligned in the molds, the entire mold-magnet assemblies were placed in a temperature-controlled oven at 80 °C for 10 h to ensure full cure of the epoxy and 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 composites [18]. This built-in residual compressive stress was shown to be effective in creating a preferred non-180° domain state in the as-prepared composites even though no external stress was used [4,5]. The effect is similar to the case of applying an external prestress to assert an initial non-180° domain state in monolithic Terfenol-D [19]. After being demolded, the particulate ($Sm_{1-x}Nd_xFe_{1.55}$) volume fraction of the composites was determined to be 0.5 based on Archimedes' principle and rule-of-mixture formulation for density expressed below:

$$\rho_c = v_f\rho_a + (1 - v_f)\rho_e, \quad (1)$$

where ρ_c, ρ_a, and ρ_e are the densities of the composite, alloy (=8300 kg/m^3), and epoxy (1097 kg/m^3), respectively; and v_f is the particulate volume fraction of the composites.

An in-house automated magnetostrictive measurement system was used to measure the quasistatic magnetomechanical properties of the composites and alloys in the longitudinal direction at room temperature and with zero stress bias [4]. These included the magnetization–magnetic field (M–H) curves and the magnetostriction–magnetic field (λ–H) curves. The M–H and λ–H curves were acquired by energizing a water-cooled electromagnet to provide a cyclic magnetic field (H) with a maximum amplitude of 500 kA/m at a quasistatic frequency of 0.05 Hz and then measuring the corresponding magnetic flux density (B) and magnetostriction (λ) using a search coil wrapped around the samples and a strain gauge attached to the center of the samples, respectively. It is noted in the measurement that the longitudinal direction of the samples was aligned parallel to the direction of H so that the measured B and λ were essentially the longitudinal values. The magnetization (M) was calculated using

$$M = \frac{B}{\mu_0} - H, \quad (2)$$

where $\mu_0 = 4\pi \times 10^{-7}$ H/m is the permeability of free space. The piezomagnetic coefficient (d_{33}) was obtained from the λ–H plots using

$$d_{33} = \frac{\partial\lambda}{\partial H} \quad (3)$$

3. Results and discussion

Fig. 1(a) shows the magnetization–magnetic field ($4\pi M$–H) curve of the epoxy-bonded $Sm_{0.6}Nd_{0.4}Fe_{1.55}$ composite, while Fig. 1(b) plots the maximum magnetization ($4\pi M_{max}$) of the $Sm_{1-x}Nd_xFe_{1.55}$ ($0 \le x \le 0.56$) composites and alloys as a function of the Nd content x. It is noted from Fig. 1(a) that the saturation magnetization state is not obtained at the maximum available H of 500 kA/m. Hence, the value of $4\pi M_{max}$ appeared in Fig. 1(b) corresponds to that of $4\pi M$ at the maximum available H of 500 kA/m in Fig. 1(a). Moreover, since $4\pi M_{max}$ of the composites increases slowly with increasing x in Fig. 1(b), only the $4\pi M$–H curve of the $Sm_{0.6}Nd_{0.4}Fe_{1.55}$ composite is included in Fig. 1(a). It is clear that the $Sm_{0.6}Nd_{0.4}Fe_{1.55}$ composite, which is similar to other composites with different x (not shown), exhibits soft magnetic characteristics. For comparison, $4\pi M_{max}$ of the alloys demonstrates a similar quantitative increasing trend with the composites for all x. As the composites with different x have the same particulate volume fraction of 0.5, we believe that the increasing trend observed in the composites is mainly due to the intrinsic magnetic properties of the alloys governed by the larger 4f moment of Nd^{3+} ion compared to the Sm^{3+} ion [17].

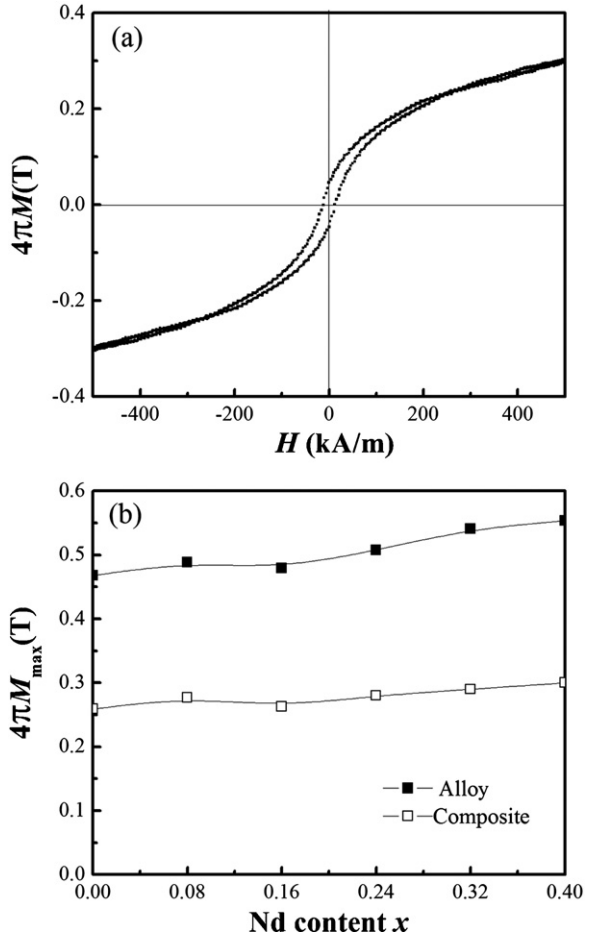

Fig. 1. (a) Magnetization–magnetic field ($4\pi M$–H) curve of the epoxy-bonded $Sm_{0.6}Nd_{0.4}Fe_{1.55}$ composite. (b) Maximum magnetization ($4\pi M_{max}$) of the $Sm_{1-x}Nd_xFe_{1.55}$ ($0 \le x \le 0.56$) composites and alloys as a function of the Nd content x.

The magnetostriction–magnetic field (λ–H) curves of the epoxy-bonded $Sm_{1-x}Nd_xFe_{1.55}$ ($0 \le x \le 0.56$) composites are shown in Fig. 2(a), while the dependence of the Nd content x on maximum magnetostriction (λ_{max}) (i.e., the value of λ at H = 500 kA/m) for the composites and the alloys is plotted in Fig. 2(b). From Fig. 2(a), the saturation magnetostriction state is not achieved at the maximally applied H of 500 kA/m. This reflects the existence of relatively high magnetocrystalline anisotropy in the $Sm_{1-x}Nd_xFe_{1.55}$ ($0 \le x \le 0.56$) alloys and their composites. From Fig. 2(b), λ_{max} increases initially, reaches its maximum at x = 0.08, and then decreases, with increasing x. This indicates that the small substitution of Nd for Sm in $SmFe_2$ can lead to a reduction in the magnetocrystalline anisotropy of $SmFe_2$ and hence an enhancement in λ_{max} [14]. However, the

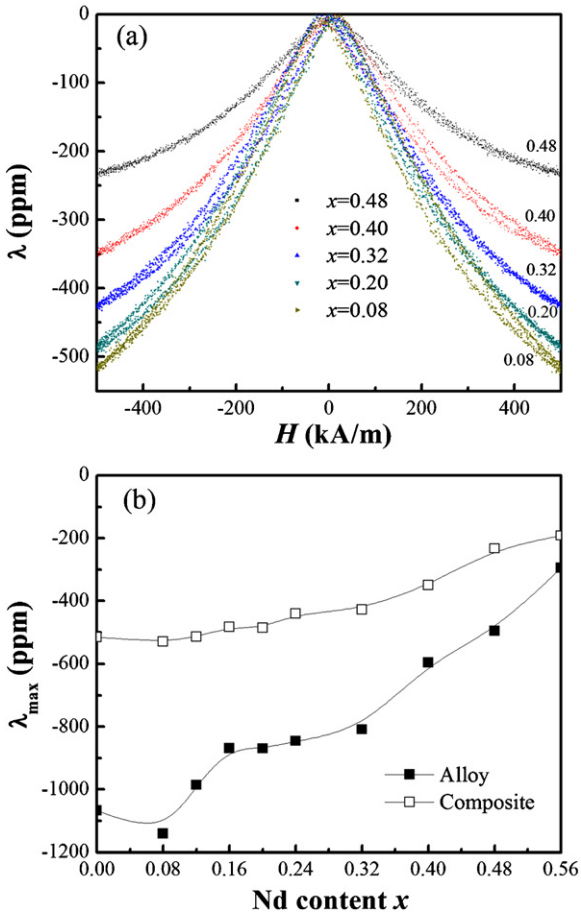

Fig. 2. (a) Magnetostriction–magnetic field (λ–H) curves of the epoxy-bonded $Sm_{1-x}Nd_xFe_{1.55}$ ($0 \le x \le 0.56$) composites. (b) Dependence of the Nd content x on maximum magnetostriction (λ_{max}) for the composites and the alloys.

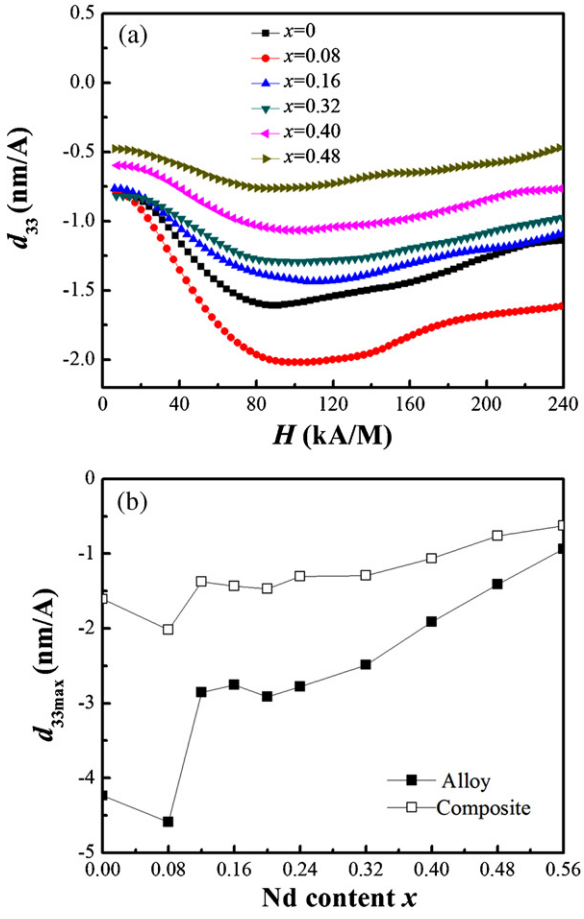

Fig. 3. (a) Piezomagnetic coefficient–magnetic field (d_{33}–H) curves of the epoxy-bonded $Sm_{1-x}Nd_xFe_{1.55}$ ($0 \le x \le 0.56$) composites. (b) Dependence of the Nd content x on maximum piezomagnetic coefficient (d_{33max}) for the composites and the alloys.

enhancement of λ_{max} in the composites is not as obvious as that in the alloys. Among the various composites, the $Sm_{0.92}Nd_{0.08}Fe_{1.55}$ one exhibits the largest λ_{max} of −530 ppm because of the compensation of magnetocrystalline anisotropy between Sm^{3+} and Nd^{3+} ions. Due to the fact that the $SmFe_2$ alloy carries a negative magnetostriction sign in opposition to the positive magnetostriction sign carried by the $NdFe_2$ alloy, there is a general reduction in λ_{max} for x in excess of 0.08 [14]. In general, the composites show a similar quantitative trend in λ_{max}, but with smaller values than the alloys for all x. This is a result of the reduced particulate volume fraction to 0.5 in the composites compared to unity in the alloys.

Fig. 3 shows the piezomagnetic coefficient–magnetic field (d_{33}–H) curves of the epoxy-bonded $Sm_{1-x}Nd_xFe_{1.55}$ ($0 \le x \le 0.56$) composites and the dependence of the Nd content x on maximum piezomagnetic coefficient (d_{33max}) for the composites and the alloys. The d_{33}–H curves in Fig. 3(a) for the composites display similar quantitative increasing and decreasing trends, with the maximum d_{33} of about −2 nm/A at 100 kA/m for the $Sm_{0.92}Nd_{0.08}Fe_{1.55}$ composite. The initial increase in d_{33} with the increase in H can be attributed to the increase in non-180° domain-wall motion. The H at which d_{33} is maximized indicates the actuation of the composites in the "burst region" of the λ–H curves (Fig. 2(a)) where the non-180° domain-wall motion is maximum. Above this critical H, the domain saturation occurs, resulting in the decrease in d_{33} value. As shown in Fig. 3(b), the composites show a similar quantitative trend in d_{33max}, but also with smaller values than the alloys for all x owing to the reduced particulate volume fraction to 0.5.

To further understand the magnetization process of $Sm_{0.92}Nd_{0.08}Fe_{1.55}$ composite and alloy, the dependence of λ/λ_{max} on M/M_{max} is shown in Fig. 4. The values of λ_{max} and M_{max} are obtained at H = 500 kA/m from Fig. 2(b) and Fig. 1(b), respectively. It is observed that the values of λ/λ_{max} of the composite are generally larger than those of the alloy. Since the 180° domain-wall motion results in changes in M without accompanying λ,

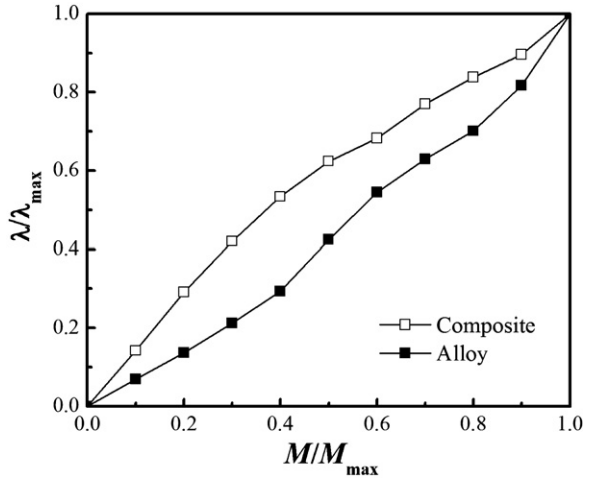

Fig. 4. Dependence of λ/λ_{max} on M/M_{max} for the $Sm_{0.92}Nd_{0.08}Fe_{1.55}$ composite and alloy.

while the non-180° domain-wall motion leads to changes in λ rather than those in M. Therefore, it is believed that the enhanced λ/λ_{max}–M/M_{max} response in the composite compared to the alloy originates from the increased non-180° domain-wall motion in the composite during the magnetization process. Moreover, this increased non-180° domain-wall motion results from the axial residual compressive stress developed through the cure of the epoxy matrix in the composite as stated in Section 1 [4,5].

4. Conclusion

We have fabricated pseudo-1–3 magnetostrictive particulate composites of 0.5 particulate volume fraction by embedding and aligning light rare earth (Sm and Nd)-based magnetostrictive $Sm_{1-x}Nd_xFe_{1.55}$ particles with the Nd content x of 0–0.56 and randomly distributed sizes of 10–180 μm in a passive epoxy matrix. Investigations into the dependence of their quasistatic magnetomechanical properties as function of the magnetic field (H) and the Nd content x have shown similar qualitative trends with their alloys. The $Sm_{0.92}Nd_{0.08}Fe_{1.55}$ composite has demonstrated the largest unsaturated magnetostriction (λ_{max}) of −530 ppm at 500 kA/m and the highest piezomagnetic coefficient (d_{33}) of −2 nm/A at 100 kA/m, both at room temperature. These attractive magnetomechanical properties make the composite interesting for practical applications.

Acknowledgments

This work was supported by the Hong Kong Research Grants Council of the HKSAR Government (PolyU 5266/08E), The Hong Kong Polytechnic University (G-U741), the National Natural Science Foundation of China (50831006 and 50571098), and the National Basic Research Program (2010CB934603) of the Ministry of Science and Technology of China.

References

[1] A.E. Clark, in: E.P. Wohlfarth (Ed.), Ferromagnetic Materials, vol. 1, North-Holland, Amsterdam, 1980, p. 531.
[2] H.T. Savage, R. Abbundi, A.E. Clark, O.D. McMasters, J. Magn. Magn. Mater. 15 (1980) 609.
[3] N. Nersessian, S.W. Or, G.P. Carman, J. Magn. Magn. Mater. 263 (2003) 101.
[4] S.W. Or, N. Nersessian, G.P. Carman, IEEE Trans. Magn. 40 (2004) 71.
[5] S.W. Or, T.L. Li, H.L.W. Chan, J. Appl. Phys. 97 (2005) 10M308.
[6] L. Sandlund, M. Fahlander, T. Cedell, A.E. Clark, J.B. Restorff, J. Appl. Phys. 75 (1994) 5656.
[7] T.A. Duenas, G.P. Carman, J. Appl. Phys. 87 (2000) 4696.
[8] T.A. Duenas, G.P. Carman, J. Appl. Phys. 90 (2001) 2433.
[9] A.E. Clark, AIP Conf. Proc. 18 (1974) 1015.
[10] R. Abbundi, A.E. Clark, O.D. McMasters, J. Appl. Phys. 53 (1982) 2664.
[11] X.K. Lv, S.W. Or, W. Liu, X.H. Liu, Z.D. Zhang, J. Alloys Compd. 476 (2009) 24.
[12] W.J. Ren, Z.D. Zhang, X.P. Song, X.G. Zhao, X.M. Jin, Appl. Phys. Lett. 82 (2003) 2664.
[13] Y.G. Shi, S.L. Tang, Y.J. Huang, L.Y. Lv, Y.W. Du, Appl. Phys. Lett. 90 (2007) 142515.
[14] F. Yang, W. Liu, X.K. Lv, B. Li, S.Q. Li, J. Li, Z.D. Zhang, J. Magn. Magn. Mater. 322 (2010) 2095.
[15] V. Hari Babu, G. Markandeyulu, A. Subrahmanyam, Appl. Phys. Lett. 90 (2007) 252513.
[16] B.W. Wang, W.J. Lee, J.S. Song, B.K. Min, Y.M. Hao, J. Appl. Phys. 91 (2002) 9246.
[17] F.E. Pinkerton, J.F. Herbst, T.W. Capehart, M.S. Meyer, W.A. Fellberg, J. Appl. Phys. 85 (1999) 1654.
[18] T.A. Duenas, Giant magnetostrictive composites, Ph.D. dissertation, Univ. California, Los Angeles, 2001.
[19] G. Engdahl, Handbook of Giant Magnetostrictive Materials, Academic, San Diego, CA, 2000.