



Polymer content and particle size effects on polymer-bonded Terfenol-D/PZT magnetoelectric composites



Yang Song^{a,b,*}, Bo Liu^{a,**}, De'an Pan^a, Lirong Xu^a, Alex A. Volinsky^b, Shengen Zhang^a

^a Institute for Advanced Materials and Technology, University of Science and Technology Beijing, Beijing 100083, China

^b Department of Mechanical Engineering, University of South Florida, Tampa, FL 33620, USA

ARTICLE INFO

Article history:

Received 25 January 2016

Accepted 28 March 2016

Available online 30 March 2016

Keywords:

Magnetoelectric composite

Terfenol-D

Polymer content

Particle size

ABSTRACT

Polymer-bonded cylindrical Terfenol-D/PZT magnetoelectric (ME) composites have been prepared with varying polymer contents and Terfenol-D particle sizes. The axial ME voltage coefficient, $\alpha_{E,A}$, was investigated for studying the effects of polymer content and particle size on the composite ME performance. The Terfenol-D particles with four different size ranges ($< 75 \mu\text{m}$, $75\text{--}100 \mu\text{m}$, $100\text{--}150 \mu\text{m}$ and $150\text{--}180 \mu\text{m}$) were used. Additionally, the epoxy content of the composites was studied with five different weight ratios of 9 wt%, 12 wt%, 14 wt%, 16 wt% and 19 wt%. The composite with an epoxy content of 0.14 wt% and $100\text{--}150 \mu\text{m}$ particle range exhibited better overall ME performance. This investigation promotes selecting the appropriate polymer content and particle size to optimize fabrication of the polymer-bonded ME composites for high ME performance to meet practical applications.

© 2016 Elsevier B.V. All rights reserved.

1. Introduction

The magnetoelectric (ME) effect is defined as the induced dielectric polarization under applied magnetic field (H) through interfacial strain coupling of the two phases, or as the induced magnetization in the presence of an applied electric field [1]. Magnetoelectric composites made by combining piezoelectric (PE) and piezomagnetic (PM) materials can lead to remarkable ME effects at room temperature, compared with the single phase magnetoelectrics [2,3]. Recently, ME composites have drawn much attention as a popular research topic because of the excellent ME performance at room temperature for potential applications in multifunctional devices, such as memory devices, tunable microwave devices and sensors [4–6].

Giant magnetostrictive material, Terfenol-D ($\text{Tb}_{1-x}\text{Dy}_x\text{Fe}_{2-y}$), alloy is one of the best PM material candidates for ME composites [7]. However, some Terfenol-D properties hinder ME composite applications, including low mechanical strength, high eddy current losses at high working frequencies and fabrication size limits [8]. To solve the aforementioned problems, polymer-bonded ME composites are considered to have distinct advantages [9]. They are highly flexible, non-brittle and allow simple manufacturing processes at room temperature with various shapes and sizes. Polymer-bonded Terfenol-D ME composites that were earlier

reported by Cewen Nan et al. Ma and Nan [10] demonstrated Terfenol-D-epoxy (TDE) medium as a PM phase for the ME composite by using 73 vol% epoxy binding with Terfenol-D particles at room temperature to solve the brittleness and high eddy current losses problems of the Terfenol-D alloy. Zuo et al. [11] realized that the polymer phase content is directly influencing the ME properties as a non-functional phase. They fabricated Terfenol-D/PZT laminated composites with 97 wt% Terfenol-D particle (randomly distributed sizes $< 180 \mu\text{m}$) and 3 wt% epoxy binder, using warm compaction ($130 \text{ }^\circ\text{C}$, 154 MPa), which resulted in excellent voltage coefficient performance. Based on these studies, the authors investigated a one-step compression method for making TDE/PZT cylindrical ME composites, which were prepared by using $< 180 \mu\text{m}$ Terfenol-D particles and PZT cylinder with 12 wt% epoxy binder [12]. However, only a few studies have focused on the effects of polymer content and particle size on polymer-bonded magnetoelectric composites. In an effort to optimize the polymer-bonded magnetoelectric composites, polymer-bonded TDE/PZT cylinder ME composites with various polymer content and particle size have been investigated. The axial mode voltage coefficient ($\alpha_{E,A}$) of the samples was examined.

2. Experiment

The Terfenol-D particles were obtained by crushing bulk Terfenol-D single crystal alloy in argon atmosphere. The XRD pattern of the particles is shown in Fig. 1(a). The proposed Terfenol-D/PZT cylindrical ME composites were made up of the aforementioned

* Corresponding author at: Institute for Advanced Materials and Technology, University of Science and Technology Beijing, Beijing 100083, China.

** Corresponding author.

E-mail addresses: yang@mail.usf.edu (Y. Song), liubo@ustb.edu.cn (B. Liu).

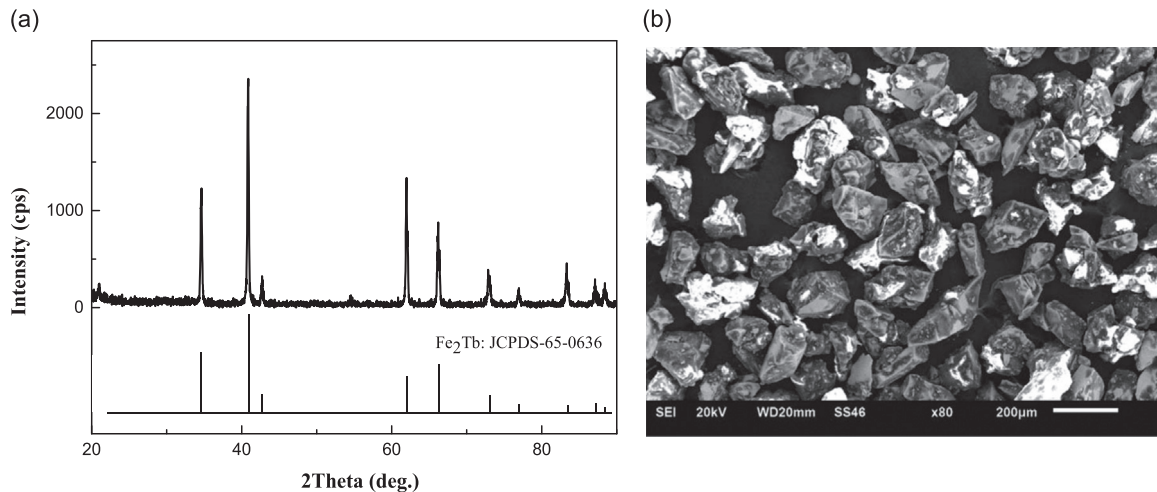


Fig. 1. (a) XRD pattern of the crushed Terfenol-D particles; (b) Terfenol-D particles with the 150–180 μm size, observed by SEM.

Terfenol-D particles and the PZT cylinder. The PZT cylinder was polarized along the radial direction. The Terfenol-D particles were compacted by uniaxial compression in the PZT cylinder, fabricated at 2 MPa pressure. The Terfenol-D-epoxy needed 24 h for curing. Detailed preparation method and schematic of the Terfenol-D/PZT cylindrical composite are shown elsewhere [12]. The ME performance of the Terfenol-D/PZT composites was obtained using the ME measurement system, where constant (H_{DC}) and alternating (δH) magnetic fields were applied in the axial mode. The ME voltage coefficient was calculated as $\alpha_E = \delta V / (t_{PE} \cdot \delta H)$, where t_{PE} is the thickness of PZT and δH is the amplitude of the AC magnetic field generated by the Helmholtz coils. The AC current flowing through the coil with the applied magnetic field amplitude of $\delta H = 1.2$ Oe was equal to 1 A. The axial mode ME voltage coefficient, $\alpha_{E,A}$, was measured when H_{DC} and δH were applied along the axial direction of the cylinder.

3. Results and discussion

The microstructure of crushed Terfenol-D particles after sieving was examined by X-ray diffraction (XRD). The XRD pattern in Fig. 1 (a) of the particles is almost identical to the standard Terfenol-D XRD pattern, meaning that the crystalline phases were not changed during the fabrication process [13]. The Terfenol-D particles with four different size ranges (< 75 , 75–100, 100–150 and 150–180 μm) were obtained using a set of two sieves with different meshes. Hence, the < 75 μm range indicates the particles which passed through the sieve with an opening size of 75 μm . The 75–100 μm range indicates that the particles passed through a 100 μm opening sieve, but were blocked by the 75 μm sieve. The particle size distribution is ideally uniform after sieving, noted by Fig. 1(b), for the 150–180 μm particle size range.

The epoxy content of the composites was studied with five different weight ratios of 9 wt%, 12 wt%, 14 wt%, 16 wt% and 19 wt%. Fig. 2 shows the ME voltage coefficient, $\alpha_{E,A}$, dependency on the volume ratio of various epoxy content with the 150–180 μm Terfenol-D particle size. As seen in Fig. 2, the ME voltage coefficient is greatly influenced by the epoxy content. The ME voltage coefficient reached the maximum value of $\alpha_{E,A} = 3.37$ V/cm·Oe when the epoxy content was 14 wt%. The ME voltage coefficient was degraded when the epoxy content was below 14 wt%. This could be due to the epoxy's inability to effectively play the binder and the lubricant roles at lower epoxy content [14]. When epoxy content increased over 14 wt%, the ME performance decreased

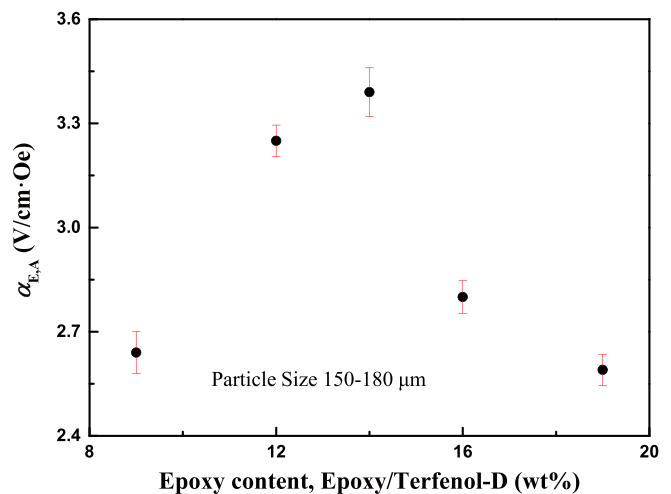


Fig. 2. The ME voltage coefficient, $\alpha_{E,A}$, dependence on the volume ratio of epoxy content. The results are for the sample with the particle size of 150–180 μm .

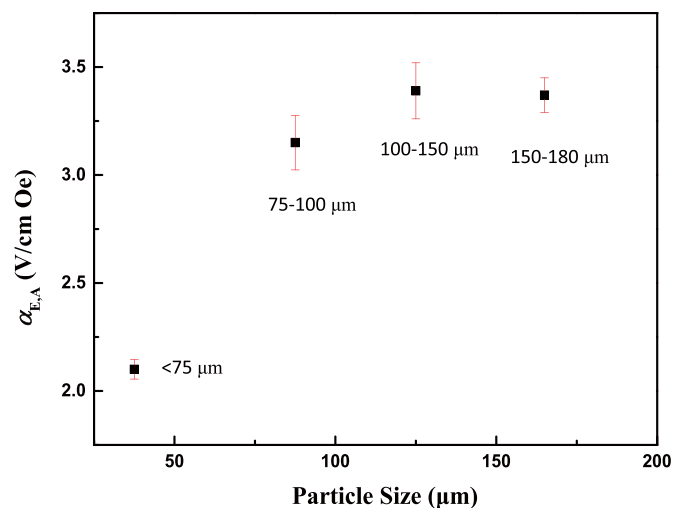


Fig. 3. The ME voltage coefficient, $\alpha_{E,A}$, dependence on the Terfenol-D particle size. The results are for the samples fabricated with the epoxy content of 0.14 wt%.

rapidly due to the dilution effect by the non-functional polymer phase [15].

Fig. 3 shows the ME voltage coefficient, $\alpha_{E,A}$, particle size

dependence for different Terfenol-D particle ranges of $< 75 \mu\text{m}$, $75\text{--}100 \mu\text{m}$, $100\text{--}150 \mu\text{m}$ and $150\text{--}180 \mu\text{m}$, with an epoxy content of 0.14 wt%. As seen in Fig. 3, The ME voltage coefficient of the $< 75 \mu\text{m}$ particle size, $\alpha_{E,A} = 2.1 \text{ V/cm Oe}$, is much lower than others. When the particle size is between 100 and $150 \mu\text{m}$, the ME voltage coefficient is the highest, up to 3.4 V/cm Oe . This result demonstrated that the ME voltage coefficient was also degraded when Terfenol-D particles were small. When particle size is between 100 and $180 \mu\text{m}$, the composite can get roughly 1.6 times higher ME performance, compared with the particle size less than $75 \mu\text{m}$.

4. Conclusions

In summary, the axial ME voltage coefficient of the polymer-bonded cylindrical Terfenol-D/PZT ME composites was studied. The voltage coefficient can be distinctly influenced by both polymer content and particle size. The composite with an epoxy content of 0.14 wt% and $100\text{--}150 \mu\text{m}$ particle sizes exhibited better overall ME performance. These findings promote selecting the appropriate polymer content and particle size to optimize fabrication of the polymer-bonded ME composites for high ME performance to meet practical applications.

Acknowledgements

This work was supported by the National Natural Science Foundation of China (U1360202, 51472030 and 51502014), by the

Beijing Nova Program (Z141103001814006), by the Fundamental Research Funds for the Central Universities (Project no.: FRF-TP-14-001C1) and the China Postdoctoral Science Foundation Funded Project (Project no.: 2014M560885). Alex Volinsky acknowledges support from the National Science Foundation under the IRES 1358088 grant. Yang Song acknowledges support from the China Scholarship Council (CSC No.: 201506460047).

References

- [1] W. Eerenstein, N.D. Mathur, J.F. Scott, *Nature* 442 (2006) 759–765.
- [2] M. Bibes, A. Barthelemy, *Nat. Mater.* 7 (2008) 425–426.
- [3] H. Yang, G. Zhang, Y. Lin, F. Wang, *Mater. Lett.* (2015).
- [4] J.H. Lee, L. Fang, E. Vlahos, X. Ke, Y.W. Jung, L.F. Kourkoutis, et al., *Nature* 466 (2010) 954–958.
- [5] Z. Shi, Y. Tong, S. Deng, H. Xue, S. Yang, Y. Lu, et al., *Appl. Phys. Lett.* 103 (2013) 032903.
- [6] B. Liu, Y. Fang, Z. Han, S. Yan, W. Zhou, B. Qian, et al., *Mater. Lett.* 164 (2016) 425–427.
- [7] M.-C. Lu, L. Mei, D.-Y. Jeong, J. Xiang, H. Xie, Q. Zhang, *Appl. Phys. Lett.* 106 (2015) 112905.
- [8] S. Lim, S. Kim, S. Kang, J. Park, J. Nam, D. Son, *J. Magn. Magn. Mater.* 191 (1999) 113–121.
- [9] P. Martins, S. Lanceros-Méndez, *Adv. Funct. Mater.* 23 (2013) 3371–3385.
- [10] J. Ma, Z. Shi, C.W. Nan, *Adv. Mater.* 19 (2007) 2571–2573.
- [11] Z.J. Zuo, D. Pan, Y.M. Jia, J.J. Tian, S.G. Zhang, L.J. Qiao, *J. Alloy. Compd.* 587 (2014) 287–289.
- [12] Y. Song, Da Pan, J. Wang, Z. Zuo, S. Zhang, B. Liu, et al., *AIP Adv.* 5 (2015) 037104.
- [13] M. Liu, S. Li, Z. Zhou, S. Beguhn, J. Lou, F. Xu, et al., *J. Appl. Phys.* 112 (2012) 063917.
- [14] A.K. Kadiyala, J. Bijwe, *Wear* 301 (2013) 802–809.
- [15] J. Liu, Z. Pan, X. Song, Z. Zhang, W. Ren, *J. Appl. Phys.* 117 (2015) 17A914.