

Effect of Dimension Control of Piezoelectric Layer on the Performance of Magnetolectric Laminate Composite

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Abstract Laminate composites composed of 0.95Pb(Zr_{0.52}Ti_{0.48})O₃-0.05Pb(Mn_{1/3}Sb_{2/3})O₃ piezoelectric ceramic and Fe-Si-B based magnetostrictive amorphous alloy are fabricated, and the effect of control of the areal dimensions and the thickness of the piezoelectric layer on the magnetolectric (ME) properties of the laminate composites is studied. As the aspect ratio of the piezoelectric layer and the magnetostrictive layer increases, the maximum value of the ME voltage coefficient (α_{ME}) increases and the intensity of the DC magnetic field at which the maximum α_{ME} value appears decreases. Moreover, as the thickness of the piezoelectric layer decreases, α_{ME} tends to increase. The ME composites exhibit α_{ME} values higher than 1 Vcm⁻¹Oe⁻¹ even at the non-resonance frequency of 1 kHz. This study shows that, apart from the inherent characteristics of the piezoelectric composition, small thicknesses and high aspect ratios of the piezoelectric layer are important dimensional determinants for achieving high ME performance of the piezoelectric-magnetostrictive laminate composite.

Key words magnetolectric, piezoelectric, magnetostriction.

1. Introduction

Magnetolectric (ME) material is a kind of multiferroic material whose electric polarization can be varied with change in magnetization, or vice versa. Various electronic devices such as magnetic field sensors, tunable phase shifters and filters, optical components, and energy harvesters have been proposed based on ME characteristics.¹⁻⁵⁾

Among the different types of ME materials, single-phase compounds have been known to possess weak ME properties even at low temperatures,^{6,7)} whereas two-phase ME composites have been observed to exhibit giant ME properties owing to the strain coupling between the piezoelectric and magnetostrictive phases.⁸⁻¹²⁾ In particular, laminate-type ME composites with 2-2 connectivity have been reported to have higher ME voltage coefficient values defined as $\alpha_{ME} = E_{AC}/H_{AC}$ (E_{AC} : AC electric field generated from the piezoelectric layer, H_{AC} : AC magnetic field applied to the magnetostrictive layer) than the other types.¹⁻³⁾ Employing the ME laminate composite structure, Wang et al. reported a very high magnetic field sensitivity using a 33-mode macro-fiber composite piezo-

electric layer,¹³⁾ and recently, Annapureddy et al. reported high-performance energy harvester applications using a 32-mode single-crystal piezoelectric layer.⁵⁾

The ME performance of the piezoelectric-magnetostrictive laminate composite is greatly influenced by the piezoelectric voltage coefficient, dielectric loss, and mechanical loss of the piezoelectric layer as it is a product property owing to strain coupling between the piezoelectric and magnetostrictive layers.^{5,14)} Meanwhile, when designing actual devices such as sensors, filters, and memories, it is necessary to understand the influence of the material dimensions as well as that of the material properties on the ME performance.

In this study, the effect of aspect ratio and thickness of the piezoelectric layer on the ME characteristics was investigated based on the laminate composites composed of 0.95Pb(Zr_{0.52}Ti_{0.48})O₃-0.05Pb(Mn_{1/3}Sb_{2/3})O₃ piezoelectric ceramic layer and Fe-Si-B based (Metglas) magnetostrictive amorphous metal layer. The areal dimensions of the piezoelectric layer and the magnetostrictive layer were controlled to be the same, and the thickness of the magnetostrictive layer was fixed to verify the effect of

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controlling only the thickness of the piezoelectric layer. The effects of the control of the aspect ratio and thickness of the piezoelectric layer and their causes are explained based on the ME voltage coefficient data and its integral value as a function of DC magnetic field.

2. Experimental Procedure

To prepare $0.95\text{Pb}(\text{Zr}_{0.52}\text{Ti}_{0.48})\text{O}_3\text{-}0.05\text{Pb}(\text{Mn}_{1/3}\text{Sb}_{2/3})\text{O}_3$ (PZT-PMS) piezoelectric layers, reagent grade raw powders of PbO , ZrO_2 , TiO_2 , MnO_2 , and Sb_2O_3 (all from Sigma-Aldrich) were mixed according to the stoichiometric composition and milled with ethyl alcohol and yttria-stabilized zirconia (YSZ) balls in a polyethylene jar for 24 h. Subsequently, the powder mixture was calcined at 850°C for 4 h. The calcined powder was ball-milled again with ethyl alcohol and YSZ balls for 24 h and dried at 80°C . The dried powder was pressed to a rectangular-shaped green body under 100 MPa, and the green body was sintered at 1200°C for 4 h in air atmosphere. The sintered PZT-PMS sample was cut into thin rectangular plates of various dimensions. An Ag electrode was formed on the top and bottom surfaces of the PZT-PMS plates, and electrical poling process was conducted by applying a DC electric field of 3 kVmm^{-1} for 30 min at 120°C in a silicone oil bath. For the measurement of 33- and planar-mode piezoelectric properties, a disk-shaped PZT-PMS sample of diameter 12 mm and thickness 1 mm was also prepared in a similar manner. The structural properties of the sintered PZT-PMS sample were examined using X-ray diffraction (XRD, D-MAX/2500, Rigaku) and scanning electron microscopy (SEM, JSM-6500F, JEOL), and the piezoelectric and dielectric properties were measured using d_{33} -meter (YE2730A, Sinocera Piezotronics) and impedance analyzer (IM3570, Hioki).

The piezoelectric-magnetostrictive laminate composites were prepared by attaching Metglas sheets (2605SA1, Metglas Inc.) on the top and bottom surfaces of the poled PZT-PMS plate using epoxy adhesive (DP460, 3M). To measure the ME voltage coefficients, the laminate composite was placed at the center of a Helmholtz coil located at the center of the electromagnet. The voltage induced on the laminate composite was monitored using a lock-in amplifier (SR860, Stanford Research System).

3. Result and Discussion

Fig. 1 shows the XRD pattern and fractured surface SEM image of the sintered PZT-PMS ceramic. The plane indices of the XRD peaks in Fig. 1 are all based on a cubic structure to simplify phase identification, although the PZT-PMS is a morphotropic phase boundary composition between rhombohedral and tetragonal phases. A

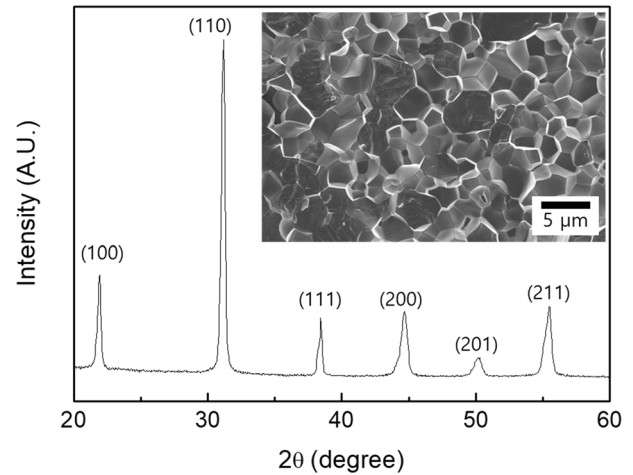


Fig. 1. XRD pattern and SEM image of PZT-PMS ceramic sintered at 1200°C .

Table 1. Piezoelectric and dielectric properties of PZT-PMS ceramic sintered at 1200°C .

d_{33} (pCN^{-1})	k_p	g_{33} (VmN^{-1})	Q_m	ϵ_3^T/ϵ_0	$\tan\delta$
235	0.52	0.049	652	542	0.003

homogeneous perovskite structure was well formed without any secondary phases and a dense microstructure was developed with an average grain size of $3\ \mu\text{m}$.

The sintered PZT-PMS specimen presented good piezoelectric properties as shown in Table 1. Moderate piezoelectric charge coefficient (d_{33}) of $235\ \text{pC}\text{N}^{-1}$ and electro-mechanical coupling coefficient (k_p) of 0.52 were obtained from the PZT-PMS. In particular, the piezoelectric voltage coefficient (g_{33}) and mechanical quality factor of the PZT-PMS were as high as $0.049\ \text{VmN}^{-1}$ and 652, respectively, demonstrating that the PZT-PMS is very suitable as a piezoelectric layer material in ME laminate composites.¹⁴⁾ A low dissipation factor ($\tan\delta$) of 0.003 is also very promising for high-performance ME devices.⁵⁾

Fig. 2 shows the constitution schematic of the piezoelectric-magnetostrictive laminate composite structure. The areal dimensions of the piezoelectric layer were designed to be the same as those of the magnetostrictive layers, and the aspect ratio (A/R) of the piezoelectric layer was defined as L/W (L : length, W : width). To confirm the effect of A/R control on the ME characteristics, we fabricated three laminate composite samples with various A/R s of 16 ($L = 16\ \text{mm}$, $W = 1\ \text{mm}$), 5 ($L = 25\ \text{mm}$, $W = 5\ \text{mm}$), and 1.8 ($L = 16\ \text{mm}$, $W = 9\ \text{mm}$), maintaining the thickness of the piezoelectric layer (t_p) at 1 mm. Two laminate composite samples with different t_p s of 0.45 mm and 0.25 mm were additionally prepared maintaining the A/R at 1.8, to confirm the effect of thickness control. In this study, the thickness of the magnetostrictive layers

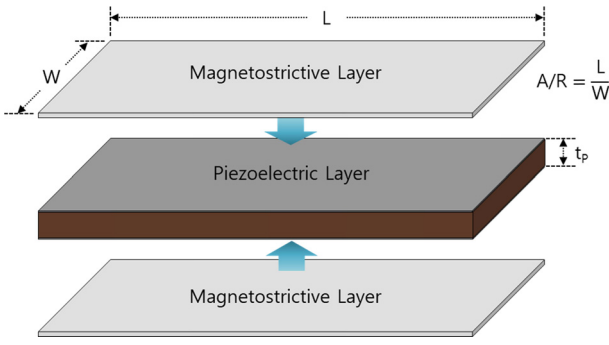


Fig. 2. Schematic diagram of the magnetoelastic laminate composite fabricated in this study.

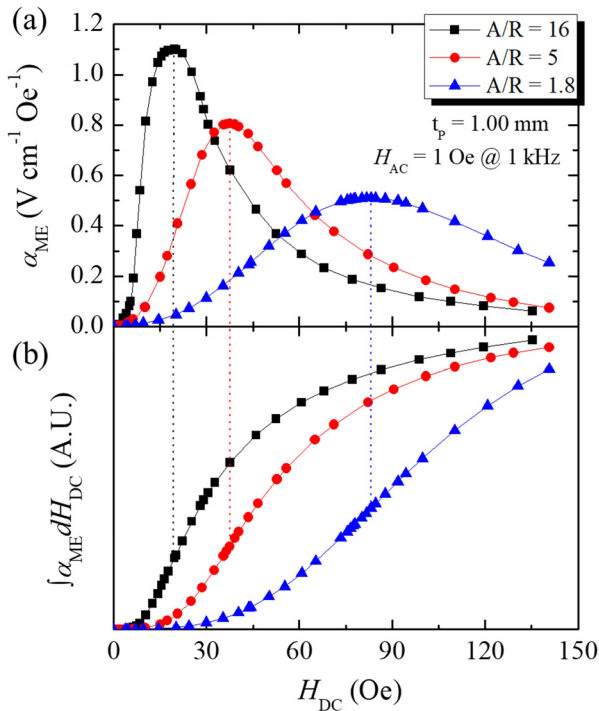


Fig. 3. (a) Magnetoelastic voltage coefficient of the laminate composites with various aspect ratios as a function of DC magnetic field, (b) integral values of the magnetoelastic voltage coefficient data in the same DC magnetic field range.

was fixed at $75\ \mu\text{m}$ (three-layered laminate of $25\text{-}\mu\text{m}$ -thick sheets) for all specimens.

Fig. 3(a) shows the ME voltage coefficient (α_{ME}) data of the laminate composites with various A/R s as a function of DC magnetic field (H_{DC}), measured at 1 kHz. All the composite samples represent a typical α_{ME} curve, exhibiting a maximum value at a specific H_{DC} . However, the maximum α_{ME} values and the H_{DC} values at which the maximum α_{ME} is observed are different for each sample. The maximum α_{ME} value of the laminate samples increased with the increase in A/R , whereas the

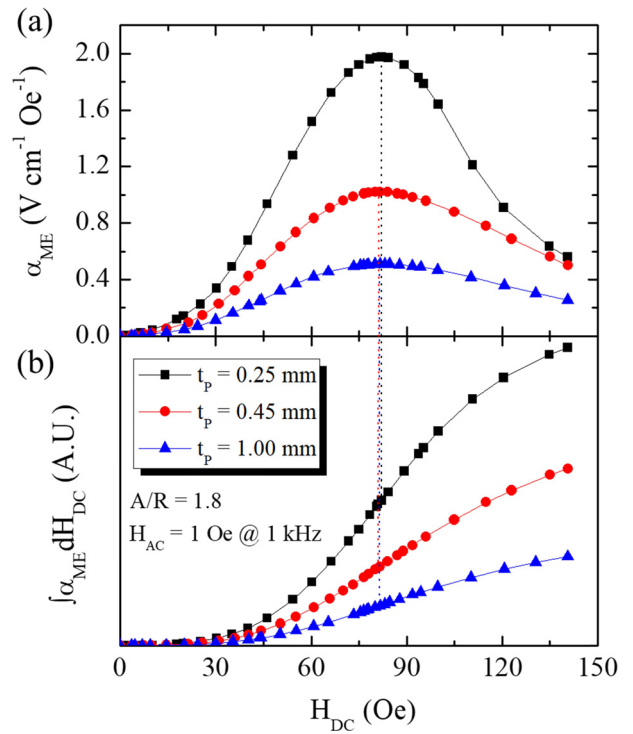


Fig. 4. (a) Magnetoelastic voltage coefficient of the laminate composites with various thicknesses of piezoelectric layer as a function of DC magnetic field, (b) integral values of the magnetoelastic voltage coefficient data in the same DC magnetic field range.

H_{DC} value at which the maximum α_{ME} was observed decreased with the increase in A/R .

It is well known that the α_{ME} of the laminate composite is directly proportional to the piezomagnetic coefficient of the magnetostrictive layers.^{1,14} The piezomagnetic coefficient (q_{11}) is defined as $q_{11} = d\lambda_1/dH_1$, where λ_1 is the magnetostriction in the longitudinal direction and H_1 is the DC magnetic field in the same direction. Therefore, the effective magnetostriction behaviors for each laminate composite can be predicted by integrating α_{ME} with respect to H_{DC} . As shown in Fig. 3(b), the integration curve of the specimen with A/R of 16 exhibits the sharpest increase along with the fastest saturation with the increase in H_{DC} , whereas the specimen with A/R of 1.8 exhibits the integration curve with the gentlest slope. As α_{ME} is the AC output value, it is maximized at the H_{DC} at which a small alternation in magnetic field (H_{AC}) induces the largest change in magnetostriction, i.e., where the slope of the integration curve is maximum. In Fig. 3(b), the specimen with A/R of 16 displays the steepest slope of the integration curve at the smallest H_{DC} ($\sim 19.2\ \text{Oe}$) among the three integration curves; therefore, the highest strain could be transferred to the piezoelectric layer under the same magnitude of H_{AC} (1 Oe), resulting in the highest α_{ME} value ($\sim 1.1\ \text{Vcm}^{-1}\text{Oe}^{-1}$).

As the ME response of the laminate composite is a product property, the magnetostriction of the composite mainly depends on the magnetostrictive materials and its characteristics are closely related to the magnetization of the magnetostrictive layer. It has been reported that the magnetic flux density generated from the magnetostrictive layer increases with the increase in the A/R of the magnetostrictive layer, owing to the reduction of the demagnetization field.^{15,16)} An increase in the magnetic flux density indicates an increase in the magnetization of the magnetostrictive layer. Furthermore, the magnetostriction is known to be proportional to the square of magnetization.¹⁷⁾ Therefore, the slope of λ vs. H_{DC} curve can be increased by employing a higher A/R of the magnetostrictive layer.

Fig. 4(a) displays the α_{ME} vs. H_{DC} curves of the laminate composites (A/R = 1.8) with various t_p s measured at 1 kHz. Overall, the α_{ME} value exhibits a tendency to increase with a decrease in t_p . The maximum α_{ME} of the specimen with t_p of 0.25 mm was as high as $1.98 \text{ Vcm}^{-1} \text{ Oe}^{-1}$, almost four times higher than that of the case with $t_p = 1.00 \text{ mm}$. As mentioned above in Fig. 3, the ME performance of the laminate composite is associated with the slope of the λ vs. H_{DC} curve. The specimen with t_p of 0.25 mm shows the steepest slope of the integration curve of α_{ME} as shown in Fig. 4(b); therefore, the highest α_{ME} can be obtained. A steep slope of the λ curve indicates that the magnetostrictive layers of the laminate composite are susceptible to deformation in response to the change in the magnetic field, resulting in a high piezomagnetic coefficient q . This might be because the total elastic modulus of the laminate composite decreases as the thickness of the piezoelectric layer decreases; further, the strain generated in the magnetostrictive layers is effectively transferred to the piezoelectric layer owing to the increased volume fraction of the magnetostrictive layers. Meanwhile, the H_{DC} at which the maximum α_{ME} is observed is almost the same at approximately 82 Oe in all three specimens. Therefore, the H_{DC} at which the maximum α_{ME} appears depends only on the A/R of the laminate composite (or magnetostrictive layer) and is independent of t_p .

4. Conclusion

The effect of variation of A/R and t_p of the piezoelectric layer on the ME characteristics of piezoelectric-magnetostrictive laminate composites was investigated. As the A/R increases, the maximum value of α_{ME} increases and the H_{DC} at which the maximum α_{ME} appears decreases. Moreover, α_{ME} tends to increase as the thickness of the piezoelectric layer decreases. It could be explained that the enhancement of the ME perfor-

mance with the increase in the A/R is caused by the increase in the slope of the magnetostriction curve owing to the increase in the magnetic flux density. The increase in the α_{ME} with the decrease in t_p appears to be caused by the increase in the magnetostriction owing to the decrease in the elastic modulus of the laminate and the increase in the volume fraction of the magnetostrictive layers. The results of this study suggest that it is advantageous to increase the A/R of the piezoelectric layer and reduce the thickness of the piezoelectric layer to obtain a high-performance ME laminate composite.

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References

1. C. W. Nan, M. I. Bichurin, S. Dong, D. Viehland and G. Srinivasan, *J. Appl. Phys.*, **103**, 031101 (2008).
2. G. Srinivasan, *Annu. Rev. Mater. Res.*, **40**, 153 (2010).
3. M. Fiebig, *J. Phys. D: Appl. Phys.*, **38**, R123 (2005).
4. C. W. Nan, G. Liu, Y. Lin and H. Chen, *Phys. Rev. Lett.*, **94**, 197203 (2005).
5. V. Annapureddy, M. Kim, H. Palneedi, H.-Y. Lee, S.-Y. Choi, W.-H. Yoon, D.-S. Park, J.-J. Choi, B.-D. Hahn, C.-W. Ahn, J.-W. Kim, D.-Y. Jeong, J. Ryu, *Adv. Energy Mater.*, **6**, 1601244 (2016).
6. J. Ryu, A. V. Carazo, K. Uchino and H. E. Kim, *Jpn. J. Appl. Phys.*, **40**, 4948 (2001).
7. R. A. Islam and S. Priya, *Integr. Ferroelectr.*, **82**, 1 (2006).
8. Y. B. Kamble, S. S. Chougule and B. Chougule, *J. Alloy Compd.*, **476**, 733 (2009).
9. J. W. Nie, G. Y. Xu, Y. Yang and C. W. Cheng, *Mater. Chem. Phys.*, **115**, 400 (2009).
10. Y. J. Li, X. M. Chen, Y. Q. Lin and Y. H. Tang, *J. Eur. Ceram. Soc.*, **26**, 2839 (2006).
11. D. R. Patil and B. K. Chougule, *J. Alloy Compd.*, **458**, 335 (2008).
12. S. T. Zhang, L. Y. Ding, M. H. Lu, Z. L. Luo and Y. F. Chen, *Solid State Comm.*, **148**, 420 (2008).
13. Y. Wang, D. Gray, D. Berry, J. Gao, M. Li, J. Li and D. Viehland, *Adv. Mater.*, **23**, 4111 (2011).
14. K.-H. Cho, C.S. Park and S. Priya, *Appl. Phys. Lett.*, **97**, 182902 (2010).
15. Z. Fang, S. G. Lu, F. Li, S. Datta, Q. M. Zhang and M. El Tahchi, *Appl. Phys. Lett.*, **95**, 112903 (2009).
16. H. Palneedi, S.-M. Na, G.-T. Hwang, M. Peddigari, K. W. Shin, K. H. Kim and J. Ryu, *J. Alloy Compd.*, **765**, 764 (2018).
17. R. E. Newnham, *Properties of Materials: Anisotropy, Symmetry, Structure*, Oxford University Press Inc., New York (2005).