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# Influence of AI Particle Size and Lead Zirconate Titanate (PZT) Volume Fraction on the Dielectric Properties of PZT-Epoxy-Aluminum Composites

Two-phase PZT-epoxy piezoelectric composites and three phase PZT-epoxy-Al composites were fabricated using a poling voltage of 0.2 kV/mm. The influence of aluminum inclusion size (nano and micron) and (lead zirconate titanate) PZT volume fraction on the dielectric properties of the three phase PZT-epoxy-aluminum composites were experimentally investigated. In general, dielectric and piezoelectric properties of the PZTepoxy matrix were improved with the addition of aluminum particles. Composites that were comprised of micron scale aluminum inclusions demonstrated higher piezoelectric  $d_{33}$ -strain-coefficients, and higher dielectric loss compared to composites that were comprised of nanosize aluminum inclusions. Specifically, composites comprised of micron sized aluminum particles and PZT volume fractions of 20%, 30%, and 40% had dielectric constants equal to 405.7, 661.4, and 727.8 (pC/N), respectively, while composites comprised of nanosize aluminum particles with the same PZT volume fractions, had dielectric constants equal to 233.28, 568.81, and 657.41 (pC/N), respectively. The electromechanical properties of the composites are influenced by several factors: inclusion agglomeration, contact resistance between particles, and air voids. These composites may be useful for several applications: structural health monitoring, energy harvesting, and acoustic liners. [DOI: 10.1115/1.4004812]

Keywords: composite, PZT-epoxy, PZT-Al-epoxy, dielectric loss

#### 1 Introduction

Two-phase composite piezoelectric polymers (CPPs), so-called 0-3 composites comprised of piezoelectric particles embedded within a continuous matrix, have attracted much attention due to their flexibility, ease of processing and use in embedded passive devices [1-12], such as capacitors. Integration of embedded passive components into printed circuit boards generally results in enhanced electrical performance of the device, improved reliability, reduction of device size, faster switching speed, and lower production costs [3,4,9]. Two-phase composites: metalpolymer and ceramic-polymer have been extensively studied [1,10,11,13–22] for application to coupling or by-pass capacitor technology, wherein emphasis has been placed on achievement of high effective dielectric constants via analysis based on percolation theory and mixing rules. However, as dielectric materials, these two-phase composites have relatively low values of dielectric constant compared to three phase 0-3-0 composite piezoelectric materials comprised of piezoelectric and electrically conductive particles embedded within a continuous matrix [1,2,23]. Thus, in order to enhance the dielectric properties of two-phase polymer-based composites, workers have investigated three phase composites consisting of a piezoelectric ceramic, metal, and polymer. Choi et al. [24] found that BaTiO<sub>3</sub>-PMMA-Ni composites comprised of 10% volume fraction of nickel (40  $\mu$ m in size) had dielectric constants equal to ~110 pC/N

compared to the two-phase CPP, BaTiO<sub>3</sub>–PMM, which had a dielectric constant equal to  $\sim 14.4 \text{ pC/N}$  [25]. In addition to higher dielectric constants, these three phase composites have low processing temperatures [24] and improved loss factors, which make them suitable for damping materials [1,16,19,20,23,26].

Inclusion of conductive fillers within polymer matrixes has been demonstrated by several other workers [16,23,24,27,28]. All have reported that the polymer matrix conductivity was enhanced by the electrically conductive filler. Ma and Wang [19] compared the microstructure and dielectric properties of epoxy-based damping composites that contained carbon nanotubes (CNTs) and PMN PZT piezoceramics. They concluded that the composites exhibited a percolation threshold in the range of 1.0-1.5 g CNTs per 100 g epoxy. They also concluded that in the region of the percolation threshold, a continuous electroconductive network was formed, and that beyond the percolation threshold, these materials demonstrated dynamic mechanical loss factors that were superior to those below the percolation threshold, and those without semiconductive inclusions. Tian and Wang [20] also examined the performance of epoxy/multiwalled carbon nanotube/piezoelectric ceramic composites as rigid damping materials. Their results were similar to Ma and Wang [19], where the percolation threshold was found to be in the range of 1.0-1.5 g CNTs per 100 g epoxy. They too concluded that loss factors were improved with the incorporation of CNT and (PZT), where the amount of CNT was above the critical electrical percolation loading. Hori et al. [26] found that three phase composites produced superior damping loss values in comparison to two-phase composites for specific volume fractions of piezoelectric and conductive material. They fabricated samples that were comprised of PZT, carbon black and epoxy. The three phase composite achieved a maximum value of damping loss

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Fig. 1 Plot of the effective dielectric constant of the two-phase composite,  $\varepsilon$ , as a function of PZT volume fraction

~0.08 for 0.51 wt.% of carbon black, which corresponded to PZT/CB/EP (69.7/0.5/29.8). This value of damping loss was a substantial increase from the single phase -100% epoxy and two-phase, PZT-epoxy composites (70/30 in wt.%) that had loss factors of 0.035 and 0.05, respectively. They found that when the carbon black content was  $\le 0.45$  wt.%, the blends behaved electrically as insulators, wherein the electric energy generated from PZT was not fully dissipated. However, for carbon black content ranges from 0.45 to 0.51 wt.%, the generated electric current flowed in the blends through carbon black particles, which resulted in enhanced damping loss values.

The size of the piezoelectric particles within the composite also has an influence on the dielectric constant of the three phase composites. The dielectric constant of the three phase CPP [7,8,27], BaTiO<sub>3</sub>–PMMA–Ni comprised of micron size BaTiO<sub>3</sub> particles was found to be ~150 pC/N in comparison to 100 pC/N for three phase composites with nanosize BaTiO<sub>3</sub> particles. The effects of the piezoelectric particle size on the bulk properties of the composite were attributed to the increase in percolation threshold of nickel that was caused when individual or aggregate BaTiO<sub>3</sub> particles surrounded the nickel particles [27].

Hence, the dielectric behavior of the composite is dependent upon the size, shape, and spatial distribution of the filler particles within the host matrix, the adhesion and the interaction between the phases, and the processing method [1,7,22,24,27,29–31]. In this paper, we report on the effects of the size of the conductive particle on the dielectric properties of the piezoelectric-epoxyconductor composite. Specifically, the influence of the size of the Al particle on the dielectric constant, strain coefficient and tan  $\delta$  is observed. The PZT-epoxy-Al composites examined comprised of micron and nanosize aluminum particles.

#### 2 Experimental

PZT powder manufactured by READE International (CAS #12626-81-2, EC-76) and epoxy resin (DGEBA, Epofix<sup>TM</sup> Cold-Setting Embedding Resin) manufactured by Electron Microscopy Sciences were mixed by hand using a stainless steel rod and stirred for 15 min. The mixture of PZT and epoxy was then ball milled for 1 h. Aluminum particles: nanosize, 99.95% purity, spherical morphology (APS: 18 nm) and micron, 99.97% purity spherical morphology, 200 mesh were supplied by MTI and Acros-Fisher Scientific, respectively. The volume fraction of aluminum was held constant at 20%. The aluminum particles were dispersed in ethanol and then stirred with a mechanical stirrer for 15 min. The mixture of PZT and epoxy was then added to the Al dispersion in ethanol mixture. An epoxy hardener was added, after the ethanol was evaporated at room temperature for approximately 12 min. The final mixture was then poured into a

circular mold that was 6 mm thick with a radius equal to 7 mm. The mold, containing the sample, was placed in between the top and bottom platens of a Carver Hydraulic press, and subjected to 500 psi for 5 min at room temperature. The samples were cured at 75 °C for 8 h. The samples were polished and subsequently painted on the top and bottom surfaces with conductive silver paint. The samples were then poled at 0.2 kV/mm at 1200 °C for 10 min in silicone oil. After poling, the sample was wrapped in Al foil for 24 h to get rid of residual charges. The dielectric constant, dielectric loss tangent, and piezoelectric strain coefficient at 110 Hz were recorded using a commercial d<sub>33</sub>-meter from Piezotest, and the impedance from 100 to 1500 kHz was recorded for each sample using a HP 4194 A Impedance analyzer. The fractured surface morphology of the samples was studied with the help of a scanning electron microscope (FESEM ZEISS 982). Two samples were prepared for each volume fraction of PZT, e.g., 10%, 20%, 30%, 40%, 50%, 60%, and 70%. The values presented in the figures are the average values from the two samples prepared.

#### **3** Results and Discussion

Two-phase PZT-epoxy composites were fabricated using the methodology described above (without inclusion of aluminum particles), where the volume fraction of PZT was varied from 10% to 70%. The dielectric constant of the two-phase composite is plotted as a function of volume fraction of PZT in Fig. 1. As expected, the dielectric constant of the composite increased with the volume fraction of PZT. The dielectric constant plot as a function of piezoelectric volume fraction followed the same trend as Refs. [27]and [30], wherein the plotted results fall between the Maxwell Garnett approximation

$$\varepsilon = \varepsilon_{\text{Epoxy}} \left[ 1 + \frac{1 + 3\phi_{\text{PZT}}\beta}{1 - \phi_{\text{PZT}}\beta} \right]$$
(1)

and the Bruggeman's effective medium approximation [27]

$$(1 - \phi_{\rm PZT})\frac{\varepsilon_{\rm Epoxy} - \varepsilon}{\varepsilon_{\rm Epoxy} + 2\varepsilon} + \phi_{\rm PZT}\frac{\varepsilon_{\rm PZT} - \varepsilon}{\varepsilon_{\rm PZT} - \varepsilon} = 0$$
(2)

In Eqs. (1) and (2),  $\phi_{PZT}$  is the volume fraction of PZT,  $\beta = (\epsilon_{PZT} - \epsilon_{Epoxy})/(\epsilon_{PZT} + 2 \epsilon_{Epoxy})$ , and  $\epsilon, \epsilon_{PZT}$ , and  $\epsilon_{Epoxy}$  represent the dielectric constant of the two-phase composite, PZT and epoxy, respectively. Both analytical models can be used to determine the effective dielectric constant of a matrix-based composite with embedded inclusions [27]. In general, the Maxwell expression underestimates [27] the effective dielectric constant,

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Fig. 2 SEM micrographs of two-phase (PZT/epoxy) composite with PZT volume fraction of 50% and magnifications of (a) 1000 and (b) 5000

while Bruggeman's expression overestimates this value as the volume fraction of the piezoelectric material increases. For our data, the Maxwell expression underestimates and Bruggeman's expression overestimates the empirical results for  $\phi_{PZT} > 30\%$ . This is due to the aggregation of PZT and epoxy particles in the composite along with the increase in PZT volume fraction, which results in continuous percolation clusters in the composites [27]. A SEM image that details the distribution of PZT within the epoxy matrix is provided in Figs. 2(*a*) and 2(*b*). The SEM micrographs of the PZT-epoxy composite are magnified 1000 and 5000 times, respectively. The arrows indicate the PZT and epoxy matrix. In Fig. 2(*b*), evidence of PZT clustering is seen, which supports the explanation for the underestimation of the Maxwell Garnett approximation.

In Fig. 3, a plot of the effective dielectric constant as a function of PZT volume fraction is provided for the PZT-epoxy and the three phase composites with micron and nanosize Al inclusions. The volume fraction of the Al inclusions is held constant at 20% for all three phase composites. As expected, the dielectric constants of the three phase composites are greater than those of the PZT-epoxy. This is attributed to the addition of the aluminum particles. This enhancement can be explained by the following power law [24,27]:

$$\varepsilon_{3-\text{Phase}} = \varepsilon \left| \frac{\phi_c - \phi}{\phi_c} \right|^{-q} \tag{3}$$

where  $\varepsilon$  is the dielectric constant of the PZT-epoxy two-phase composite,  $\phi_c$  is the percolation threshold volume fraction,  $\phi$  is the volume fraction of the aluminum, and q is the critical exponent. A value of 1.3 was determined for the critical exponent, q, when a value of 16% [27,30] for the percolation threshold of aluminum,  $\phi_c$ , and the experimental values for the dielectric constant were substituted into Eq. (3).

The effective dielectric constant for the composite that incorporates microsize aluminum particles presents higher effective dielectric constants for equivalent volume fractions of PZT than the composites with nanosize aluminum particles. For example, at volume fractions of 20%, 30%, and 40% of PZT, the dielectric constants of the microcomposites are 405.74, 661.38, and 727.78 (pC/N) as compared to 233.28, 568.81, and 657.41 (pC/N), respectively, for the nanocomposite. Differences in the piezoelectric strain coefficient and loss factor, tan  $\delta$ , are also observed when aluminum particles are added to the two-phase composite, and when the size of the aluminum particles is varied. Tan  $\delta$  is an important parameter in the characterization of the macromolecular viscoelasticity of the material. It represents the damping capacity of the material and describes the ability of the material to convert mechanical energy into heat energy, when subjected to an external load. In general, tan  $\delta = E''/E'$ , where tan  $\delta$  is the phase angle between the stress and strain [20], while E' and E" are the elastic storage modulus and elastic loss modulus, respectively. The results provided in Fig. 4 indicate that all of the composites show an increase in loss factor at higher concentrations of PZT. This



Fig. 3 A comparison of the dielectric constants of the two-phase and three phase microcomposite and nanocomposite

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Fig. 4 A comparison of the dielectric loss of the three phase microcomposite and nanocomposite with the two-phase PZT-epoxy composite with increasing PZT volume fraction shows that microcomposites have the highest dielectric loss a compared to the nanocomposite and two-phase composite

trend agrees well with those noted by Refs. [19] and [20]. The experimental values also follow the power law [24]:

$$\tan \delta = \tan \delta_{\rm o} \left| \frac{\phi_c - \phi}{\phi_c} \right|^{-r} \tag{4}$$

In Eq. (4), tan  $\delta_o$  is the loss tangent of the two-phase matrix and r is direct current conductivity of the composites. The experimental values of the dielectric loss tangent are in good agreement with Eq. (4) and follow the same trend as Refs. [27] and [30], r may be determined from the empirical date provided. The composites that have the micron size aluminum particles present higher values of tan  $\delta$  than the composites that incorporate nanosize aluminum constituents. For example, the tan  $\delta$  values for a PZT volume fraction equal to 70% are 0.014, 0.0273, and 0.0457 for the PZT-epoxy, PZT-epoxy-Al/nano, and PZT-epoxy-Al/microcomposites, respectively.

In Fig. 5, a plot of the piezoelectric strain coefficient,  $d_{33}$ , as a function of PZT volume fraction is presented. As evidenced by other workers [32,33],  $d_{33}$  increases with the volume fraction of PZT in composite piezoelectric materials. Similar to the loss factor results presented in Fig. 4, the strain coefficient of the composites containing nanosize particles of aluminum is lower than those containing micron size particles of aluminum. The strain coefficient is a function of the capacitance of the material, which

is proportional to the number of charges that are stored on the surface of the sample under an applied electric field. The inclusion of dielectric fillers result in higher permittivity of the composite and ultimately higher piezoelectric coefficients. In fact, the  $d_{33}$  coefficient of the microcomposite is almost three times that of the nano-composite for PZT volume fractions of 40% and 50%.

In Figs. 6(a) and 6(b), the distribution of the Al microparticles in the PZT-epoxy matrix can be observed. In these figures, clustering of the Al is observed. In Figs. 7(a) and 7(b), the three phase composite comprised of nanosize Al inclusions are provided at magnifications of 1000 and 5000. The aluminum particles in the nanometer range have more surface area to interact with the PZTepoxy matrix than that of the microsized Al inclusions. Thus, the interfacial effect [27] between the materials becomes more prominent as the size of the conductive inclusion diminishes. Agglomeration of Al nanoparticles is circled in the SEM micrographs. Both interfacial effects and agglomeration are important factors governing the dielectric and piezoelectric characteristics of the composite.

The difference in performance of the composites comprised of nano and micron size aluminum particles is the result of several factors. Contact resistance between the conductive material and the polymer matrix and dielectric material reduce the conductivity of the conductive materials themselves. Smaller size conductive materials provide higher contact surface area per unit volume of



Fig. 5 A comparison of the piezoelectric strain coefficient  $d_{33}$  of the three phase microcomposite and nanocomposite with the two-phase PZT-epoxy composite with increasing PZT volume fraction. Microcomposites have the highest dielectric loss as compared to the nanocomposite and two-phase composite.

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Fig. 6 SEM micrographs of PZT-epoxy-AI microcomposite with PZT volume fraction of 50% and magnifications of (a) 1000 and (b) 5000



Fig. 7 SEM micrographs of PZT-epoxy-AI microcomposite with PZT volume fraction of 50% and magnifications of (a) 1000 and (b) 5000

composite material than composites comprised of micron sized materials. This results in reduced values in strain coefficient for composites comprised of nanoconductive particles in comparison to those comprised of micron size particles. Also, composite homogeneity is a critical parameter in achieving optimal performance and is a challenge to achieve due to space charge effects. Adequate dispersion of PZT and conductive filler within the polymer matrix, and minimal air voids are desirable. However, agglomeration of PZT and conductive filler material within the matrix is common [21] as evidenced by the SEM images. The agglomeration of the constituent materials can lead to reduced mechanical, electrical, and conductive properties in the materials. Agglomeration was observed in all of the sets of composite materials, in particular those containing nanosize aluminum constituents. Also, addition of ceramic materials tends to introduce flaws, in the form of agglomerations that contribute to scattering of results [21].

## 4 Conclusions

Piezoelectric composites comprised of PZT, epoxy, and Al were fabricated, and the size of the aluminum particles was varied from nano to micro in size. The dependence of the dielectric and piezoelectric characteristics of the composite on the volume fraction of PZT has been established. The enhancement of dielectric and piezoelectric properties resulting from the addition of Al

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inclusions and increase of PZT volume fraction has been shown. A comparative study on the microcomposite and nanocomposite showed that the influence of the interfacial effect and Al nanoparticle agglomeration on the dielectric and piezoelectric characteristics of the bulk three phase composite. The difference in properties of the microcomposite and nanocomposite proves that the dielectric and piezoelectric properties of these particulate composites can be tailored by varying inclusion particle size, while the other parameters remain fixed. The ease of processing, flexibility, and good dielectric behavior of the three-phase piezoelectric particulate composites potentially makes the composites suitable for practical applications in energy harvesting, acoustic dampening, and structural health monitoring.

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#### References

- Bai, Y., Cheng, Z. Y., Bharti, V., Xu, H. S., and Zhang, Q. M., 2000, "High-Dielectric-Constant Ceramic-Powder Polymer Composites," Appl. Phys. Lett., 76(25), p. 3804–3807.
- [2] Kuo, D. H., Chang, C. C., Su, T. Y., Wang, W. K., and Lin, B. Y., 2001, "Dielectric Behaviours of Multi-Doped BaTiO<sub>3</sub>/Epoxy Composites," J. Eur. Ceram. Soc., 21, pp. 1171–1177.

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- [3] Funer, R., 2002, "Embedding Resistors and Capacitors," Printed Circuit Des., 19(12), p. 8.
- [4] Ulrich, R., 2004, "Matching Embedded Capacitor Dielectrics to Applications," Circlut World, 30(1), p. 20.
- [5] Bhattacharya, S. K., and Tummala, R. R., 2000, "Next Generation Integral Passives: Materials, Processes, and Integration of Resistors and Capacitors on PWB Substrates," J. Mater. Sci.: Mater. Electron., 11(3), pp. 253–268.
- [6] Bhattacharya, S. K., and Tummala, R. R., 2002, "Epoxy Nanocomposite Capacitors for Application as MCM-L Compatible Integral Passives," ASME J. Electron. Packag., 124(1), pp. 1–6.
  [7] Choi, H. W., Heo, Y. W., Lee, J. H., Kim, J. J., Lee, H. Y., Park, E. T., and
- [7] Choi, H. W., Heo, Y. W., Lee, J. H., Kim, J. J., Lee, H. Y., Park, E. T., and Chung, Y. K., 2006, "Effects of BaTiO<sub>3</sub> on Dielectric Behavior of BaTiO<sub>3</sub>-Ni-Polymethyl Methacrylate Composites," Appl. Phys. Lett., 89(13), 132910.
- [8] Choi, H. W., Heo, Y. W., Lee, J. H., Kim, J. J., Lee, H. Y., Park, E. T., and Chung, Y. K., 2007, "Effects of Ni Particle Size on Dielectric Properties of PMMA-Ni-BaTiO<sub>3</sub> Composites," Integr. Ferroelectr., 87, p. 85.
- [9] Kim, H., Sun, B. K., and Kim, J., 2004, "Suppression of GHz Range Power/ Ground Inductive Impedance and Simultaneous Switching Noise Using Embedded Film Capacitors in Multilayer Packages and PCBs," IEEE Compon. Lett., 14(2), p. 71.
- [10] Li, L., Takahashi, A., Hao, J. J., Kikuchi, R., Hayakawa, T., Tsurumi, T. A., and Kakimoto, M. A., 2005, "Novel Polymer-Ceramic Nanocomposite Based on New Concepts for Embedded Capacitor Application (I). IEEE Trans. Compon. Packag. Technol., 28(4), p. 754.
- [11] Ogitani, S., Bidstrup-Allen, S. A., and Kohl, P. A., 2000, "Factors Influencing the Permittivity of Polymer/Ceramic Composites for Embedded Capacitors," IEEE Trans. Adv. Packag., 23(2), pp. 313–322.
  [12] Rao, Y., and Wong, C. P., 2004, "Material Characterization of a High-Dielec-
- [12] Rao, Y., and Wong, C. P., 2004, "Material Characterization of a High-Dielectric-Constant Polymer-Ceramic Composite for Embedded Capacitor for RF Applications," J. Appl. Polym. Sci., 92(4), pp. 2228–2231.
- [13] Cai, L. F., Mai, Y. L., Rong, M. Z., Ruan, W. H., and Zhang, M. Q., 2007, "Interfacial Effects in Nano-Silica/Polypropylene Composites Fabricated by In-Situ Chemical Blowing," eXPRESS Polym. Lett., 1(1), pp. 2–7.
  [14] Nan, C. W., Li, M., and Huang, J. H., 2001, "Calculations of Giant Magneto-Definition of Content of Content
- [14] Nan, C. W., Li, M., and Huang, J. H., 2001, "Calculations of Giant Magnetoelectric Effects in Ferroic Composites of Rare-Earth-Iron Alloys and Ferroelectric Polymers," Phys. Rev. B, 63(14), 144415.
- [15] Nan, C.-W., Liu, L., Cai, N., Zhai, J., Ye, Y., Lin, Y. H., Dong, L. J., and Xiong, C. X., 2002, "A Three-Phase Magnetoelectric Composite of Piezoelectric Ceramics, Rare-Earth Iron Alloys, and Polymer," Appl. Phys. Lett., 81(20), pp. 3831–3834.
- [16] Qi, L., Lee, B. I., Samuels, W. D., Exarhos, G. J., and Sam J., Parler, G., 2006, "Three-Phase Percolative Silver–BaTiO<sub>3</sub>–Epoxy Nanocomposites With High Dielectric Constants," J. Appl. Polym. Sci., **102**, pp. 967–971.
- [17] Ho, C. H., Poon, Y. M., and Shin, F. G., 2006, "New Explicit Formulas for the Effective Piezoelectric Coefficients of Binary 0-3 Composites," J. Electroceram., 16, pp. 283–288.
- [18] Newnham, R. E., Skinner, D. P., Klicker, K. A., Bhalla, A. S., Hardiman, B., and Gururaja, T. R., 1980, "Ferroelectric Ceramic-Plastic Composites for Piezoelectric and Pyroelectric Applications," Ferroelectrics, 27(1–4), pp. 49–55.

- [19] Ma, M., and Wang, X. D., 2009, "Preparation, Microstructure and Properties of Epoxy-Based Composites Containing Carbon Nanotubes and PMN-PZT Piezoceramics as Rigid Piezo-Damping Materials," Mater. Chem. Phys., 116(1), pp. 191–197.
- [20] Tian, S., and Wang, X. D., 2008, "Fabrication and Performances of Epoxy/Multi-Walled Carbon Nanotubes/Piezoelectric Ceramic Composites as Rigid Piezo-Damping Materials," J. Mater. Sci., 43(14), pp. 4979–4987.
- [21] Tsantzalis, S., Karapappas, P., Vavouliotis, A., Tsotra, P., Paipetis, A., Kostopoulos, V., and Friedrich, K., 2007, "Enhancement of the Mechanical Performance of an Epoxy Resin and Fiber Reinforced Epoxy Resin Composites by the Introduction of CNF and PZT Particles at the Microscale," Composites, Part A, 38(4), pp. 1076–1081.
- [22] Doyle, W. T., and Jacobs, I. S., 1992, "The Influence of Particle Shape on Dielectric Enhancement in Metal-Insulator Composites," J. Appl. Phys., 71(8), pp. 3926–3937.
- [23] Yao, S.-H., Dang, Z.-M., Jiang, M.-J., and Bai, J., 2008, "BaTiO<sub>3</sub>-Carbon Nanotube/Polyvinylidene Fluoride Three-Phase Composites With High Dielectric Constant and Low Dielectric Loss," Appl. Phys. Lett., 93(18), 182905.
- [24] Choi, H.-W., Heo, Y.-W., Lee, J.-H., Kim, J.-J., Lee, H.-Y., Park, E.-T., and Chung, Y.-K., 2006, "Effects of BaTiO<sub>3</sub> on Dielectric Behavior of BaTiO<sub>3</sub>-Ni-Polymethyl Methacrylate Composites," Appl. Phys. Lett., 89(13), 132910.
- [25] Choi, H.-W., Heo, Y.-W., Lee, J.-H., Kim, J.-J., Lee, H.-Y., Park, E.-T., and Chung, Y.-K., 2006, "Effects of BaTiO<sub>3</sub> on Dielectric Behavior of BaTiO<sub>3</sub>-Ni-Polymethyl Methacrylate Composites," Appl. Phys. Lett., 89(13), 132910.
- [26] Hori, M., Aoki, T., Ohira, Y., and Yano, S., 2001, "New Type of Mechanical Damping Composites Composed of Piezoelectric Ceramics, Carbon Black and Epoxy Resin," Composites, Part A, 32(2), pp. 287–290.
- [27] Dang, Z. M., Shen, Y., and Nan, C.W., 2002, "Dielectric Behavior of Three-Phase Percolative Ni-BaTiO<sub>3</sub> Polyvinylidenmne Fluoride Composites," Appl. Phys. Lett., 81(25), pp. 4814–4817.
- [28] Miller, S. G., 2008, Effects of Nanoparticle and Matrix Interphase on Nanocomposite Properties, The Graduate Faculty of The University of Akron, University of Akron, p. 191, Akron, OH.
- [29] Ahmad, Z., Prasad, A., and Prasad, K., 2009, "A Comparative Approach to Predicting Effective Dielectric, Piezoelectric and Elastic Properties of PZT/PVDF Composites," Physica B, 404(20), pp. 3637–3644.
- [30] Dang, Z. M., Fan, L. Z., Shen, Y., and Nan, C. W., 2003, "Dielectric Behavior of Novel Three-Phase MWNTs/BaTiO<sub>3</sub>/PVDF Composites," Mater. Sci. Eng., B, 103(2), pp. 140–144.
- [31] Huang, C., Zhang, Q. M., deBotton, G., and Bhattacharya, K., 2004, "All-Organic Dielectric-Percolative Three-Component Composite Materials With High Electromechanical Response," Appl. Phys. Lett., 84(22), pp. 4391–4394.
- [32] Venkatragavaraj, E., Satish, B., Vinod, P. R., and Vijaya, M. S., 2001, "Piezoelectric Properties of Ferroelectric PZT-Polymer Composites," J. Phys. D, 34(4), p. 487.
- [33] Burianova, L., Hana, P., Panos, S., Kulek, J., and Tyagur, Y. I., 2000, "Piezoelectric, Dielectric and Pyroelectric Properties of 0-3 Ceramic-Polymer Composites," Ferroelectrics, 241(1–4), pp. 59–66.