

EFFECTIVE MEDIUM THEORY OF THE MECHANICAL, DIELECTRIC AND PIEZOELECTRIC PROPERTIES OF FERROELECTRIC CERAMIC-POLYMER COMPOSITE

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ABSTRACT. An effective medium theory of ferroelectric ceramic-polymer composite materials which treats both components symmetrically has been investigated to demonstrate the role played by the microgeometry of inclusions on dielectric, mechanical and piezoelectric properties of 0-3 composites. The limits of the various theoretical predictions as a function of the geometric parameters n_1 and n_2 are determined. We have observed that the geometric effect is more significant for the component with low values of the various material constants. Comparisons of predictions of the effective medium theory and spherical particle dispersion theory with several experimental results on the magnitude and loss tangents of elastic, dielectric, and piezoelectric coupling coefficients are given. The predictions of the effective medium theory on all the coupling constants magnitude and loss tangent are in good accord with experiment. The theory enables predictions of dielectric and elastic constants of composites from piezoelectric constant data and vice versa.

INTRODUCTION

Composites of the $\text{Pb}(\text{Zr}, \text{Ti})\text{O}_3$ or the PZT family with polymers are important both scientifically and industrially [1-10]. They are selected for the best individual properties and put together in a manner designed to make maximum use of these properties. A production of an electric response due to mechanical excitation (piezoelectricity) or thermal excitation (pyroelectricity) is revealed in such composite systems. In the direct piezoelectric effect an applied mechanical force is coupled to an electrical response in an acentric material. It is through these coupled properties that composite materials are expected to play a vital role in device design. Such composite materials are used as transducers for electric keyboards and pressure sensors.

In the study of ferroelectric ceramic-polymer composites, the concept of phase connectivity, or the number of dimensions in which each component phase is self-connected between the limiting surfaces of the composite, has proved to be very useful in the understanding of their properties as compared with those of the phases. Among the ten possible connectivities for two phase composite systems, those with 0-3 connectivity are very useful because of their potential use as soft transducers. A composite with 0-3 connectivity consists of a three dimensionally connected polymer matrix loaded with piezoelectrically active ceramic particles. Ceramic rods, one-

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dimensionally connected between electrodes and held in a three-dimensionally connected polymer matrix are called 3-1 composites [1,9].

Piezoelectric, dielectric and mechanical properties of a composite are affected by several factors among which are composition, shape, connectivity, properties of the individual components, temperature and frequency of measurement [3-12]. In some materials not only are the properties of the separate phases modified, but composites may exhibit completely new couplings not found in the separate phases. One of the inherent problems in composite systems has been in predicting their macroscopic properties using the properties of the constituents.

Theoretical investigations on polycrystalline ceramics [8] and binary systems [5-6] have been performed with regard to the piezoelectric constant, the dielectric constant, and the elastic constant. However, some of the methods are very complicated and may not be appropriate for applying the theories to such a composite. Spherical particle dispersion theories which uses Maxwell-Garnet (MG) type approximations [13-14] are commonly used to interpret experimental data of composite materials [11-12, 15-17]. It is well known that MG type theories are too simple to take care of all the important factors which influence composite properties. A strong piezoelectricity is needed for a composite system to be a useful one. This could be achieved by preparing a composite with a large PZT volume fraction. So it is hard to apply MG type theories to such a composite. At high ceramic composition of interest the role of inclusion and host interchanges. This makes it necessary to consider the shape of the matrix in developing a theory of composite materials. The effect of inclusion shape in the composite dielectric [18-19], elastic [18] and piezoelectric d-constant [18] via the permittivity has been demonstrated. In an earlier paper [20] it was shown that the elastic local field also depends on the shape of the inclusion which influences all the piezoelectric coupling coefficients.

In this work we have investigated an effective medium theory developed in the earlier publication to demonstrate the role played by shape and connectivity on properties of composites. We have observed that shape and connectivity effects are inseparable within the limits of the theory. We have compared the predictions from the theory with experimental data. Predictions are also made on the complex dielectric, elastic and piezoelectric properties of composites. The predictions on the magnitude and loss tangents of these constants are compared with experimental results [11, 18].

In the next section predictions of the effective medium theory are derived and compared against the spherical particle dispersion theory (MG). This is then followed by the presentation of the results and discussion. The important conclusions are summarized in the last section.

EFFECTIVE MEDIUM THEORY

The model used to develop the effective medium theory is a symmetrical effective medium which can automatically change the role of matrix and inclusion phases depending on the composition of the system. Such models are generally classified as Bruggeman Approximation (BA) [21]. Figure 1 shows a representation of the model. In the first case a polymer inclusion (phase 1), the blank ellipsoid, is placed in an effective medium. In the second case a ceramic inclusion (phase 2), the lined ellipsoid, is placed in an effective medium. In the Bruggeman Approximation the effective medium is expressed as a linear combination of the two cases. In the following, we shall give the

effective medium theory expressions for the dielectric, elastic and piezoelectric properties.

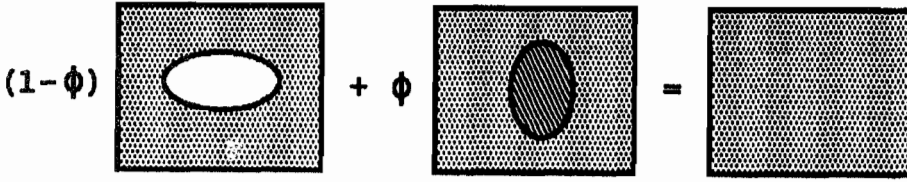


Figure 1. Representation of Bruggeman Approximation. The blank ellipsoid, lined ellipsoid and dotted regions represent polymer inclusion, ceramic inclusion and effective medium, respectively.

$$E_d^i = -n_i \frac{\epsilon_i - \epsilon}{(1 - n_i)\epsilon + n_i\epsilon_i} E, \quad i = 1, 2 \quad (1)$$

Dielectric properties. The depolarization field in phase i , E_d^i , due to a field E in the effective medium is given as

where ϵ_i and ϵ are the dielectric constants of component i and the composite, respectively, n_i is the geometric factor for phase i . n_i takes 0(prolate), 1/3(sphere), 1(oblate) and intermediate values for intermediate shapes. The electric field in phase i , E_i , is given by

$$E_i = E + E_d^i = \frac{\epsilon E}{(1 - n_i)\epsilon + n_i\epsilon_i} \quad (2)$$

Using the dielectric equations for the single components: $D_i = \epsilon_i E_i$ and the effective medium dielectric relations: $D = \epsilon E$, $D = \phi D_2 + (1 - \phi)D_1$ and $E = \phi E_2 + (1 - \phi)E_1$, where ϕ is the volume fraction of the ceramic inclusion (phase 2), the effective medium dielectric constant, ϵ , is given as a solution of the quadratic equation

$$\phi \frac{\epsilon}{(1 - n_2)\epsilon + n_2\epsilon_2} + (1 - \phi) \frac{\epsilon}{(1 - n_1)\epsilon + n_1\epsilon_1} = 1 \quad (3)$$

Elastic properties. The effective medium elastic properties are calculated by assuming the composite and the individual components to be incompressible. In addition their elastic properties are assumed to be one dimensional and similar to their analogous dielectric properties. This allows the use of similar equations for similar elastic and dielectric properties: $\epsilon \rightarrow C$ (elastic constant), $E \rightarrow S$ (strain) and $D \rightarrow T$ (stress). This leads to the strain analogue of the depolarization field, which we shall denote by S_d^i , and is given by

$$S_d^i = -4n_i \frac{(C_i - C)}{3(1-n_i)C + 4n_i C_i} S \quad i = 1, 2 \quad (4)$$

where C_i and C are the elastic constants of component i and the composite, respectively, n_i is the geometric factor for phase i . The strain in phase i , S_i , is given by

$$S_i = S + S_d^i = \frac{(3+n_i)}{4n_i C_i + 3(1-n_i)C} C S \quad (5)$$

Using the expressions for the elastic properties of the single components: $T_i = C_i S_i$, and the effective medium elastic relations: $T = CS$, $T = \phi T_2 + (1-\phi)T_1$ and $S = \phi S_2 + (1-\phi)S_1$ the effective medium elastic constant, C , is given as a solution of the quadratic equation

$$\phi \frac{(3+n_2)C}{3(1-n_2)C + 4n_2 C_2} + (1-\phi) \frac{(3+n_1)C}{3(1-n_1)C + 4n_1 C_1} = 1 \quad (6)$$

Piezoelectric polarization. When a stress T or strain S is applied to a composite system of discrete nonpiezoelectric phase (phase 1) and a piezoelectric phase (phase 2), a piezoelectric polarization P_2 is induced by the stress T or strain S in the latter which in turn produces an electric displacement D and an electric field E in the system. The observed polarization of the system caused by P_2 is a function of the dielectric constants (ϵ_1, ϵ_2) and elastic constants (C_1, C_2) of the two phases. The cross coupling effect between the second-rank tensor elastic variables (T stress, S strain) and the simple vector dielectric variables (D electric displacement, E electric field) define four third-rank tensor piezoelectric constants for the composite: d, e, g and h , and are given by [12,22]

$$\begin{aligned} d &= \left(\frac{D}{T}\right)_E = \left(\frac{S}{E}\right)_T, & e &= \left(\frac{D}{S}\right)_E = -\left(\frac{T}{E}\right)_S \\ g &= \left(\frac{E}{T}\right)_D = \left(\frac{S}{D}\right)_T, & h &= -\left(\frac{E}{S}\right)_D = -\left(\frac{T}{D}\right)_S \end{aligned} \quad (7)$$

When one or both the components of a composite are piezoelectric, we can observe the piezoelectric effect as a gross property of composites. In order to relate the gross piezoelectric constants to that of the components we assume that the components constants are one dimensional and relate to the dielectric variables along one polar axis, and the elastic variables along the direction perpendicular (or parallel) to the polar axis. The dielectric constants are approximated to be independent of mechanical states and the elastic constants are approximated to be independent of elastic states. The composite piezoelectric coefficients are calculated from the corresponding

values of the components as

$$\begin{aligned}
 d &= \sum_{i=1}^2 \phi_i L_{E_i} L_{T_i} d_i, & e &= \sum_{i=1}^2 \phi_i L_{S_i} L_{E_i} e_i \\
 g &= \sum_{i=1}^2 \phi_i L_{D_i} L_{T_i} g_i, & h &= \sum_{i=1}^2 \phi_i L_{S_i} L_{D_i} h_i
 \end{aligned}
 \tag{8}$$

where d_i, e_i, g_i and h_i are piezoelectric coefficients of the individual components and the various L 's are local field coefficients defined as follows

$$L_{E_i} = \frac{E_i}{E}, \quad L_{D_i} = \frac{D_i}{D}, \quad L_{T_i} = \frac{T_i}{T}, \quad L_{S_i} = \frac{S_i}{S}
 \tag{9}$$

and have the explicit expressions

$$\begin{aligned}
 L_{E_1} &= \frac{\alpha_2}{\phi\alpha_1 + (1-\phi)\alpha_2}, & L_{E_2} &= \frac{\alpha_1}{\phi\alpha_1 + (1-\phi)\alpha_2} \\
 L_{D_1} &= \frac{\alpha_2\epsilon_1}{\phi\alpha_1\epsilon_2 + (1-\phi)\alpha_2\epsilon_1}, & L_{D_2} &= \frac{\alpha_1\epsilon_2}{\phi\alpha_1\epsilon_2 + (1-\phi)\alpha_2\epsilon_1},
 \end{aligned}
 \tag{10}$$

where $\alpha_1 = (1-n_1)\epsilon + n_1\epsilon_1$, $\alpha_2 = (1-n_2)\epsilon + n_2\epsilon_2$ and

$$\begin{aligned}
 L_{T_1} &= \frac{\beta_2 C_1}{\phi\beta_1 C_2 + (1-\phi)\beta_2 C_1}, & L_{T_2} &= \frac{\beta_1 C_2}{\phi\beta_1 C_2 + (1-\phi)\beta_2 C_1} \\
 L_{S_1} &= \frac{\beta_2}{\phi\beta_1 + (1-\phi)\beta_2}, & L_{S_2} &= \frac{\beta_1}{\phi\beta_1 + (1-\phi)\beta_2},
 \end{aligned}
 \tag{11}$$

where $\beta_1 = (3+n_2)[3(1-n_1)C + 4n_1C_1]$, $\beta_2 = (3+n_1)[3(1-n_2)C + 4n_2C_2]$.

For comparison we shall give the expressions for dielectric constant, elastic constant, and local field coefficients derived via spherical particle dispersion theory as follows.

$$\epsilon = \frac{2\epsilon_1 + \epsilon_2 - 2\phi(\epsilon_1 - \epsilon_2)}{2\epsilon_1 + \epsilon_2 + \phi(\epsilon_1 - \epsilon_2)} \epsilon_1, \quad C = \frac{3C_1 + C_2 - 2\phi(C_1 - C_2)}{2C_1 + C_2 + \phi(C_1 - C_2)} C_1
 \tag{12}$$

and

$$\begin{aligned}
 L_{E_1} &= \frac{2\epsilon_1 + \epsilon_2}{2\epsilon_1 + \epsilon_2 + \phi(\epsilon_1 - \epsilon_2)}, & L_{E_2} &= \frac{3\epsilon_1}{2\epsilon_1 + \epsilon_2 + \phi(\epsilon_1 - \epsilon_2)}, & L_{D_1} &= \frac{2\epsilon_1 + \epsilon_2}{2\epsilon_1 + \epsilon_2 - 2\phi(\epsilon_1 - \epsilon_2)}, \\
 L_{D_2} &= \frac{3\epsilon_2}{2\epsilon_1 + \epsilon_2 - 2\phi(\epsilon_1 - \epsilon_2)}, & L_{T_1} &= \frac{3C_1 + 2C_2}{3C_1 + 2C_2 - 3\phi(C_1 - C_2)}, & L_{T_2} &= \frac{5C_2}{3C_1 + 2C_2 - 3\phi(C_1 - C_2)} \quad (13) \\
 L_{S_1} &= \frac{3C_1 + 2C_2}{3C_1 + 2C_2 - 2\phi(C_1 - C_2)}, & L_{S_2} &= \frac{5C_1}{3C_1 + 2C_2 + 2\phi(C_1 - C_2)}
 \end{aligned}$$

RESULTS AND DISCUSSION

We have proposed an effective medium theory of ferroelectric ceramic-polymer composite materials which can treat both components symmetrically, and have demonstrated the role played by the microgeometry of inclusions on dielectric, mechanical and piezoelectric properties of 0-3 type composites. The limits of the dielectric, elastic and the local field coefficients used to predict the four piezoelectric coefficients of the effective medium theory as a function of the geometric parameters is determined and presented in Table 1. The predictions reduce to that of a parallel

Table 1. Limits of the effective medium predictions as a function of the effective geometric parameters n_1 and n_2 .

Parallel: $n_1 = n_2 = 0$	Series: $n_1 = n_2 = 1$
$\epsilon = \phi\epsilon_2 + (1-\phi)\epsilon_1, \quad C = \phi C_2 + (1-\phi)C_1$	$\frac{1}{\epsilon} = \frac{\phi}{\epsilon_2} + \frac{(1-\phi)}{\epsilon_1}, \quad \frac{1}{C} = \frac{\phi}{C_2} + \frac{(1-\phi)}{C_1}$
$L_{E_1} = 1, \quad L_{E_2} = 1$	$L_{E_1} = \frac{\epsilon_1}{\phi\epsilon_2 + (1-\phi)\epsilon_1}, \quad L_{E_2} = \frac{\epsilon_2}{\phi\epsilon_2 + (1-\phi)\epsilon_1}$
$L_{D_1} = \frac{\epsilon_1}{\phi\epsilon_2 + (1-\phi)\epsilon_1}, \quad L_{D_2} = \frac{\epsilon_2}{\phi\epsilon_2 + (1-\phi)\epsilon_1}$	$L_{D_1} = 1, \quad L_{D_2} = 1$
$L_{T_1} = \frac{C_1}{\phi C_2 + (1-\phi)C_1}, \quad L_{T_2} = \frac{C_2}{\phi C_2 + (1-\phi)C_1}$	$L_{T_1} = 1, \quad L_{T_2} = 1$
$L_{S_1} = 1, \quad L_{S_2} = 1$	$L_{S_1} = \frac{C_2}{\phi C_2 + (1-\phi)C_1}, \quad L_{S_2} = \frac{C_1}{\phi C_2 + (1-\phi)C_1}$

model when n_1 and n_2 go to 0, a series model when n_1 and n_2 go to 1, a pure component 1 when n_1 goes to 1 and n_2 goes to 0 and a pure component 2 when n_1 goes to 0 and n_2 goes to 1. It appears that the geometric parameters, n_1 and n_2 , are effective parameters which can take care of shape of inclusion and certain aspects of connectivity. For this particular circumstance the effect of shape and connectivity on the mechanical, electrical and piezoelectric properties are

indistinguishable. In Figures 2-4 we present the predictions from the theory on the effect of microgeometry on the dielectric, elastic and dielectric constants of a composite.

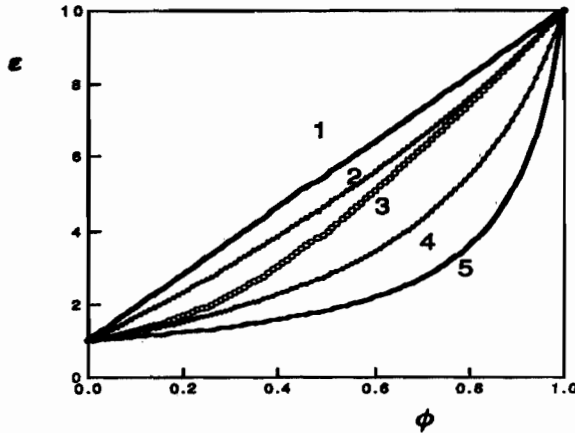


Figure 2. Effective medium theory predictions on the effect of inclusion microgeometry on the composite dielectric constant. 1(Parallel: $n_1 = 0, n_2 = 0$), 2(Spherical matrix: $n_1 = 1/3, n_2 = 0.05$), 3($n_1 = 1/3, n_2 = 1/3$), 4(Spherical inclusions: $n_1 = 0.83, n_2 = 1/3$), 5 (Series: $n_1 = 1, n_2 = 1$).

Figure 2 shows comparison of predictions of the effective medium theory via equation 3 on the dielectric constants of five different models as a function of the ceramic volume fraction, 1(Parallel: $n_1 = 0, n_2 = 0$), 2(Spherical matrix: $n_1 = 1/3, n_2 = 0.05$), 3($n_1 = 1/3, n_2 = 1/3$), 4(Spherical inclusions: $n_1 = 0.83, n_2 = 1/3$), 5(Series: $n_1 = 1, n_2 = 1$). The dielectric constants data for the two pure phases used in these calculations are given in Table 2.

Table 2. Parameters used in the theoretical calculations [18].

	ϵ	$C/(\text{GN m}^{-2})$	$d/(\text{pC N}^{-1})$
PZT	1850	6.32	184
PVDF	8.9	0.79	-

All the effective medium predictions are intermediate between the two extremes, parallel and series models. The shape effect is significant in the ceramic composition range of interest $\phi \sim 0.7$. We shall present next the predictions of the effective medium theory on effect of microgeometry on the elastic and piezoelectric properties of a composite at a constant ceramic composition of interest.

For a given ceramic volume fraction ϕ , the composite elastic constant is expected to depend on the microgeometry of the inclusions and the host. Equation 6 gives the composition and microgeometry dependence of the composite elastic constant predicted by the effective medium

theory. Figure 3 shows the geometric dependence of the composite elastic constant at a constant ceramic composition of $\phi = 0.67$. The elastic constants data for the two pure phases used in these calculations are given in Table 2. The dielectric constants of the two pure phases differ by a factor of about 200, in contrast to the elastic constants of the two pure phases which differ by a factor of about 10. The relatively small difference in the magnitude of the elastic constants for the two pure phases compared to that of the dielectric constants appears to make the geometric dependence of the composite elastic constant relatively weak compared to that of the dielectric constant.

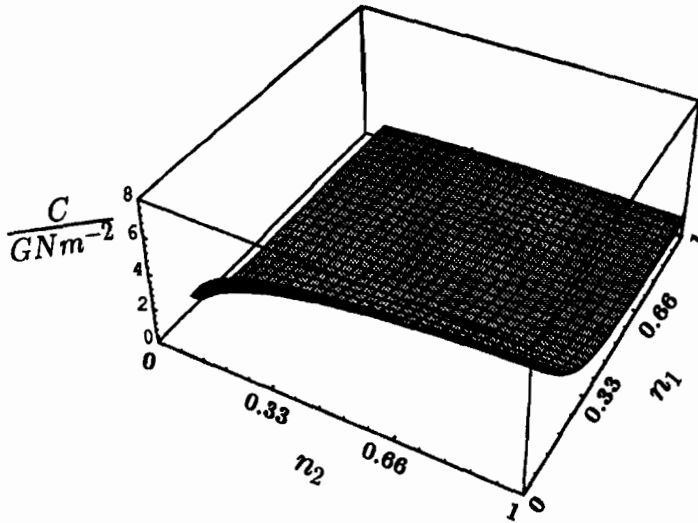


Figure 3. Effective medium predictions on the effect of inclusions microgeometry on the composite elastic constant for a ceramic composition of 0.67.

In the experimental determination of the longitudinal piezoelectric d-constant [18], the stress and the electric displacement are measured perpendicular to each other. Consequently we have used two separate sets of geometric parameters for the dielectric and elastic properties to define the piezoelectric d-constant. Figure 4 shows the piezoelectric d-constant of the composite predicted via equations 8, 10 and 11 as a function of n_1 of the dielectric constant and n_1 of the elastic constant when n_2 of the elastic constant and n_2 of the dielectric constant are kept fixed at 0.5. The piezoelectric d-constants for the two pure phases used in this prediction are given in Table 2 where the ceramic volume fraction was taken to be $\phi = 0.67$. The microgeometry in the dielectric properties is more significant in determining the d-constant than that of the elastic constant. In general, the microgeometry of the component with low dielectric, elastic and piezoelectric coefficients appear to play a more significant role in controlling the corresponding values in the composite.

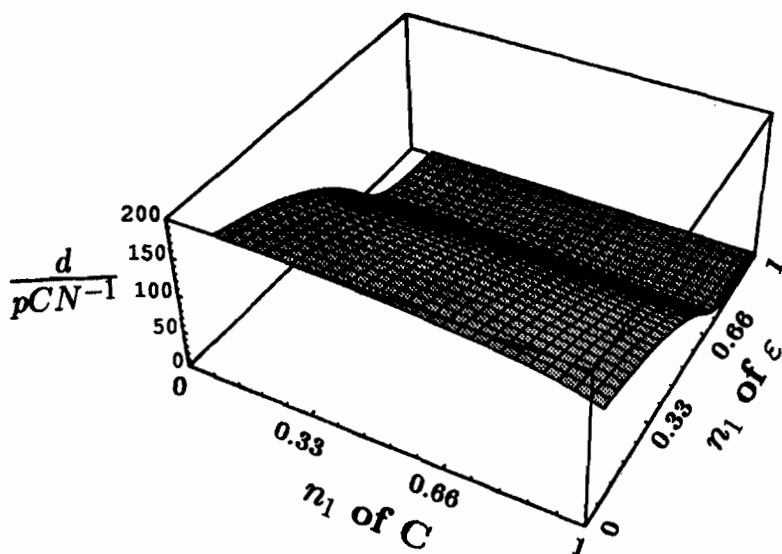


Figure 4. Effective medium predictions on the effect of inclusions microgeometry on the composite piezoelectric d -constant for a ceramic composition of 0.67.

Next, we present a comparison of theoretical predictions of the elastic, dielectric and piezoelectric coupling constants with the experimental data obtained from Reference 18 on a binary system consisting of polyvinylidene fluoride (PVDF) and lead zirconate titanate (PZT) powder at various compositions. The powder is expected to have, on the average a spherical shape and to be uniformly distributed in the polymer matrix. The single component coupling constants used in the theoretical calculations are given in Table 2. The values of the effective geometric parameters, n_1 and n_2 , which best fit the experimental data on the stress free dielectric constant are 0.60 and 0.20, respectively. Figure 5A compares predictions for the dielectric constant via equation 3 (solid line), equation 12 (dashed line) and experimental data [18] (open circles). The effective medium theory predictions are in good accord with the experimental data over all the composition range studied. For uniformly distributed spherical inclusions at low composition range, n_1 is expected to take values close to 1 and n_2 close to 0.33 (see Figure 2). Close to zero values of n_2 and values much lower than 1 of n_1 for ϵ implies that the PZT particles are connected across the electrodes. The specifications for the preparation of the sample describes how the composite film was prepared by pressing the film perpendicular to the film surface. The PZT sample, which was once distributed homogeneously, would make electrical contact in the normal direction of the surface of the film during the pressing process. The values of n_1 and n_2 that fit the experimental data are consistent with this explanation.

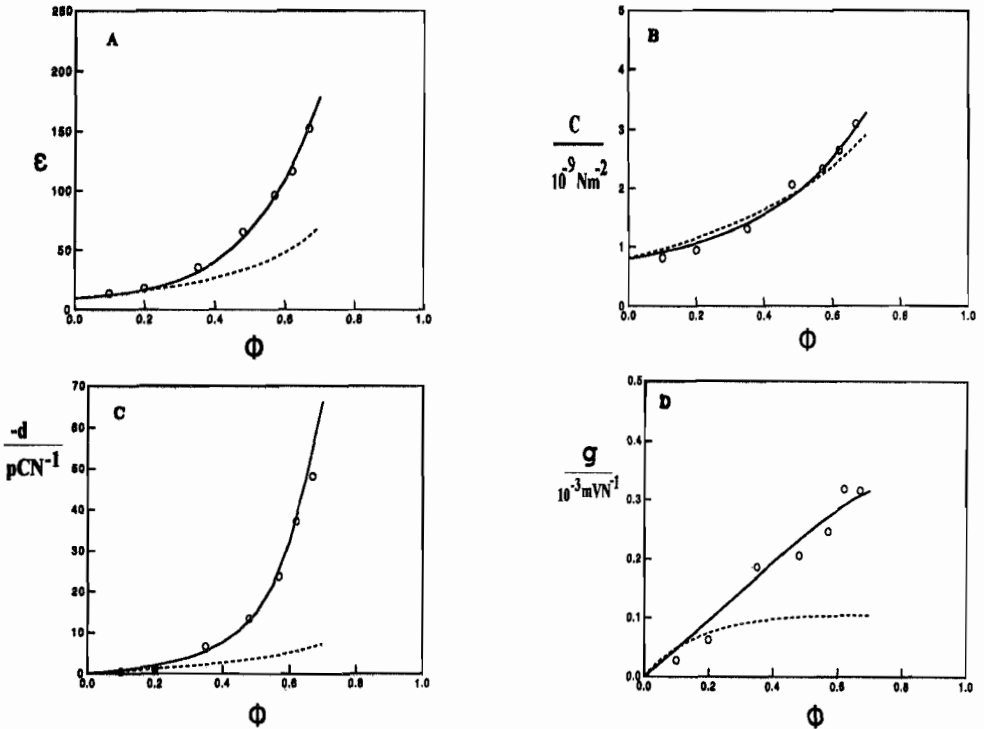


Figure 5. PZT volume fraction dependence of A) dielectric constant ϵ , B) elastic constant C , C) piezoelectric d-constant, and D) piezoelectric g-constant in a PVDF matrix composite. The effective medium theory predictions, the spherical particle dispersion theory predictions and the experimental data [18] are represented by solid line, dashed line and points, respectively.

The comparison of the predictions from theory and experimental data of the dependence of the elastic constant on a PZT volume fraction is shown in Figure 5B. The geometric parameters n_1 and n_2 evaluated via equation 6 (solid line) to fit the experimental data are 0.6 and 0.8, respectively. The values of the geometric parameters appear to be significantly different from those of the dielectric constant. This implies that the inclusions are elongated and perpendicular to the applied stress. This is consistent with the predictions for the inclusions geometry based on the dielectric constant. The dielectric constant is measured along the thickness direction and the elastic constant is measured along the length direction.

Considering the relationships among the four piezoelectric coupling constants: $e = dC$, $g = d/\epsilon$, and $h = e/\epsilon$, and the mechanical and dielectric local field coefficients: $L_T = (C_2/C)L_S$ and $L_D = (\epsilon_2/\epsilon)L_E$ we have used the geometric parameters determined to fit the dielectric constant data ($n_1 = 0.6$ and $n_2 = 0.2$) and elastic constant data ($n_1 = 0.6$ and $n_2 = 0.8$) to predict and

compare against the experimental data for the piezoelectric d- and g-coupling constants of the same composite [18]. Figures 5C and 5D compare predictions of the effective medium theory defined via equations 8, 10 and 11, the spherical particle dispersion theory defined via equations 8 and 13, and experimental results for the piezoelectric d- and g- coefficients, respectively. Overall the composition range studied from the experimental data is well predicted by the effective medium theory.

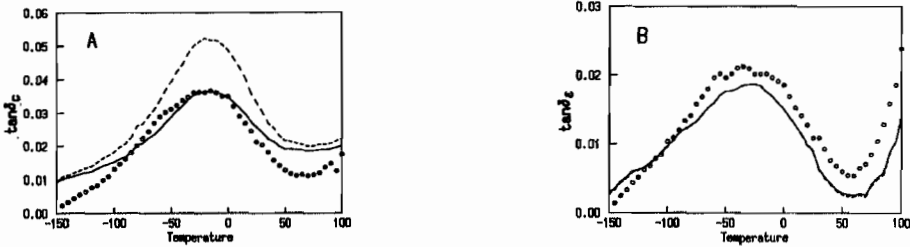


Figure 6. Comparison of predictions of effective medium theory (solid lines), spherical particle theory (dashed lines) with experimental results [5] (open circles) on the loss tangents of a 0.131 volume fraction PZT- epoxy resin composite in the local mode dispersion region of epoxy resin: A) $\tan \delta_c$ for $n_1 = 0.11$ and $n_2 = 0.33$, B) $\tan \delta_e$ for $n_1 = 0.26$ and $n_2 = 0.33$.

The elastic, dielectric and piezoelectric coupling constants of composites depend upon temperature and frequency of measurement [23]. The piezoelectric constants show relaxations where thermal relaxations in mechanical and dielectric properties take place. For sinusoidal excitations the relaxing dielectric and elastic constants are expressed as complex quantities in the form $\epsilon^* = \epsilon' - j\epsilon''$, $C^* = C' + jC''$, the relaxing complex piezoelectric constants are expressed as $d^* = d' - jd''$, $e^* = e' - je''$, $g^* = g' - jg''$, $h^* = h' - jh''$ and their loss tangent is defined in the form ($\tan \delta_x = x''/x'$ where x stands for any of the six complex quantities).

The effective medium equations developed are generally valid at any temperature if the corresponding values of the single component constants are used. Figure 6(A-B) compare predictions of theory with experimental results [11] on the loss tangents of elastic and dielectric constants of a 0.131 volume fraction PZT-epoxy resin composite in the local mode dispersion region of an epoxy resin. Values for the effective geometric parameters which fit the experimental data are $\epsilon(n_1 = 0.26$ and $n_2 = 0.33)$ and $C(n_1 = 0.11$ and $n_2 = 0.33)$. Both theories give good predictions on $\tan \delta_e$. However, they differ widely for the predictions of $\tan \delta_c$. The geometric parameters which fit $\tan \delta_e$ and $\tan \delta_c$, are also found to best fit the experimental data on $\tan \delta_d$, $\tan \delta_g$, $\tan \delta_e$ and $\tan \delta_h$ when used in the appropriate local field coefficients. The results for the loss tangents of the piezoelectric coupling constants are shown in Figure 7(A-D). The spherical

particle dispersion theory predictions differ widely from the experimental results. In particular the g -constant was predicted to be non-relaxing against the experimental results. These differences show the effect of the inclusions microgeometry on the relaxation properties of the various composite coupling constants.

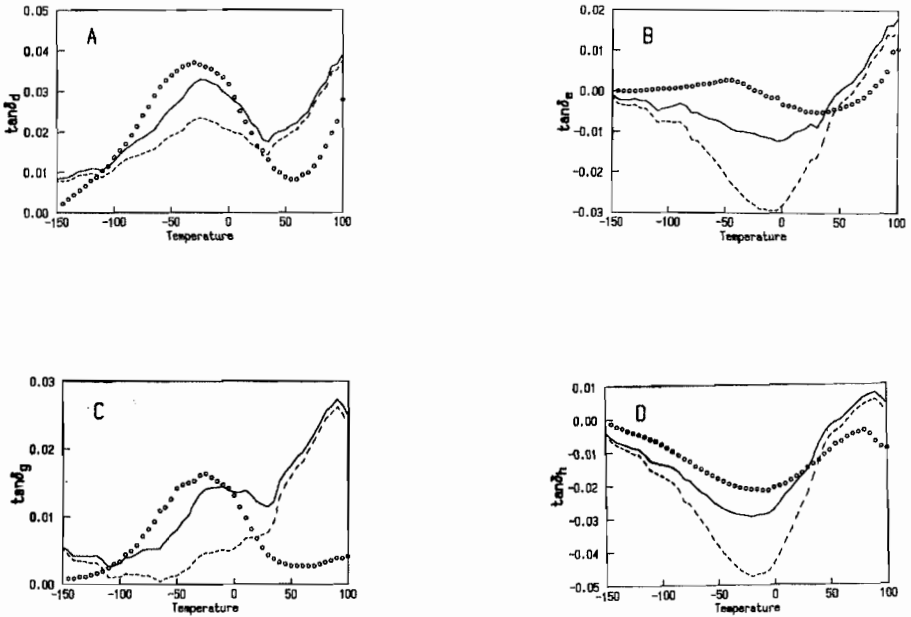


Figure 7. Comparison of predictions of effective medium theory (solid lines, spherical particle theory (dashed lines) with experimental results [11] (open circles) on the loss tangents: A) $\tan \delta_d$, B) $\tan \delta_g$, C) $\tan \delta_e$ and D) $\tan \delta_h$ for a 0.131 volume fraction PZT-epoxy resin composite. The geometric parameters used to fit the effective medium theory are $n_1 = 0.11$ and $n_2 = 0.33$ for the elastic constant and $n_1 = 0.26$ and $n_2 = 0.33$ for the dielectric constant.

CONCLUSIONS

In this work we have shown the strong influence of the geometric effect in determining the magnitude and relaxational behavior of electrical, mechanical and piezoelectric properties of ferroelectric ceramic-polymer composite materials by comparing the effective medium predictions against spherical particle dispersion theory and experimental data from various composite materials. We have observed that the geometric effect is more significant for the component with

low values for the dielectric, elastic and piezoelectric coupling constants. The effective medium theory could be used to interpret and understand experimental data from composite materials. It could also be used to predict dielectric and elastic properties from piezoelectric constant data and vice versa.

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