Dielectric response of spherically anisotropic graded piezoelectric composites

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(Received 18 April 2007; accepted 5 August 2007; published online 1 October 2007)

A graded piezoelectric composite consisting of a spherically anisotropic graded piezoelectric inclusion imbedded in an infinite nonpiezoelectric matrix, with the physical properties of the graded spherical inclusion having a power-law profile with respect to the radial variable r, is studied theoretically. Under an external uniform electric field, the electric displacement field and the elastic stress tensor field of this spherically anisotropic graded piezoelectric composite are derived exactly by means of displacement separation technique, based on the governing equations in the dilute limit. A piezoelectric response mechanism, in which the effective piezoelectric response vanishes along the z direction (or x, y directions), is revealed in this kind of graded piezoelectric composites. Furthermore, it is found that the effective dielectric constant decreases (or increases) with the volume fraction p of the inclusions if the exponent parameter k of the grading profile is larger (or smaller) than a critical value. © 2007 American Institute of Physics. [DOI: 10.1063/1.2785020]

I. INTRODUCTION

The effective property of piezoelectric composites has been extensively investigated by many authors using various methods¹⁻⁸ because they have many applications in ultrasonic transducers for ocean acoustics, biomedical imaging, and electronic sensors and monitors for naval and space navigations.^{9,10} In order to satisfy the needs of engineering design of suitable materials, scientists and engineers have studied and fabricated several functionally graded materials in the laboratory.¹¹ These materials usually have variations in their spatial composition, microstructure, or other microproperties so that they have physical properties varying from point to point within the composite materials. In fact, many natural materials have graded properties, such as the graded dielectric properties in living cells and in bamboo. Already, some researchers have investigated the electrical, thermal, and mechanical properties of some graded composites.¹²⁻¹⁸ However, there are few theoretical works on the effective coupling properties of graded piezoelectric composites as they usually only focused on homogenous piezoelectric composites.¹⁻⁶ Also, it is very difficult to derive exact solutions for the local displacement and electric fields, in general, for graded piezoelectric composites under arbitrary external electric and strain fields. However, since graded piezoelectric composites have the advantage of providing controllable effective piezoelectric properties in engineering applications, and before the fabrication of a special kind of piezoelectric composites for specific needs, engineers would like to be able to estimate its effective properties, it is valuable to study the effective dielectric and piezoelectric responses of these graded piezoelectric composites theoretically.

Many piezoelectric materials, for example, piezoceramics, exhibit transverse isotropy, with the unique axis aligned along their poling direction. However, if the ceramic spherical inclusion is poled along its radial direction, the composite will possess a special kind of transverse isotropy on the spherical surface, called spherically anisotropy (or called spherical isotropy, in some literatures).^{19–21} For these spherically anisotropic composites, there are relatively few works investigating their effective properties under different external fields. In particular, the effective property response problem of a spherically anisotropic graded piezoelectric composite is not yet solved although some authors have studied the free vibration and crack problems of some geometrically structural graded piezoelectric materials, such as graded spherical shell, graded cylindrical shell, and laminate plate.^{19,22–24} In this paper, as an example, we have investigated theoretically the effective responses of a graded piezoelectric composite, which comprises of a spherically anisotropic graded piezoelectric inclusion embedded in an infinite nonpiezoelectric matrix. The spherical inclusion is assumed to have a power-law dependence on the radial variable r, namely, $\varepsilon_{ii}(r) = \varepsilon_{ii}^0 r^k$, $e_{ij}(r) = e_{ij}^0 r^k$, and $c_{ij}(r) = c_{ij}^0 r^k$. Furthermore, the effects of the power-law parameter k on the effective coupling response under an external electric field will be discussed in details. One of our results is that a vanishing

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effective piezoelectric response mechanism is revealed in this kind of spherically anisotropic graded piezoelectric composite.

In Sec. II, analytical solutions of the elastic displacement and electric potentials are derived exactly for a spherically anisotropic graded piezoelectric inclusion in an isotropic nonpiezoelectric matrix under an external uniform electric field along the z direction. In Sec. III, formulas for calculating the effective dielectric and piezoelectric responses are given in the dilute limit, and numerical results are given for discussing the effects of the power-law parameter k on the effective dielectric properties. In Sec. IV, a brief conclusion is given.

II. ANALYTICAL SOLUTIONS

Consider an infinite isotropic nonpiezoelectric matrix containing a spherically anisotropic graded piezoelectric inclusion in spherical coordinates (θ, φ, r) , with the origin of the coordinate system located at the center of the spherical inclusion. The constitutive equations in the graded inclusion region Ω_i and the nonpiezoelectric host region Ω_h are, respectively,

$$\sigma_{ij}^{i} = c_{ijkl}^{i} \gamma_{kl}^{j} - e_{kij}^{i} E_{k}^{i}, \quad D_{i}^{i} = e_{ikl}^{i} \gamma_{kl}^{j} + \varepsilon_{ik}^{i} E_{k}^{i}, \quad \text{in } \Omega_{i}, \quad (1)$$

$$\sigma_{ij}^{h} = c_{ijkl}^{h} \gamma_{kl}^{h}, \quad D_{i}^{h} = \varepsilon_{ik}^{h} E_{k}^{h}, \quad \text{in } \Omega_{h}, \tag{2}$$

where the subscripts i, j, k, l=1, 2, 3 denote the θ, φ, r directions, respectively, and the superscripts i and h denote quantities of the inclusion and the host regions, respectively. The quantities σ , γ, D , and E are the stress, strain, electric displacement, and electric field, respectively, and c, e, and ε are the elastic stiffness, piezoelectric coefficient, and dielectric constant, respectively. If there is no body forces and no free electric charges, the governing equations in the inclusion and host regions are $\sigma_{ij,j}=0$ and $D_{i,i}=0$, and the boundary conditions at the two-phase interface are the continuity of elastic displacement, lectric potential, normal traction, and electric displacement. In spherical coordinates, the governing equations are ¹⁹

$$\frac{\partial \sigma_{rr}}{\partial r} + \frac{1}{r} \frac{\partial \sigma_{r\theta}}{\partial \theta} + \frac{1}{r \sin \theta} \frac{\partial \sigma_{\varphi r}}{\partial \varphi} + \frac{2\sigma_{rr} - \sigma_{\theta\theta} - \sigma_{\varphi\varphi} + \sigma_{r\theta} \cot \theta}{r} = 0,$$

$$\frac{\partial \sigma_{r\theta}}{\partial r} + \frac{1}{r} \frac{\partial \sigma_{\theta\theta}}{\partial \theta} + \frac{1}{r \sin \theta} \frac{\partial \sigma_{\theta\varphi}}{\partial \varphi} + \frac{(\sigma_{\theta\theta} - \sigma_{\varphi\varphi})\cot \theta + 3\sigma_{r\theta}}{r}$$
$$= 0,$$

$$\frac{1}{r^2}\frac{\partial}{\partial r}(r^2D_r) + \frac{1}{r\sin\theta}\frac{\partial(D_\theta\sin\theta)}{\partial\theta} + \frac{1}{r\sin\theta}\frac{\partial D_\varphi}{\partial\varphi} = 0.$$
 (3)

In general, for this spherically anisotropic graded piezoelectric material, the constitutive relations in the inclusion region in spherical coordinates are as follows:¹⁹

$$\sigma_{\theta\theta} = c_{11}\gamma_{\theta\theta} + c_{12}\gamma_{\varphi\varphi} + c_{13}\gamma_{rr} - e_{31}E_r,$$

$$\sigma_{\varphi\varphi} = c_{12}\gamma_{\theta\theta} + c_{11}\gamma_{\varphi\varphi} + c_{13}\gamma_{rr} - e_{31}E_r,$$

$$\sigma_{rr} = c_{13}\gamma_{\theta\theta} + c_{13}\gamma_{\varphi\varphi} + c_{33}\gamma_{rr} - e_{33}E_r,$$

$$\sigma_{\varphi r} = 2c_{44}\gamma_{\varphi r} - e_{15}E_{\varphi}, \quad \sigma_{r\theta} = 2c_{44}\gamma_{r\theta} - e_{15}E_{\theta},$$

$$\sigma_{\theta\varphi} = (c_{11} - c_{12})\gamma_{\varphi\theta}, \quad D_{\theta} = 2e_{15}\gamma_{r\theta} + \varepsilon_{11}E_{\theta},$$

$$D_{\varphi} = 2e_{15}\gamma_{\varphi r} + \varepsilon_{11}E_{\varphi},$$

$$D_r = e_{31}\gamma_{\theta\theta} + e_{31}\gamma_{\varphi\varphi} + e_{33}\gamma_{rr} + \varepsilon_{33}E_r,$$
(4)

where we have omitted the superscript *i* (inclusion) from above equations for convenience. The subscripts *i*, *j* in the coefficients c_{ij} and e_{ij} in Eq. (4) are the replacement indices according to Nye's rule.¹⁹ Because the matrix is an isotropic elastic and dielectric material, the constitutive relations of host region can be obtained from Eq. (4) by omitting the piezoelectric coefficients e_{ij} and letting $c_{ijkl}=\lambda \delta_{ij}\delta_{kl}$ $+\mu(\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk})$ and $\varepsilon_{11}=\varepsilon_{22}=\varepsilon_{33}$, where λ and μ are the Lame constants and δ_{ij} is the Kronecker delta. The strain tensor γ can be expressed in terms of the elastic displacements u_r , u_{θ} , and $u_{\varphi}^{(1)}$,

$$\begin{split} \gamma_{rr} &= \partial u_r / \partial r, \quad \gamma_{\theta\theta} = r^{-1} (\partial u_{\theta} / \partial \theta + u_r), \\ \gamma_{\varphi\varphi} &= r^{-1} (u_r + u_{\theta} \cot \theta + \sin \theta^{-1} \partial u_{\varphi} / \partial \varphi), \\ \gamma_{r\theta} &= (r^{-1} \partial u_r / \partial \theta + \partial u_{\theta} / \partial r - r^{-1} u_{\theta}) / 2, \\ \gamma_{\theta\varphi} &= r^{-1} (\sin \theta^{-1} \partial u_{\theta} / \partial \varphi + \partial u_{\varphi} / \partial \theta - u_{\varphi} \cot \theta) / 2, \\ \gamma_{\varphi r} &= (\partial u_{\varphi} / \partial r - r^{-1} u_{\varphi} + r^{-1} \sin \theta^{-1} \partial u_r / \partial \varphi) / 2. \end{split}$$
(5)

In the following, the analytical solutions of the elastic displacement *u* and the electric potential Φ in this graded piezoelectric composite under an external uniform electric field E_0 along the \hat{z} directions (or \hat{x} direction) will be derived by displacement separation method. We assume that the physical properties of a spherically anisotropic graded piezoelectric inclusion with radius *a* obey the same power law along the radial direction: $\varepsilon_{ij} = \varepsilon_{ij}^0 r^k$, $e_{ij} = e_{ij}^0 r^k$, and $c_{ij} = c_{ij}^0 r^k$. Here ε_{ij}^0 , e_{ij}^0 , and c_{ij}^0 are constants. In order to solve the governing equations $\sigma_{ij,j}=0$ and $D_{i,i}=0$ in the inclusion region, we introduce three unknown functions $\psi(\theta, \varphi, r), G(\theta, \varphi, r)$, and $W(\theta, \varphi, r)$ for the displacement components, ^{20,22,25}

$$u_{\theta} = -\frac{1}{\sin\theta} \frac{\partial\psi}{\partial\varphi} - \frac{\partial G}{\partial\theta}, \quad u_{\varphi} = \frac{\partial\psi}{\partial\theta} - \frac{1}{\sin\theta} \frac{\partial G}{\partial\varphi}, \quad u_{r} = W.$$
(6)

Substituting Eq. (6) into Eq. (4), it is easy to get four partial differential equations governing $\psi(\theta, \varphi, r), G(\theta, \varphi, r),$

 $W(\theta, \varphi, r)$, and hence the electric potential unknown function $\Phi(\theta, \varphi, r)$. Furthermore, we assume that general solutions of these four unknown functions ψ , *G*, *W*, and Φ take the following forms:

$$\psi = \sum_{n=1}^{\infty} \psi_n(r) Y_n^m(\theta, \varphi), \quad G = \sum_{n=1}^{\infty} G_n(r) Y_n^m(\theta, \varphi),$$
$$W = \sum_{n=1}^{\infty} W_n(r) Y_n^m(\theta, \varphi), \quad \Phi = \frac{e_{33}^0}{\varepsilon_{33}^0} \sum_{n=1}^{\infty} \Phi_n(r) Y_n^m(\theta, \varphi), \quad (7)$$

where $Y_n^m(\theta, \varphi)$ are the spherical harmonics and $\psi_n(r)$, $G_n(r)$, $W_n(r)$, and $\Phi_n(r)$ are unknown functions. Substituting Eqs. (7) and (6) into Eq. (3), we get a set of linear differential equations for the unknown functions $\psi_n(r)$, $G_n(r)$, $W_n(r)$, and $\Phi_n(r)$,

$$-f_1W_n - f_2r\frac{\partial W_n}{\partial r} - r^2\frac{\partial^2 W_n}{\partial r^2} + f_3r\frac{\partial \Phi_n}{\partial r} + r^2\frac{\partial^2 \Phi_n}{\partial r^2} - f_4\Phi_n$$
$$-f_5\frac{r\partial G_n}{\partial r} - f_6G_n = 0,$$
$$-f_7\psi_n + f_3r\frac{\partial \psi_n}{\partial r} + r^2\frac{\partial^2 \psi_n}{\partial r^2} = 0,$$
$$-f_8G_n + f_3r\frac{\partial G_n}{\partial r} + r^2\frac{\partial^2 G_n}{\partial r^2} - f_9r\frac{\partial W_n}{\partial r} - f_{10}W_n - f_{11}r\frac{\partial \Phi_n}{\partial r}$$
$$-f_{12}\Phi_n = 0,$$
$$f_{13}r\frac{\partial \Phi_n}{\partial r} + f_{14}r^2\frac{\partial^2 \Phi_n}{\partial r^2} - f_{15}\Phi_n + f_{16}W_n + f_3r\frac{\partial W_n}{\partial r}$$

$$+ r^{2} \frac{\partial^{2} W_{n}}{\partial r^{2}} + f_{17} G_{n} + f_{18} r \frac{\partial G_{n}}{\partial r} = 0, \qquad (8)$$

where

$$\begin{split} f_1 &= [2(k+1)e_{31}^0 - n(n+1)e_{15}^0]/e_{33}^0, \\ f_2 &= (k+2) + 2e_{31}^0/e_{33}^0, \\ f_3 &= (k+2), \quad f_4 = n(n+1)\frac{\varepsilon_{11}^0}{\varepsilon_{33}^0}, \\ f_5 &= n(n+1)(e_{31}^0 + e_{15}^0)/e_{33}^0, \\ f_6 &= n(n+1)[(k+1)e_{31}^0 - e_{15}^0]/e_{33}^0, \\ f_7 &= \left[k+2+\frac{1}{2}(c_{11}^0/c_{44}^0 - c_{12}^0/c_{44}^0)(n^2+n-2)\right], \\ f_8 &= [n(n+1)c_{11}^0/c_{44}^0 + k+2 - c_{11}^0/c_{44}^0 + c_{12}^0/c_{44}^0], \\ f_9 &= (c_{13}^0/c_{44}^0 + 1), \\ f_{10} &= (k+2+c_{12}^0/c_{44}^0 + c_{11}^0/c_{44}^0), \end{split}$$

$$\begin{split} f_{11} &= (1 + e_{31}^{0}/e_{15}^{0}) \frac{e_{33}^{0}e_{15}^{0}}{\varepsilon_{33}^{0}c_{44}^{0}}, \quad f_{12} &= (k+2) \frac{e_{33}^{0}e_{15}^{0}}{\varepsilon_{33}^{0}c_{44}^{0}}, \\ f_{13} &= (2 + k - 2e_{31}^{0}/e_{33}^{0}) \frac{e_{33}^{0}e_{33}^{0}}{\varepsilon_{33}^{0}c_{33}^{0}}, \\ f_{14} &= \frac{e_{33}^{0}e_{33}^{0}}{\varepsilon_{33}^{0}c_{33}^{0}}, \quad f_{15} &= n(n+1) \frac{e_{33}^{0}e_{15}^{0}}{\varepsilon_{33}^{0}c_{33}^{0}}, \\ f_{16} &= [2(c_{13}^{0} + kc_{13}^{0} - c_{11}^{0} - c_{12}^{0}) - n(n+1)c_{44}^{0}]/c_{33}^{0}, \\ f_{17} &= n(n+1)(kc_{13}^{0} + c_{13}^{0} - c_{11}^{0} - c_{12}^{0} - c_{44}^{0})/c_{33}^{0}, \\ f_{18} &= n(n+1)(c_{44}^{0} + c_{13}^{0})/c_{33}^{0}. \end{split}$$

Now, we assume that the unknown functions $\psi_n(r)$, $G_n(r)$, $W_n(r)$, and $\Phi_n(r)$ can be expressed in the following forms:

$$G_{n}(r) = A_{n}r^{\nu_{n}(k)}, \quad W_{n}(r) = B_{n}r^{\nu_{n}(k)},$$
$$\Phi_{n}(r) = C_{n}r^{\nu_{n}(k)}, \quad \psi_{n}(r) = D_{n}r^{\lambda_{n}(k)}, \quad (9)$$

where A_n , B_n , C_n , D_n , and $\nu_n(k)$ are undetermined constants. Substituting Eq. (9) into Eq. (8), we obtained a set of linear equations,

$$F_{ii}X_i = 0 \tag{10}$$

where $X_j = (A_n, B_n, C_n)^T$, $F_{11} = -f_5 \nu_n - f_6$, $F_{12} = -f_2 \nu_n - f_1$ $-\nu_n(\nu_n - 1)$, $F_{13} = f_3 \nu_n - f_4 + \nu_n(\nu_n - 1)$, $F_{21} = f_3 \nu_n - f_8 + \nu_n(\nu_n - 1)$, $F_{22} = -f_9 \nu_n - f_{10}$, $F_{23} = -f_{11} \nu_n - f_{12}$, $F_{31} = f_{18} \nu_n + f_{17}$, $F_{32} = f_3 \nu_n + f_{16} + \nu_n(\nu_n - 1)$, and $F_{33} = f_{13} \nu_n - f_{15} + f_{14} \nu_n(\nu_n - 1)$.

For Eq. (10) to have nonzero solutions, the eigenvalues $\nu_n(k)$ should be determined from the eigenequation $|F_{ij}|=0$. Also, for a stable graded material, the eigenvalues $\nu_n(k)$ should have non-negative real part solutions. This means that valid solutions of $\nu_n(k)$ inside the spherical inclusion, denoted as $\nu_{ni}(k)$, must have $\operatorname{Re}(\nu_{ni}) \ge 0$ (i=1,2,3). The following relations between A_n , B_n , and C_n are obtained from Eq. (10):

$$B_{ni} = K_{ni}^1 A_{ni}, \quad C_{ni} = K_{ni}^2 A_{ni} \quad (i = 1, 2, 3),$$
 (11)

where the matrices K_{ni}^1 and K_{ni}^2 are obtained from Eq. (10). It is noted that eigenvalue λ_n can be determined from the equation $[-f_7+f_3\lambda_n+\lambda_n(\lambda_n-1)]D_n=0$ for nonzero D_n . However, with the boundary conditions, we can demonstrate that the eigenvalue λ_n and the coefficient D_n should have no effects on the stress and the electric fields in the composite under an external field, i.e., the coefficients D_n should be zero. Thus, the unknown functions $G_n(r)$, $W_n(r)$, and $\Phi_n(r)$ can be rewritten in the following forms with the unknown coefficients A_{ni} (*i*=1,2,3):

$$G_n(r) = \sum_{i=1}^3 A_{ni} r^{\nu_{ni}(k)}, \quad W_n(r) = \sum_{i=1}^3 K_{ni}^1 A_{ni} r^{\nu_{ni}(k)},$$
$$\Phi_n(r) = \sum_{i=1}^3 K_{ni}^2 A_{ni} r^{\nu_{ni}(k)}, \quad \psi_n(r) = D_n r^{\lambda_{n1}(k)}.$$

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If an external electric field E_0 is applied along the \hat{z} direction, the solution should be independent of the variable φ because of the axial symmetry of the spherical inclusion. In the inclusion region, the displacement and the electric potentials can be expressed in terms of the Legendre polynomials $P_n(\cos \theta)$ as follows:

$$\Phi = \frac{e_{33}^0}{\varepsilon_{33}^0} \sum_{n=1}^{\infty} \sum_{i=1}^3 K_{ni}^2 A_{ni} r^{\nu_{ni}(k)} P_n(\cos \theta),$$

$$u_{\theta} = -\sum_{n=1}^{\infty} \sum_{i=1}^3 A_{ni} r^{\nu_{ni}(k)} \frac{\partial}{\partial \theta} P_n(\cos \theta),$$

$$u_{\varphi} = \sum_{n=1}^{\infty} D_n r^{\lambda_{n1}(k)} \frac{\partial}{\partial \theta} P_n(\cos \theta),$$

$$u_r = \sum_{n=1}^{\infty} \sum_{i=1}^3 K_{ni}^1 A_{ni} r^{\nu_{ni}(k)} P_n(\cos \theta).$$
 (12)

In the host region, because the matrix is a pure elastic and dielectric isotropic material, the nonzero elastic displacement u^h and the electric potential Φ^h under an external electric field along \hat{z} direction can be found by using Goodier's results,²⁶

$$u_{r}^{h} = [-2H_{1}/r^{3} + (\alpha_{-1} - 2)H_{2}/r]\cos\theta,$$

$$u_{\theta}^{h} = -(H_{1}/r^{3} + H_{2}/r)\sin\theta,$$

$$\Phi^{h} = (-E_{0}r + H_{3}/r^{2})\cos\theta,$$
 (13)

where $\alpha_{-1} = (10-12\nu)/(3-4\nu)$, $\nu = \lambda^h/2(\mu^h + \lambda^h)$. λ^h and μ^h are Lame constants of the host material. The unknown coefficients H_i (*i*=1,2,3) can be determined by the boundary conditions. Now, we apply the boundary conditions at the two-phase interface to solve for the unknown coefficients A_{ni} and H_i (*i*=1,2,3). The continuity boundary conditions for a spherical inclusion with radius *a* are as follows:

$$\begin{split} u_r^i(r) &= u_r^h(r)|_{r=a}, \qquad u_{\theta}^i(r) = u_{\theta}^h(r)|_{r=a}, \\ u_{\varphi}^i(r) &= u_{\varphi}^h(r)|_{r=a}, \qquad \sigma_{rr}^i(r) = \sigma_{rr}^h(r)|_{r=a}, \\ \sigma_{r\theta}^i(r) &= \sigma_{r\theta}^h(r)|_{r=a}, \qquad \sigma_{r\varphi}^i(r) = \sigma_{r\varphi}^h(r)|_{r=a}, \\ \Phi^i(r) &= \Phi^h(r)|_{r=a}, \qquad D_r^i(r) = D_r^h(r)|_{r=a}. \end{split}$$

Here, note that the two boundary conditions about the φ direction $u_{\varphi}^{i}(r) = u_{\varphi}^{h}(r)|_{r=a}$ and $\sigma_{r\varphi}^{i}(r) = \sigma_{r\varphi}^{h}(r)|_{r=a}$ are satisfied automatically. A set of closed-form equations for the determination of the six unknown coefficients A_{ni} and H_i (i = 1, 2, 3) can be found,

$$\sum_{i=1}^{3} K_{1i}^{1} a^{\nu_{1i}(k)} A_{1i} + 2a^{-3}H_1 + (2 - \alpha_{-1})H_2/a = 0,$$

$$\sum_{i=1}^{3} a^{\nu_{1i}(k)} A_{1i} + a^{-3}H_1 + a^{-1}H_2 = 0,$$

$$\sum_{i=1}^{3} \frac{e_{33}^{0}}{\varepsilon_{33}^{0}} K_{1i}^{2} a^{\nu_{1i}(k)} A_{1i} - a^{-2} H_{3} = -E_{0}a,$$

$$\sum_{i=1}^{3} m_{1i} a^{\nu_{1i}(k)} A_{1i} - h_{0} a^{-k-1} H_{2} - 12 \mu^{h} a^{-k-3} H_{1} = 0,$$

$$\sum_{i=1}^{3} m_{2i} a^{\nu_{1i}(k)} A_{1i} + 6 \mu^{h} a^{-k-3} H_{1} + h_{1} a^{-k-1} H_{2} = 0,$$

$$\sum_{i=1}^{3} m_{3i} a^{\nu_{1i}(k)} A_{1i} - 2\varepsilon^{h} a^{-k-2} H_{3} = a\varepsilon^{h} E_{0},$$
(14)

where

$$\begin{split} m_{1i} &= \left[\left(c_{33}^{0} K_{1i}^{1} + e_{33}^{0} \frac{e_{33}^{0}}{\varepsilon_{33}^{0}} K_{1i}^{2} \right) \nu_{1i} + 2c_{13}^{0} (1 + K_{1i}^{1}) \right], \\ m_{2i} &= \left(c_{44}^{0} K_{1i}^{1} + c_{44}^{0} + e_{15}^{0} \frac{e_{33}^{0}}{\varepsilon_{33}^{0}} K_{1i}^{2} - c_{44}^{0} \nu_{1i} \right), \\ m_{3i} &= \left[2e_{31}^{0} (1 + K_{1i}^{1}) + e_{33}^{0} (K_{1i}^{1} - K_{1i}^{2}) \nu_{1i} \right], \\ h_{0} &= 2\mu^{h} \left(\frac{2 - 8\nu^{h}}{1 - 2\nu^{h}} + \frac{-1 + 3\nu^{h}}{1 - 2\nu^{h}} \alpha_{-1} \right), \\ h_{1} &= \mu^{h} (4 - \alpha_{-1}). \end{split}$$

Once the unknown coefficients A_{1i} and H_i are obtained from Eq. (14), the elastic displacement and the electric potentials in the spherical inclusion region and the host region can be determined electric field. For example, in the inclusion region, we have

$$\Phi = \frac{e_{33}^0}{\varepsilon_{33}^0} \sum_{i=1}^3 K_{1i}^2 A_{1i} r^{\nu_{1i}(k)} \cos \theta, \quad u_\theta = \sum_{i=1}^3 A_{1i} r^{\nu_{1i}(k)} \sin \theta,$$
$$u_\varphi = 0, \quad u_r = \sum_{i=1}^3 K_{1i}^1 A_{1i} r^{\nu_{1i}(k)} \cos \theta. \tag{15}$$

III. EFFECTIVE RESPONSE

The strain tensor field and the electric field in the inclusion region can be derived by means of the solutions of the elastic displacement and the electric potentials obtained in Sec. II, for an external electric field along the \hat{z} direction. The effective dielectric and piezoelectric responses ε^e and e^e can be estimated by using the effective constitutive relations, defined by $\bar{\sigma}_{ij} = c^e_{ijkl} \bar{\gamma}_{kl} - e^e_{kij} \bar{E}_k$ and $\bar{D}_i = e^e_{ikl} \bar{\gamma}_{kl} + \varepsilon^e_{ik} \bar{E}_k$, where $\bar{A} = (1/V) \int_{\Omega_i + \Omega_h} A dV$ and V is the total volume occupied by the inclusion Ω_i and the host Ω_h regions,

$$\frac{1}{V} \int_{\Omega_i} \left[(c^i_{ijkl} - c^h_{ijkl}) \gamma_{kl} - (e^i_{kij} - e^h_{kij}) E_k \right] dV$$
$$= \bar{\sigma}_{ij} - c^h_{ijkl} \bar{\gamma}_{kl} + e^h_{kij} \bar{E}_k, \tag{16}$$

$$\frac{1}{V} \int_{\Omega_i} \left[(e^i_{ikl} - e^h_{ikl}) \gamma_{kl} + (\varepsilon^i_{ik} - \varepsilon^h_{ik}) E_k \right] dV$$
$$= \overline{D}_i - e^h_{ikl} \overline{\gamma}_{kl} - \varepsilon^h_{ik} \overline{E}_k.$$
(17)

Furthermore, we have obtained the following formulas by substituting the effective constitutive relations into Eqs. (16) and (17):

$$e^{e}_{ikl}\overline{\gamma}_{kl} + \varepsilon^{e}_{ik}\overline{E}_{k} = \varepsilon^{h}_{ik}\overline{E}_{k} + \frac{1}{V} \int_{\Omega_{i}} \left[e^{i}_{ikl}\gamma_{kl} + (\varepsilon^{i}_{ik} - \varepsilon^{h}_{ik})E_{k} \right] dV, \quad (18)$$
$$c^{e}_{ijkl}\overline{\gamma}_{kl} - e^{e}_{kij}\overline{E}_{k} = c^{h}_{ijkl}\overline{\gamma}_{kl} + \frac{1}{V} \int_{\Omega_{i}} \left[(c^{i}_{ijkl} - c^{h}_{ijkl})\gamma_{kl} - e^{i}_{kij}E_{k} \right] dV.$$

$$V \int_{\Omega_i} \left[\left(C_{ijkl} - C_{ijkl} \right) \gamma_{kl} - C_{kij} D_k \right] dV.$$
(19)

Because the external electric field E_0 is applied to the graded piezoelectric composite along *z* direction and the applied external strain tensor γ_{kl}^0 is zero, the effective dielectric response ε_{zz}^e and piezoelectric responses e_{zij}^e can be estimated in the dilute limit as follows:

$$\varepsilon_{zz}^{e} = \varepsilon_{zz}^{h} + \frac{1}{VE_{0}} \int_{\Omega_{i}} [e_{zkl}^{i} \gamma_{kl} + (\varepsilon_{zk}^{i} - \varepsilon_{zk}^{h})E_{k}]dV, \qquad (20)$$

$$e_{zij}^{e} = -\frac{1}{VE_{0}} \int_{\Omega_{i}} [(c_{ijkl}^{i} - c_{ijkl}^{h})\gamma_{kl} - e_{kij}^{i}E_{k}]dV \quad (i, j = x, y, z), \qquad (21)$$

Here, note that the quantities in Eqs. (20) and (21) are represented in Cartesian coordinates. Using transformations from spherical coordinates $(u_{\theta}, u_{\varphi}, u_r)$ to Cartesian coordinates (u_x, u_y, u_z) (for example, the z-component transformation formula is $u_z = u_r \cos \theta - u_{\theta} \sin \theta$), in the inclusion region, we have

$$e_{zkl}\gamma_{kl} + \varepsilon_{zk}E_{k} = D_{z} = D_{r}\cos\theta - D_{\theta}\sin\theta$$

$$= \cos^{2}\theta\sum_{i=1}^{3} \left[e_{33}^{0}(K_{1i}^{1} - K_{1i}^{2})\nu_{1i} + 2e_{31}^{0}(K_{1i}^{1} + 1)\right]A_{1i}r^{\nu_{1i}(k) - 1 + k}$$

$$- \sin^{2}\theta\sum_{i=1}^{3} \left[\varepsilon_{11}^{0}\frac{e_{33}^{0}}{\varepsilon_{33}^{0}}K_{1i}^{2} + e_{15}^{0}(\nu_{1i} - 1 - K_{1i}^{1})\right] \times A_{1i}r^{\nu_{1i}(k) - 1 + k}, \qquad (22)$$

$$\varepsilon_{zk}^{h} E_{k} = D_{r}^{hi} \cos \theta - D_{\theta}^{hi} \sin \theta$$

$$= -\varepsilon^{h} \frac{e_{33}^{0}}{\varepsilon_{33}^{0}} \sum_{i=1}^{3} K_{1i}^{2} \nu_{1i} A_{1i} r^{\nu_{1i}(k)-1} \cos^{2} \theta$$

$$-\varepsilon^{h} \frac{e_{33}^{0}}{\varepsilon_{33}^{0}} \sum_{i=1}^{3} K_{1i}^{2} A_{1i} r^{\nu_{1i}(k)-1} \sin^{2} \theta, \qquad (23)$$

where the superscript hi denotes that the physical properties are those of the host region, while the electric field and the strain tensor field are those of the inclusion region. Substituting Eqs. (22) and (23) into Eq. (20), we obtained the effective dielectric response of graded piezoelectric composites in the dilute limit,

$$\varepsilon_{zz}^{e} = \varepsilon_{zz}^{h} + \frac{1}{VE_{0}} \int_{\Omega_{i}} \left[(e_{zkl}\gamma_{kl} + \varepsilon_{zk}E_{k}) - \varepsilon_{zk}^{h}E_{k} \right] dV$$

$$= \varepsilon^{h} + pE_{0}^{-1} \sum_{i=1}^{3} \left[e_{33}^{0} (K_{1i}^{1} - K_{1i}^{2})\nu_{1i} + 2e_{31}^{0} (K_{1i}^{1} + 1) \right] A_{1i}$$

$$\times \frac{a^{\nu_{1i}+k-1}}{\nu_{1i} + k + 2} - 2pE_{0}^{-1} \sum_{i=1}^{3} \left[\varepsilon_{11}^{0} \frac{e_{33}^{0}}{\varepsilon_{33}^{0}} K_{1i}^{2} + e_{15}^{0} (\nu_{1i} - 1 - K_{1i}^{1}) \right] A_{1i} \frac{a^{\nu_{1i}+k-1}}{\nu_{1i} + k + 2} + pE_{0}^{-1} \varepsilon^{h} \frac{e_{33}^{0}}{\varepsilon_{33}^{0}}$$

$$\times \sum_{i=1}^{3} K_{1i}^{2} A_{1i} a^{\nu_{1i}(k)-1}, \qquad (24)$$

where p is the volume fraction of the graded spherical inclusions.

Furthermore, we can estimate the effective piezoelectric response e_{zij}^{e} (*i*,*j*=*x*,*y*,*z*) by means of Eq. (21). For example, e_{zzz}^{e} in the dilute limit is given by

$$e_{zzz}^{e} = -\frac{1}{VE_{0}} \int_{\Omega_{i}} \left[(c_{zzkl}^{i} - c_{zzkl}^{h}) \gamma_{kl} - e_{kzz}^{i} E_{k} \right] dV.$$
(25)

Using tensor transformations between the spherical coordinates and the Cartesian coordinates $\sigma_{zz} = \sigma_{rr} \cos^2 \theta$ $-2\sigma_{r\theta} \cos \theta \sin \theta + \sigma_{\theta\theta} \sin^2 \theta$ in the inclusion region, we get

$$c_{zzkl}\gamma_{kl} - e_{kzz}E_k = \sigma_{zz} = \sigma_{rr}\cos^2\theta - 2\sigma_{r\theta}\cos\theta\sin\theta + \sigma_{\theta\theta}\sin^2\theta,$$
$$C_{zzkl}^h\gamma_{kl} = \sigma_{zz}^{hi} = \sigma_{rr}^{hi}\cos^2\theta - 2\sigma_{r\theta}^{hi}\cos\theta\sin\theta + \sigma_{\theta\theta}^{hi}\sin^2\theta.$$

Clearly, the stress in the inclusion region has the following relations with respect to the variable θ : $\sigma_{rr} \propto \cos \theta$, $\sigma_{r\theta} \propto \sin \theta$, and $\sigma_{\theta\theta} \propto \cos \theta$. Using the orthogonality of the trigonometric functions, we get

$$e_{zzz}^e = 0. (26)$$

Similarly, we have also obtained the effective piezoelectric responses $e_{zii}^e = 0$ (i, j = x, y, z). Note that, for an external electric field acting along the \hat{x} direction or \hat{y} direction, the effective piezoelectric constants e_{xij}^{e} and e_{yij}^{e} (i,j=x,y,z) are zero. Here, we have omitted the derivation process since it is similar to the case of an external field acting along the \hat{z} direction. The results indicate that the effective piezoelectric response vanishes due to the symmetry of this kind of spherically anisotropic piezoelectric composite system even in graded piezoelectric composites in this study. This kind of effective piezoelectric response mechanism will have important applications in designing functionally graded composite materials. Note that if the symmetry of the spherically anisotropic system is destroyed, then bulk effective piezoelectric properties will appear. For example, if a transversely isotropic piezoelectric spherical inclusion is immersed in a nonpiezoelectric matrix, the composite will have nonzero effective piezoelectric responses.²⁷



FIG. 1. Effective dielectric constant $\varepsilon_{zz}^{e}/\varepsilon^{h}$ vs the dielectric constant factor *n* of the graded inclusions for different values of the power-law parameter *k*. The elastic and piezoelectric factors *n* of the graded inclusions are fixed at 10.

In order to discuss the coupling effects of the elastic, dielectric, and piezoelectric properties of the inclusion on the effective dielectric constant, we consider again the spherically anisotropic graded piezoelectric composite. We have calculated the effective dielectric response ε_{zz}^{e} [Eq. (24)] at a small volume fraction, say, p=0.1, and taking the radius a = 1, the elastic parameters and dielectric constant of the host region $\nu=0.25$, $\mu=32$ Gpa, and $\varepsilon^{h}=6 \times 10^{-10}$ C² N⁻¹ m⁻². Also, the elastic, piezoelectric, and dielectric constants of the graded spherical inclusion in spherical coordinates are taken to be $\varepsilon_{ij}(r)=\varepsilon_{ij}^{0}r^{k}$, $e_{ij}(r)=e_{ij}^{0}r^{k}$, and $c_{ij}(r)=c_{ij}^{0}r^{k}$, here $C_{11}^{0}=16.6n$ GPa, $C_{33}^{0}=16.2n$ GPa, $C_{12}^{0}=7.7n$ GPa, $C_{44}^{0}=4.3n$ GPa, $C_{13}^{0}=7.8n$ GPa, $e_{31}^{0}=-0.44n$ C m⁻², $e_{33}^{0}=1.12n \times 10^{-9}$ C² N⁻¹ m⁻², where *n* is a dimensionless factor. In Figs. 1–3, we let the dielectric, piezoelectric, and elastic factors *n* to vary from 1 to 20, while other factors *n* are fixed at 10. In Fig. 4, the factors *n* of the dielectric, piezoelectric, and elastic constants are fixed at 10.

In order to test our formulas, in the dilute limit p=0.1, we considered a pure isotropic dielectric spherical composite



FIG. 3. Effective dielectric constant e_{zz}^{e}/e^{h} vs the elastic constant factor *n* of the graded inclusions for different values of the power-law parameter *k*, with the dielectric and piezoelectric constant factors *n* of the graded inclusion fixed at 10.

without gradient profiles, i.e., k=0 and $e_{ii}^0=0$, and compared our results with Bruggemann formula²⁸ in Fig. 1. Clearly, our results are in good agreement with Bruggemann's. Furthermore, we find that the effective dielectric constant increases with the dielectric and piezoelectric constants of the inclusion for a given value of the power-law parameter k (Figs. 1 and 2). On the contrary, the effective dielectric response decreases as the elastic constant increases (Fig. 3). However, it can be seen that, for a given set of parameters, the effective dielectric constant for smaller k is larger than those of larger k (Figs. 1–3). This implies that the power-law parameter kcan play an important role in controlling the effective dielectric response of this kind of graded piezoelectric composite. To discuss the effects of the volume fraction and the powerlaw parameter k on the effective dielectric response, Fig. 4 shows that the effective dielectric response decreases when kincreases. It also shows that for larger dielectric constant of the host material, when the value of k is larger (or smaller) than a critical value (in our case this value is about 3), the effective dielectric constant decreases (or increases) when



FIG. 2. Effective dielectric constant $\varepsilon_{zz}^{\ell}/\varepsilon^h$ vs the piezoelectric constant factor *n* of the graded inclusions for different values of the power-law parameter *k*, with the elastic and dielectric factors *n* of graded inclusions fixed at 10.



FIG. 4. Effective dielectric constant $\varepsilon_{zz}^{e}/\varepsilon^{h}$ vs the power-law parameter k of the graded inclusions for volume fractions p=0.1,0.08,0.05 and the host relative dielectric constants $\varepsilon^{h}=67.8$ and 678. The factors n for the dielectric, piezoelectric, and elastic constants of the graded inclusions are set to 10.

the volume fraction p increases. This result further shows that the power-law parameter k can enhance or reduce the effective dielectric response of this kind of graded piezoelectric composites.

IV. CONCLUSIONS

Spherically anisotropic graded piezoelectric composites having power-law graded spherical inclusions in a nonpiezoelectric matrix are investigated, and analytical solutions of the elastic displacement field and electric potential under an external electric field are derived. In the dilute limit, formulas of the effective dielectric and piezoelectric responses are given. Our results show that spherically anisotropic graded piezoelectric composite does not have bulk piezoelectric behavior due to the symmetry of the composite system and the radial polarization of the spherical inclusions. Of course, for nonsymmetric composites, either due to the physical properties or the structure, effective piezoelectric response will appear, such as in transversely isotropic piezoelectric spherical or ellipsoidal inclusions in a nonpiezoelectric matrix.²⁷ Our results also indicate that the dielectric response of the composite can be controlled by the particle piezoelectric, dielectric, elastic constants, and the power-law parameter k of the inclusion grading profile. Furthermore, we find that the piezoelectric and dielectric constants (or elastic constant) can enhance (or reduce) the effective dielectric constant. Also, the effective dielectric constant decreases (or increases) when the volume fraction p increases if the power-law parameter k is larger (or smaller) than a critical value. These properties demonstrate that there are coupling effects on the effective dielectric response due to the graded physical properties of the inclusions. As an example, we have shown that the power-law parameter of the grading profile can be used to change the effective dielectric response.

Based on our analytical solutions, the effective response at higher concentration of the inclusions can be estimated by means of effective medium approximation and other piezoelectric composites having other grading profiles, such as exponential profile, can be investigated and compared. It is also possible to develop a method for studying the effective nonlinear dielectric response of graded piezoelectric composites by further development on some existing methods for solving nonlinear composites problems.^{29–32}

ACKNOWLEDGMENTS

This work was supported by the Centre for Smart Materials and the NSFC (Grant Nos. 10374026 and 40476026).

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