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Material selection rules for optimal size-dependent flexoelectric enhancement in lead-free piezocomposites

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Abstract

A promising pathway to maximize the performance of lead-free piezocomposites involves optimizing complex coupled processes such as flexoelectricity, geometric structure of the piezocomposite material and device, and the choice of materials. Here, we present a simple example demonstrating how the performance of pieozocomposites can be boosted significantly using a combination of geometric anisotropy and optimal mechanical properties of the matrix material. The optimal design is conducive to large strain gradients that lead to considerable flexoelectric enhancement to the electromechanically coupled response of the composite to applied mechanical stimuli.

Keywords: flexoelectricity, lead-free piezocomposites, strain gradients, anisotropy, strain gradients

1 Introduction

Size dependent effects offer an interesting route to enhancing material properties through critical coupled processes which are very sensitive to the material size-scales. Flexoelectricity is one such important effect in the area of piezoelectric composite materials where considerable enhancements in the effective piezoelectric response of the material are possible at smaller length scales where the strain gradients are relatively large [1]. The coupling between the strain gradients and the electric field results in significant size-dependent piezoelectric effects leading to large enhancements at smaller material size scales. A condition which is critical to such flexoelectric enhancements is the presence of an elastic gradient in the material [2]. This means that the material geometry must enable the development of large strain gradients. This is possible through the introduction of geometric anisotropy in the material geometry by shaping the materials as, for example, truncated pyramids or triangles. On the other hand, it is also worth considering other such material designs which are amenable to 3D printing and other additive manufacturing techniques. Further, specific to the design of piezoelectric composites is the selection of the matrix materials. Flexoelectric enhancements are significantly affected by the elastic properties of the matrix and it is important for the design to take into account the effects of these properties. In this work, we consider an elastically anisotropic composite, the anisotropy being introduced through a graded piezoelectric inclusion concentration profile. Variants of this design could be amenable to additive manufacturing. We explore the size dependent enhancements to piezoelectricity in this composite structure using an RVE approach that evaluates the effective piezoelectric coefficient. In doing so, we consider environmentally friendly lead-free $BaTiO_3$ piezoelectric inclusions embedded in different polymeric matrices with varying elastic properties.

2 Methods

Here, we consider a piezocomposite with a graded inclusion concentration as illustrated in Figure 1(a). The concentration of the randomly shaped $BaTiO_3$ inclusions decreases as we move along the x_3 direction. This offers an elastic gradient which can help the development of strain gradients and flexoelectric effects. The behavior of the composite is numerically modelled using constitutive relationships derived from a common free energy function that contains the contributions of the major coupled effects – linear piezoelectricity and flexoelectricity, in addition to the energy stored in the composite due to pure electric field and elastic field effects [2, 3]. The electroelastic behavior of the composite is modelled using the following constitutive relationships:

$$\sigma_{ij} = c_{ijkl}\varepsilon_{kl} - e_{kij}E_k, \tag{1a}$$
$$\hat{\sigma}_{ijk} = \mu_{lijk}E_l, \tag{1b}$$
$$D_i = \epsilon_{ij}E_j + e_{ijk}\varepsilon_{jk} + \mu_{ijkl}\varepsilon_{jk,l}, \tag{1c}$$

where σ_{ij} and ε_{ij} are the components of the stress and strain tensors, $\hat{\sigma}_{ijk}$ are the higher order stress, E_i and D_i are the electric field and flux density vector components, $c_{ijkl}, e_{ijk}, \mu_{ijkl}$, and ϵ_{ij} are the elastic, linear piezoelectric, linear flexoelectric, and permittivity coefficients of the materials comprising the composite. These equations are further subject to the following balance equations:

$$\left(\sigma_{ij} - \hat{\sigma}_{ijk,k}\right)_{,j} + F_i = 0, \tag{2a}$$

$$D_{i,i} = 0, \tag{2b}$$

The boundary conditions applied to the composite are illustrated in Figure 1(b). These correspond to the boundary conditions to evaluate the effective piezoelectric coefficient e_{33}^{eff} of the composite [4]. As also seen in Figure 1(a), the geometry of the RVE is scaled by the factor N (the effective thickness), with respect to a reference thickness $h_0 = 50$ µm.

We consider three different classes of matrix materials – (a) Hydrogel-like materials with small elastic moduli ($E_m = 1$ MPa, ν_m=0.35), (b) Epoxy-like materials with relatively larger moduli ($E_m = 1$ GPa, $\nu_m = 0.35$), and (c) PDMS-like rubbery materials with relatively smaller elastic moduli, but large Poisson's ratios corresponding to incompressible materials ($E_m = 1$ MPa, $\nu_m = 0.499$). The other material properties considered in the model are summarized in Table 1. λ_m and μ_m correspond to the Lame's constants of the elastically isotropic matrix, given by $\lambda_m = \frac{E_m \nu_m}{(1+\nu_m)(1-2\nu_m)}$ and $\mu_m = \frac{E_m}{2(1+\nu_m)}$.

Material property	**Values for $BaTiO_3$**	**Values for matrix**
	Elastic coefficients (Moduli in Pa)	
c_{11}	275.1×10^9 [5]	$\lambda_m + 2\mu_m$
c_{13}	151.55×10^9	λ_m
c_{33}	164.8×10^9	$\lambda_m + 2\mu_m$
c_{44}	54.3×10^9	μ_m
Young's modulus, E_m	N.A.	$E_m = [1\text{MPa}, 1\text{GPa}, 1\text{MPa}]$
Poisson's ratio, ν_m	N.A.	$\nu_m = [0.35, 0.35, 0.499]$
	Relative permittivity	
ϵ_{11}/ϵ_0	1970 [5]	3.5
ϵ_{33}/ϵ_0	109	3.5
	Piezoelectric coefficients (Cm^{-2})	
e_{15}	21.3 [5]	Matrix is non-piezoelectric
e_{31}	-2.69	
e_{33}	3.65	
	Flexoelectric coefficients (Cm^{-1})	
Longitudinal, μ_{11}	1×10^{-6} [6]	1×10^{-9} [7]
Transverse, μ_{12}	1×10^{-6} [8]	1×10^{-9}
Shear, μ_{44}	0	0

Table 1: Electroelastic coefficients of the $BaTiO_3$ piezoelectric inclusions and the matrix.

Here E_m and ν_m correspond to the elastic modulus and the Poisson's ratio of the matrix, respectively.

3 Results

We evaluate the size dependent flexoelectric enhancement as $\frac{e_{33}(N)}{e_{33}^0}$, where e_{33}^0 is the effective piezoelectric coefficient evaluated at large N. These results are plotted in Figure 1(c). We notice clearly that while both the incompressible matrix and the harder epoxy-like matrices lead to very small size-dependent enhancements, the softer matrix having small elastic moduli, which are compressible (ν_m=0.35, here), show large flexoelectric enhancements. This is likely due to the favorable matrix elasticity which allows the development of deformations which give rise to large strain-gradients. Hard and compressible materials do not likely allow the development of large strain gradients. These gradients are also magnified by large contrast in the elastic properties at the interface between the soft matrix and relatively harder inclusions.

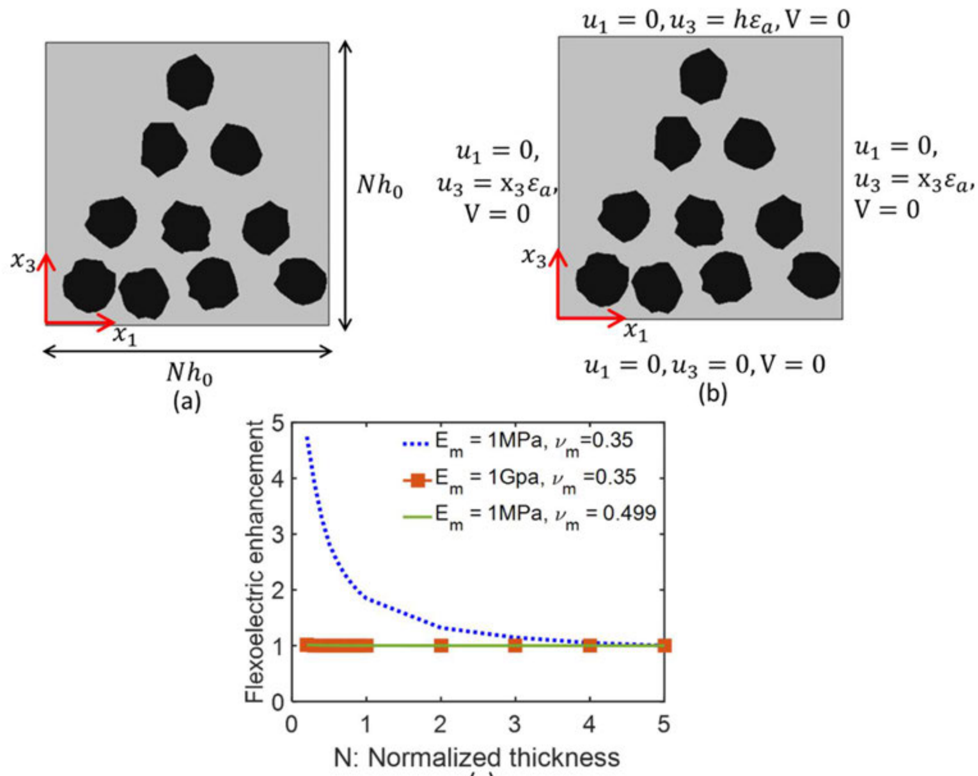

Figure 1: (a) The RVE used for the evaluation of size-dependent piezoelectric effect, (b) the boundary conditions employed where ε_a=1 × 10^{-6}, (b) the flexoelectric size dependent enhancements for different matrix materials.

4 Conclusions and Contributions

In summary, we have identified important design rules, from the perspective of material selection, to boost the piezoelectric response of geometrically anisotropic piezocomposites through the introduction of a graded inclusion concentration profile and selection of soft compressible matrix materials, which make feasible the development of large strain-gradients and therefore large flexoelectric contributions at smaller size scales. Further, these observations also point to new avenues of material design that could lead to efficient flexoelectric energy harvesting and sensing. For example, one could consider a matrix with a graded concentration of nanomaterials – carbon nanotubes, graphene, and so on – which can render the matrix elastically anisotropic and hence can make the conditions conducive to strain-gradient development. One could also think of graded metal-nanoparticle concentrations that could lead to a graded dielectric profile. The coupling between the electric field and the strains can then lead to gradients and flexoelectric behavior. Further, the design, and its variants, conceived here are amenable to additive manufacturing processes and therefore, scalable. Careful tuning of anisotropy within the material could also lead to directional sensing behavior and it could be possible to create a macroscale material with a distribution of microscopic anisotropies, therefore providing a large degree of freedom in designing anisotropic materials that might be of interest for several applications in sensing and energy harvesting of mechanical stimuli.

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