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Modeling of poling, piezoelectric, and pyroelectric properties of ferroelectric 0–3 composites

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A model for the piezoelectric and pyroelectric activities of 0–3 composites of ferroelectric particles in a linear matrix has been developed based on the polarization behavior of the inclusions. The model is applied to simulate the piezoelectric activity of a lead zirconate titanate/epoxy system polarized under different fields and the piezo- and the pyroelectric properties of lead titanate/polyvinylidene fluoride–trifluoroethylene 70/30 mol %] composites polarized with different poling times. The model predictions show reasonably good agreement with experimental data. The ability to predict the gross properties of 0–3 composites in terms of the poling field and poling time may be especially helpful in the making of practical composites with particular properties. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1596718]

I. INTRODUCTION

Composites of ferroelectric polymers and ferroelectric ceramics reveal piezoelectric and pyroelectric activity after being subjected to a high electric field which aligns the spontaneous polarization along the poling direction. These composites are useful for practical applications as they combine the high piezoelectric and pyroelectric activities of ferroelectric ceramic with excellent mechanical properties of a polymer. Applications of such composites include electromechanical transducers, e.g., microphones and hydrophones, as well as pyroelectric detectors in infrared sensors and microcalorimeters.

In our previous papers, we have theoretically studied some polarization switching behavior of multilayered[1] and 0–3 composites.[2] Effects of the permittivity and electric conductivity of the constituents have been discussed. However, there is still a lack of theoretical models to account for the effect of poling time and field, which predict the macroscopic properties of a ferroelectric composite from the polarization acquired by the constituents. Extensive experimental work is usually required to characterize and optimize composite systems which limit their application, in practice. Piezoelectric and pyroelectric activities of various 0–3 composite systems have been systematically studied and are reported in literature (e.g. see recent reviews by Dias *et al.*,[3] Tressler *et al.*,[4] and Bhimasankaram *et al.*,[5] and references therein). Most of these works were focused on technical aspects and applications, while many fundamental aspects were not fully understood. On the other hand, models for piezoelectric and pyroelectric properties of ferroelectric 0–3 composites have also been given recently.[6,7]

The emphasis of this article is to study the gross properties of such composite systems in terms of the poling field and poling time. This will be useful for the selection of materials and the poling treatment of composite systems for tailored applications. We shall give a theoretical description of the poling process of dilute 0–3 composite systems, built up from consideration of a single-inclusion problem in which a single-ferroelectric sphere is surrounded by a linear matrix medium under the action of a uniform electric field. A modified model of Miller *et al.*[8,9] is used for the description of the polarization–electric field ($P-E$) relation of the ferroelectric constituent. The predictions of the model will be compared to experimental results of the piezoelectric properties of PZT/epoxy 0–3 composites given by Furukawa *et al.*[10] and to our studies of piezo- and pyroelectric properties of lead titanate (PT)/polyvinylidene fluoride–trifluoroethylene [P(VDF–TrFE)] 0–3 composites.

II. THEORY

Consider a ferroelectric ceramic/polymer composite with 0–3 connectivity, i.e., the ferroelectric ceramic particles are not in contact with each other while the polymer phase is self-connected in three dimensions as illustrated schematically in Fig. 1. The particles are assumed to have a spherical shape.

The electric displacement D of the ceramic and polymer phase are given by[11]

$$D_i = \varepsilon_i E_i + P_i , \tag{1}$$

$$D_m = \varepsilon_m E_m + P_m , \tag{2}$$

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