Optimized Pyroelectric Properties of 0-3 Composites of PZT Particles in Polyurethane Doped with Lithium Perchlorate

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Abstract—**A substantial improvement in the performance of pyroelectric 0-3 composites of ceramic particles in a polymer matrix has been achieved by doping the polymer matrix material. Readily prepared and polarized films with various volume fractions of lead zirconate-titanate (PZT) particles in polyurethane have been doped in a solution of lithium perchlorate in acetone to increase the conductivity. With an appropriate conductivity, the dielectric permittivities of the ceramic particles and the polymer matrix become matched, resulting in an improvement of the pyroelectric coefficient from about 6 $\mu C/(m^2K)$ to about 50 $\mu C/(m^2K)$. The experimental results are explained by theoretical predictions.**

I. Introduction

Composites of ferroelectric ceramic particles in a polymer matrix are promising materials for application in pyroelectric sensors [1]. Compared to ceramic films, they have the advantage of higher process compatibility with the fabrication of integrated circuits. The performance of pyroelectric 0-3 composites, however, is limited by the dielectric mismatch between the high dielectric permittivity of the ferroelectric ceramic particles and the substantially lower dielectric permittivity of the polymer matrix. This mismatch can be reduced substantially when the matrix is partially conducting [2], resulting in a substantial increase of the pyroelectric performance.

II. Theory

The pyroelectric coefficient p of a composite of pyroelectric ceramic particles with pyroelectric coefficient p_i and relative dielectric permittivity ε_i embedded in a matrix material with dielectric permittivity ε_m generally is expressed as [3]:

$$p = \frac{\varepsilon - \varepsilon_m}{\varepsilon_i - \varepsilon_m} p_i, \tag{1}$$

where ε is the effective dielectric permittivity of the composite. Effective medium models for ε always depend on the shape and distribution of the constituents, i.e., for ε there exists no general formula like (1) for p. Recently, the Poon-Shin model was derived, which gives very good results for 0-3 composites with high ceramic volume fractions [4]:

$$\frac{\varepsilon}{\varepsilon_m} = 1 + \frac{\nu(\varepsilon_i/\varepsilon_m - 1)}{\nu + (1-\nu)[(\varepsilon_i/\varepsilon_m)(1-\nu) + \nu + 2]/3}, \tag{2}$$

where ν is the volume fraction of the inclusions. From (1), (2) follows the pyroelectric coefficient:

$$p = \frac{3\nu}{3\nu + (1-\nu)[\varepsilon_i/\varepsilon_m + 2 - \nu(\varepsilon_i/\varepsilon_m - 1)]} p_i. \tag{3}$$

When we are looking for an optimum selection for ε_m of the matrix material, (3) would suggest a ε_m as high as possible, as p becomes maximum when ε_m goes to infinity. Then, however, the impedance of the sensor material goes to zero, and so the pyroelectric power that can be coupled to an external load resistor goes to zero too. The proper optimization is to choose a ε_m for which a maximum pyroelectric power can be coupled into an external load. When a pyroelectric sensor film with relative dielectric permittivity ε, thickness d, and area A is periodically heated at circular frequency ω, a maximum pyroelectric signal power is coupled into an external load resistor, when its value R_L is chosen as:

$$R_L = \frac{d}{\omega\varepsilon_0\sqrt{\varepsilon'^2 + \varepsilon''^2} + \varepsilon''}. \tag{4}$$

Then the pyroelectric signal power P_L coupled into the load resistor is proportional to:

$$P_L \propto \frac{p^2}{\sqrt{\varepsilon'^2 + \varepsilon''^2} + \varepsilon''}. \tag{5}$$

With p and ε from (1), (2) follows that P_L is maximum for a matrix permittivity of:

$$\varepsilon_m = \varepsilon_i \sqrt{\frac{1-\nu^3}{4 + 6\nu - \nu^3}}. \tag{6}$$

For a composite with 30% ceramic volume fraction, the dielectric permittivity of the matrix material would have to be about 40% of the dielectric permittivity of the inclusions. For particles of lead zirconate titanate (PZT) dispersed in a polymer, this condition cannot be met. The

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