Optimized pyroelectric 0-3 composites of PZT particles in doped polyurethane

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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 μC/(m^{2}K) to about 40 μC/(m^{2}K) at modulation frequencies around 50 Hz. The experimental results are explained by theoretical predictions.

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 is, however, 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.

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 is generally expressed as [3]:

$$p = \frac{\varepsilon - \varepsilon_m}{\varepsilon_i - \varepsilon_m} p_i \qquad (1)$$

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

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

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

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

When we are looking for an optimum selection for ε_m of the matrix material, Eq. (3) would suggest an 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 which can be coupled to an external load resistor. The proper optimization is to choose an ε_m for which a maximum pyroelectric power can be coupled into an external load. This leads to

$$\varepsilon_m = \varepsilon_i \sqrt{\frac{1-\nu^3}{4 + 6\nu - \nu^3}} \qquad (4)$$

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 can not be met. The permittivity of polymers is by for too low resulting in a substantial dielectric mismatch in ceramic/polymer composites. It is, however, possible to introduce a dc conductivity by doping the polymer matrix and raise the imaginary part ε_m'' to the value given by Eq. (4).

The pyroelectric coefficient has been simulated for a composite of 30 % volume fraction of particles with $\varepsilon_i' = 290$, $\varepsilon_i'' = \varepsilon_i' \cdot 0.003$ and $p_i = 290$ μC/(m^{2}K) (typical values for small PZT particles) dispersed in a matrix with $\varepsilon_m' = 5$ and $\varepsilon_m'' = \varepsilon_m' \cdot 0.02$ (e.g. polyurethane). Fig. 1 shows the pyroelectric coefficient vs. frequency f for matrix conductivities $\sigma_m = 10^{-13}$ /(Ωm) (undoped) and $\sigma_m = 2 \cdot 10^{-7}$ /(Ωm). The complex permittivity of the matrix material causes a frequency

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