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THE DIELECTRIC, MECHANICAL AND PIEZOELECTRIC PROPERTIES OF 3-3 PIEZOELECTRIC COMPOSITES

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The effective material parameters of the 3-3 piezocomposites are estimated using constitutive equations corresponding to the thickness-mode oscillations of a piezocomposite plate resonator. The dielectric constant ($\varepsilon_{33}^{T}/\varepsilon_0$), piezoelectric coefficients (d_{33}, d_{31}, d_h, g_h and e_{33}), stiffness coefficient (c_{33}^{E}), electrical impedance spectrum, electromechanical thickness-mode coupling coefficient (k_t), acoustic impedance (Z_{aco}) of the 3-3 composites are evaluated as a function of piezoceramic/polymer volume fraction. The results are compared with the experimental data.

Keywords: *Piezoelectric properties, Piezocomposites, transducers, thickness-mode*

I. INTRODUCTION

Piezocomposites have several advantages over single-phase piezoceramic materials. Their superior hydrostatic performance, better acoustic impedance matching with human tissue and water, and higher electromechanical efficiency make them useful for bio-medical and underwater applications [1]. Piezocomposites consist of active piezoceramic and passive polymer materials arranged in a specific configuration called *connectivity.* In piezocomposites with 3-3 connectivity, the interpenetrating piezoceramic and polymer materials are self-connected in three dimensions. The material characteristics of a piezocomposite can be estimated by modelling a fundamental block, called '*unit cell*' which represents the entire piezocomposite. [2,3]. Bowen *et al.*[4] proposed a model, in which the '*unit cell*' is split into four volumes with series/ parallel combinations of piezoceramic and polymer components. This model assumes that only certain volumes contribute effectively to the piezoelectric charge coefficients and dielectric constant, depending on the alignment of piezoceramic component with respect to the stresses applied in the longitudinal and lateral directions. We have used this model to determine further material parameters of a 3-3 piezocomposite transducer vibrating in the thickness-mode.

II. CONSTITUTIVE RELATIONS

Consider a 3-3 piezocomposite thin plate fully electroded on the planar surfaces and poled in the thickness direction. The effective material parameters of the composite can be determined using the material parameters of the constituent phases, viz. piezoceramic and polymer. The piezoelectric charge and voltage coefficients of the piezocomposites under hydrostatic condition are given by,

$$\bar{d}_h = \bar{d}_{33} + 2\bar{d}_{31} \tag{1}$$

and

$$\bar{g}_h = \frac{\bar{d}_h}{\bar{\varepsilon}^T_{33}} \tag{2}$$

where, $\bar{d}_{33}$ and $\bar{d}_{31}$ are the piezoelectric charge coefficients and $\bar{\varepsilon}^T_{33}$ is the dielectric permittivity at constant stress, as defined in Ref [3]. The bar over the character represents the materials parameters of the composite.

The density of the composite is given by,

$$\bar{\rho} = \rho^c v^c + \rho^p v^p \tag{3}$$

where ρ and v are the density and volume fraction of the ceramic and polymer phases marked by the superscripts c and p respectively.

The electrical impedance of the composite plate vibrating in thickness-mode is given by,

$$\bar{Z} = \frac{1}{j\omega\bar{C}_0}\left[1 - \bar{k}_t^2 \frac{\tan(\alpha t)}{\alpha t}\right] \tag{4}$$

where, t is half thickness of the composite plate

$$\alpha = \omega\sqrt{\frac{\bar{\rho}}{\bar{c}^D_{33}}} \tag{5}$$

$$\omega = 2\pi f \tag{6}$$

and $\bar{C}_0$ is the clamped capacitance as given by,

$$\bar{C}_0 = \frac{\bar{\varepsilon}^S_{33} A}{2t} \tag{7}$$

where A is the surface area of the electrodes.

The thickness-mode electromechanical coupling coefficient is given by,

$$\bar{k}_t^2 = \frac{\bar{e}_{33}^2}{\bar{\varepsilon}_{33}^S \bar{c}_{33}^D} \tag{8}$$

where, $\bar{e}_{33}$ is the piezoelectric coefficient determined using charge coefficient and stiffness coefficient of the composite [5].

The acoustic impedance is given by,

$$\bar{Z}_{aco} = \sqrt{\bar{c}_{33}^D \bar{\rho}} \tag{9}$$

III. EXPERIMENTAL METHODS

3-3 piezocomposite specimens with a range of ceramic volume ratios were prepared using two different methods. Specimens with higher ceramic volume ratios were prepared using BURPS (BURnout Plastic Spheres) method [2]. PZT 5H powder was mixed with polyethylene oxide (PEO) granules at required ratios and cold-pressed to form pallets, which were then heated to 600 ^{0}C to burn out PEO and sintered at 1200 ^{0}C for 2 hours. Specimens with low ceramic volume ratios were prepared using the Reticulated Foam Method. Pieces of foam in the cylindrical shape were dipped in a PZT slurry and dried in air. They were then heated to 600 ^{0}C to remove the foam material and sintered at 1200 ^{0}C for 2 hours. The porous PZT structure thus obtained were coated with silver electrodes on the flat surfaces, poled using a corona poling technique and filled with low viscosity polymer (SpeciFix A40, Struers) under vacuum. The piezoelectric charge coefficients (d_{33} and d_{31}) were measured using Berlincourt-type Peizo Meter, capacitance was measured using LCR bridge (HP 4263B) and electrical impedance was measured using Solartron Impedance Analyser.

IV. RESULTS AND DISCUSSION

The material parameters of the 3-3 piezocomposites are estimated using the present model and are given in Figures 1-8. In all the figures, the solid lines represent the analytical data and markers represent the experimental data. Figure 1 shows the variations in the piezoelectric charge coefficients $\bar{d}_{33}$, $\bar{d}_{31}$ and $\bar{d}_h$ as a function of ceramic volume fraction.
The hydrostatic charge coefficient increases with the ceramic volume ratio and reaches a maximum at around 30-40 %. It can be seen from the figure that the experimental data agree with the analytical result. Figure 2 shows the variation of dielectric constant with ceramic volume ratio. For low ceramic volume fractions, the experimentally measured values are very

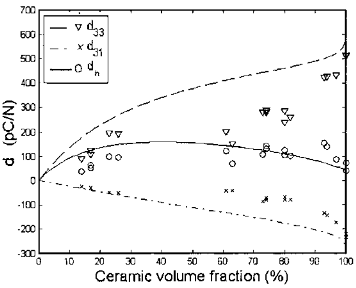

Fig.1 Piezoelectric charge coefficient as a function of ceramic volume fraction

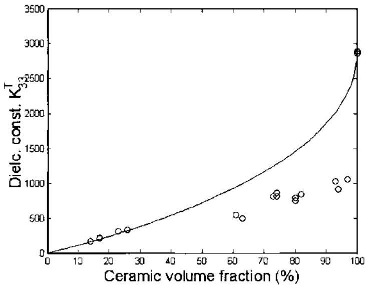

Fig.2 Dielectric constant as a function of ceramic volume fraction

close to the predicted values. The lower value of the experimental values for higher ceramic content may be due to the presence of isolated pores. This lowers the capacitance of the composites even for a small addition of polymer, because of serial connection with PZT. Similar effect of 0-3 connectivity on permittivity has also been reported in the literature [6].

Variations in the piezoelectric voltage coefficient, $\bar{g}_h$ and Figure-of-Merit (F0M), $\bar{d}_h \bar{g}_h$ are given in figure 3 and 4, respectively. The analytical curves show a maximum in the region of low ceramic volume fractions and the value of FoM is about three orders of magnitude higher than that of the

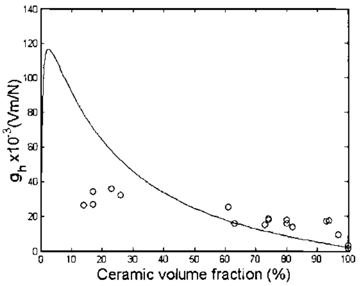

Fig.3 Piezoelectric voltage coefficient as a function of ceramic volume fraction

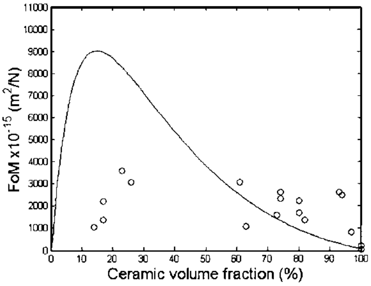

Fig.4 Figure-of-Merit as a function of ceramic volume fraction

pure ceramics. The experimentally measured values lie around the analytical curve for high ceramic volume ratios.

Figure 5 shows that the density of the piezocomposite is a linear function of the ceramic volume fraction and follows the simple 'rule of

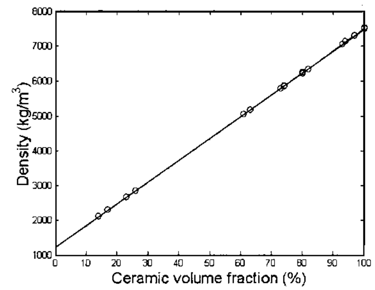

Fig.5 Density as a function of ceramic volume fraction

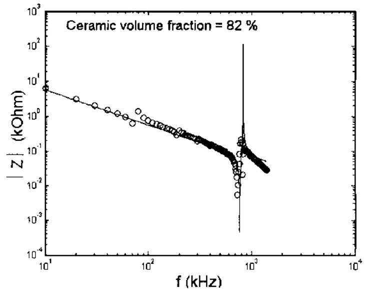

Fig.6 Electrical impedance spectrum for 82 % ceramic volume fraction

mixtures'. The electrical impedance spectra of the composite discs are estimated using Eq.4 and a representative plot for the case of 82 % ceramic volume fraction is shown in Figure 6, in which the resonance corresponds to the thickness-mode vibrations. The experimental results are in good agreement with the analytical results.

Figure 7 shows the variations in the thickness-mode electromechanical coupling coefficients ($\overline{k}_t$) of the composites. The analytical values are estimated using Eq.8 and the experimental values are obtained from the resonance (f_r) and anti resonance (f_a) frequencies of the electrical impedance spectra. $\overline{k}_t$ increases with the ceramic volume fraction, unlike the case of a 1-3 piezocomposite, for which the $\overline{k}_t$ curve shows a broad maximum for intermediate volume fractions [1]. The acoustic impedance ($\overline{Z}_{aco}$) data estimated using Eq.9 and obtained from the values of f_r and f_a are shown in Figure 8. The deviations in the experimental values of

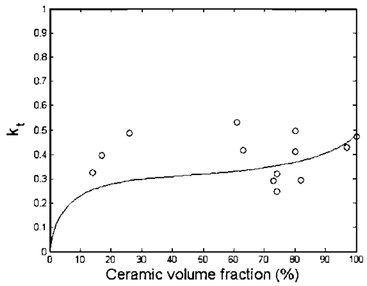

Fig.7 Thickness-mode coupling coefficient as a function of ceramic volume fraction

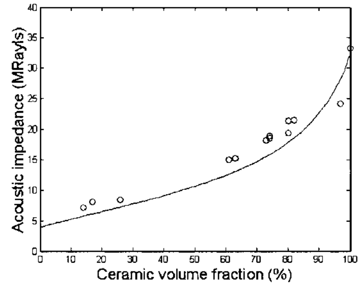

Fig.8 Acoustic impedance as a function of ceramic volume fraction

$\overline{k}_t$ and $\overline{Z}_{aco}$ from the analytical results may be due to the broadening of resonance for lower ceramic volume ratios and difficulties involved in precisely determining the resonance and anti resonance frequencies.

V. CONCLUSIONS

The effective material parameters of 3-3 piezocomposite thin discs (thickness-to-diameter ratio of 0.1) are estimated from the material parameters of the individual phases, viz, piezoceramic and polymer. Analytical results on the 3-3 piezocomposites are comparable with the experimental data. Lower values of dielectric constant for higher ceramic volume fractions could be attributed to the presence of isolated pores. Broad resonance peaks for lower ceramic volume ratios leads to difficulties in accurately determining the coupling coefficient from the experimental data.

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