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A method to estimate the material parameters of 1–3 piezocomposites by non-linear regression analysis is presented. Experimentally measured electrical impedance data are least-squares fitted to a theoretical expression which accounts for the internal losses of a piezocomposite plate resonator vibrating in the thickness-mode. The process converges rapidly and the estimated values of the material parameters are comparable with that obtained from the constituent relations of 1–3 piezocomposites. The advantage of this method is that it can be used to estimate the material parameters of lossy resonators whose characteristic frequencies cannot be determined accurately.

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# 1. INTRODUCTION

Piezocomposite transducers, composed of piezoelectric ceramic and passive polymer, have attracted considerable interest in recent years owing to their underwater and bio-medical applications [1]. Their superior hydrostatic characteristics render them useful for hydrophone applications [2, 3]. Improved acoustic matching, high electromechanical efficiency and uni-modal operation of the 1–3 piezocomposite transducers offer great potential for ultrasonic medical imaging compared with the conventional piezoceramics and piezopolymers [4]. The transducer characteristics of the piezocomposites can be optimized for a specific application by altering the ceramic/polymer volume fraction, polymer stiffness and so on. Smith and Auld [5] have proposed a model to estimate the material parameters of 1–3 piezocomposites in terms of the material parameters of the constituent phases, namely, piezoelectric ceramic and polymer. The transducer characteristics of piezocomposites have been studied thoroughly by finite element analysis [3, 6, 7].

Methods generally used to estimate the material parameters of piezoceramic plates from the experimentally measured electrical impedance data can be extended to the piezocomposite plates. In this case, the piezocomposite plate can be regarded as a homogeneous medium with effective material parameters, provided that the lateral dimension and spacing of the ceramic rods are sufficiently small compared with the approppriate acoustic wavelength [2]. Smits [8] has proposed an iterative method to determine the material coefficients of piezoelectric resonators using the electrical admittance data at three fixed frequencies. Kwok *et al.* [9] have discussed different methods of estimating the material coefficients from the measured impedance data. Electromechanical coupling coefficients of lossy piezoelectric plates could be estimated from the electrical impedance measurements, by taking into consideration the internal mechanical losses [10]. Most of these methods are based on experimental data at a few characteristic frequencies. However, in the case of lossy resonators, accurate determination of the characteristic frequencies is difficult.

In this paper, we present a method to estimate the material parameters of 1-3 piezocomposite plates by fitting the experimentally measured electrical impedance data to a theoretical model by a non-linear regression analysis.

# 2. THEORETICAL METHODS

Consider a piezoelectric composite plate with dimensions as shown in Figure 1. The electrical impedance of a lossless piezoelectric thin plate poled in the thickness direction and vibrating in one-dimensional thickness mode is given by [11]

$$Z = \frac{1}{j\omega C_0} \left[ 1 - k_t^2 \frac{\tan(\alpha_z a_z)}{\alpha_z a_z} \right], \tag{1a}$$

where

$$\alpha_z = \frac{\omega}{V_z},\tag{1b}$$

$$C_0 = \frac{2\varepsilon_{33}^S a_x a_y}{a_z},\tag{1c}$$

 $\omega = 2\pi f$ ,  $V_z$  is the phase velocity in the z direction,  $k_t$  is the thickness mode coupling coefficient,  $C_0$  is the clamped capacitance,  $\varepsilon_{33}^S$  is the clamped dielectric constant and



Figure 1. Coordinates of a 1-3 piezocomposite plate.

 $2a_x$ ,  $2a_y$  and  $2a_z$  are the dimensions of the piezoelectric plate in the x, y and z directions respectively.

### 2.1. CONSTITUTIVE EQUATIONS OF MATERIAL PARAMETERS OF 1-3 PIEZOCOMPOSITE

The electrical impedance of the piezocomposite can be estimated by replacing the material parameters appearing in equation (1) by the effective parameters of the composites represented in terms of the constituent phases. When the lateral spacing of the ceramic pillars in the polymer matrix is relatively small compared with the wavelength of the fundamental thickness mode resonance of the composite plate, the following constitutive relations can be applied to describe the characteristics of 1–3 piezocomposites [1, 5]:

$$\bar{\rho} = \rho^c v^c + \rho^p v^p, \tag{2a}$$

$$\bar{k}_t^2 = \frac{\bar{e}_{33}^2}{\bar{e}_{33}^5 \bar{c}_{33}^D},$$
(2b)

$$\bar{c}_{33}^{D} = \bar{c}_{33}^{E} + \frac{\bar{e}_{33}^{2}}{\bar{\varepsilon}_{33}^{S}},$$
(2c)

$$\bar{V}_z = \sqrt{\frac{\bar{c} \frac{D}{33}}{\bar{\rho}}},\tag{2d}$$

$$\bar{c}_{33}^{E} = v^{c} \left[ c_{33}^{E} - \frac{2v^{p}(c_{13}^{E} - c_{12})^{2}}{v^{c}(c_{11} + c_{12}) + v^{p}(c_{11}^{E} + c_{12}^{E})} \right] + v^{p}c_{11},$$
(2e)

$$\bar{\varepsilon}_{33}^{s} = v^{c} \left[ \varepsilon_{33}^{S} - \frac{2v^{p} e_{31}^{2}}{v^{c} (c_{11} + c_{12}) + v^{p} (c_{11}^{E} + c_{12}^{E})} \right] + v^{p} \varepsilon_{11},$$
(2f)

$$\bar{e}_{33} = v^c \left[ e_{33} - \frac{2v^p e_{31}(c_{13}^E - c_{12})}{v^c (c_{11} + c_{12}) + v^p (c_{11}^E + c_{12}^E)} \right].$$
 (2g)

In above equations, the superscript "c" corresponds to the ceramic phase, "p" corresponds to the polymer phase, a bar over the character corresponds to the piezocomposite and "v" corresponds to the volume fraction of ceramic/polymer.  $\rho$  is the density, the symbols  $e_{33}, e_{31}, c_{33}^D, c_{33}^E, c_{12}^E$  and  $c_{13}^E$  correspond to the ceramic phase as described in IEEE STD [12] and the symbols  $c_{11}, c_{12}$  and  $\varepsilon_{11}$  correspond to the polymer phase.

The values of the material parameters obtained using these sets of equations are used as input data in the following analysis.

#### 2.2. ELECTRICAL IMPEDANCE OF LOSSY PIEZOCOMPOSITE RESONATORS

Piezocomposites are considered to be lossy materials. Attenuation in piezocomposites is due to the presence of lossy polymer matrix [13, 14]. As the

major contribution to the total loss arises from the internal mechanical losses, it is possible to obtain reasonably accurate results by taking into account only these losses in the analysis [15]. As a first approximation, the dielectric and piezoelectric losses are considered to be small compared with the mechanical losses. Mechanical losses can be included in the electrical impedance equation by considering the stiffness and electromechanical coupling coefficients as complex parameters [10] as given by

$$\bar{\mathbf{c}}_{33}^{\mathbf{D}} = \bar{c}_{33}^{\mathbf{D}} [1 + j \tan \delta],$$
 (3a)

$$\bar{\mathbf{k}}_{t} = \frac{\bar{k}_{t}}{\sqrt{(1+j\tan\delta)}},\tag{3b}$$

where  $\tan \delta$  is the loss factor (reciprocal of the quality factor Q). The material parameters are assumed to be independent of frequency. Substituting the complex coefficients given by equation (3) and the constituent relations of the piezocomposites given by equation (2) in equation (1), we get the electrical impedance of a composite plate vibrating in the thickness mode as follows:

$$Z(\omega) = \frac{1}{j\omega\bar{C}_0} \left[ 1 - \frac{\bar{k}_t^2}{\omega a_z} \sqrt{\frac{\bar{c}_{33}^D}{\rho(1+j\tan\delta)}} \tan\left(\omega a_z \sqrt{\frac{\rho}{\bar{c}_{33}^D(1+j\tan\delta)}}\right) \right].$$
(4)

By expanding, rearranging and separating the real and imaginary parts of this equation, we get

$$Z(\omega) = R(\omega) + jX(\omega), \tag{5}$$

where

$$R(\omega) = \frac{\bar{k}_t^2 \bar{c}_{33}^D}{\omega^3 \bar{C}_0 a_z^2 \bar{\rho}} \frac{A \sinh 2B + B \sin 2A}{\cos 2A + \cosh 2B},$$
(6a)

$$X(\omega) = \frac{-1}{\omega\bar{C}_0} + \frac{\bar{k}_t^2 \bar{c}_{33}^D}{\omega^3 \bar{C}_0 a_z^2 \bar{\rho}} \frac{A \sin 2A - B \sinh 2B}{\cos 2A + \cosh 2B},$$
(6b)

$$A = \omega a_z \sqrt{\frac{\bar{\rho}}{\bar{c}_{33}^D}} \left( 1 - \frac{3}{8} \tan^2 \delta \right), \tag{6c}$$

$$B = \omega a_z \sqrt{\frac{\bar{\rho}}{\bar{c}_{33}^D}} \left( \frac{1}{2} \tan \delta - \frac{5}{16} \tan^3 \delta \right), \tag{6d}$$

Since tan  $\delta$  is small, its higher order terms are neglected in equations (6c) and (6d). The modulus of impedance and the corresponding phase angle are given by

$$|Z(\omega)| = \sqrt{R(\omega)^2 + X(\omega)^2},$$
(7a)

$$\phi(\omega) = \tan^{-1}\left(\frac{X(\omega)}{R(\omega)}\right). \tag{7b}$$

### 2.3. ESTIMATION OF COEFFICIENTS

As can be seen from the above equations, the electrical impedance  $Z(\omega)$  is a non-linear function of the material parameters namely,  $\bar{e}_{33}^{S}$ ,  $\bar{k}_{t}$ ,  $\bar{c}_{33}^{D}$  and  $\tan \delta$ , which can be determined by the non-linear regression method [13].

Let  $a_i$ , i = 1, ..., 4 ( $\bar{\epsilon}_{33}^S$ ,  $\bar{k}_t$ ,  $\bar{c}_{33}^D$  and  $\tan \delta$  respectively) be the parameters to be determined,  $a'_i$  be the approximate initial values of  $a_i$ , and  $\Delta a_i = a_i - a'_i$ . Expanding  $|Z(\omega)|$  [equation (7a)] in Taylor's series in terms of  $\Delta a_i$ , we get

$$\mathbf{Z}| = |Z(\omega, \bar{\varepsilon}_{33}^{S}, \bar{k}_{t}, \bar{c}_{33}^{D}, \tan \delta)| + \frac{\partial |Z|}{\partial \bar{\varepsilon}_{33}^{S}} \Delta \bar{\varepsilon}_{33}^{S} + \frac{\partial |Z|}{\partial \bar{k}_{t}} \Delta \bar{k}_{t} + \frac{\partial |Z|}{\partial \bar{c}_{33}^{D}} \Delta \bar{c}_{33}^{D} + \frac{\partial |Z|}{\partial (\tan \delta)} \Delta (\tan \delta) + \cdots$$
(8)

Assuming that the approximate values of the parameters  $(a'_i)$  are so chosen that the correlation factors  $(\Delta a_i)$  are small, the higher order terms in equation (8) can be neglected. The corrections to the material parameters  $(\Delta a_i)$  could be estimated by least-squares fitting the experimentally measured data to equation (8).

The sum of squares of residuals is

$$S = \sum_{i=1}^{n} v_i^2 = \sum_{i=1}^{n} \left[ |\mathbf{Z}_i| - |Z_i^{exp}| \right]^2,$$
(9)

where  $|Z_i^{exp}|$  are the experimentally measured impedance magnitude values and *n* is the number of data points. *S* is a function of the correlation factors ( $\Delta a_i$ ) rather than the parameters themselves. To set *S* to the minimum, the partial derivatives of *S* with respect to  $\Delta a_i$  are equated to zero. Hence,

$$\frac{\partial S}{\partial (\Delta \bar{\varepsilon}_{33}^S)} = 0, \qquad \frac{\partial S}{\partial (\Delta \bar{k}_t)} = 0, \qquad \frac{\partial S}{\partial (\Delta \bar{c}_{33}^D)} = 0 \quad \text{and} \quad \frac{\partial S}{\partial (\Delta (\tan \delta))} = 0.$$
(10)

These normal equations are linear functions of  $\Delta a_i$ . The four unknown parameters are obtained from the solutions of these equations. More accurate results are obtained by taking these values as a new set of initial values and repeating the procedure a few times. This process can be terminated when it has converged with a pre-determined accuracy by setting a cut-off factor (*C*) defined as

$$C \ge \frac{a_i(k) - a_i(k-1)|}{a_i(k-1)}$$
 for  $i = 1, \dots, 4$ , (11)

where  $a_i(k)$  and  $a_i(k-1)$  are the values of the parameters  $(a_i)$  at kth and (k-1)th step.

### 3. EXPERIMENTS

Piezocomposite plates fabricated with 1-3 connectivity, composed of PZT-5A and Araldite (CY 230 + HY950) with 40% and 25% ceramic volume fractions and poled in the thickness direction were used in the present study. The periodicity of the ceramic pillars and the dimensions of the piezocomposite plates used for the

present study are given in Table 1. A thin layer of silver paint coated on the faces of the plates acts as electrode. The electrical impedance of bare plates of 1-3 piezocomposites was measured.

# 4. RESULTS AND DISCUSSION

The present analysis requires a set of approximate initial values of the material parameters. They were obtained from the material parameters of the piezoceramic and polymer components given in Table 2, using the constitutive relations given by equation (2). Using these values in equation (1), the magnitude of electrical impedance was calculated as a function of frequency for a lossless 1–3 piezocomposite plate with 40% ceramic volume fraction and is shown in Figure 2. It can be seen from this figure that the fundamental and the third harmonics of the thickness mode resonance frequencies coincide with the measured spectra.

Ceramic volume fraction	40%	25%
Ceramic pillar width (mm)	0·95	0.65
Polymer width (mm)	0·55	0.65
Composite plate thickness (mm)	6	6
Composite plate width (mm)	50	50

Dimensions of the piezocomposite plates

# TABLE 2

Input material parameters of piezoceramic and polymer components

Parameters	Values	
(a) PZT-5A: $c_{11}^{E} (10^{10} \text{ N/m}^2)$ $c_{12}^{E} (10^{10} \text{ N/m}^2)$ $c_{13}^{E} (10^{10} \text{ N/m}^2)$ $c_{33}^{E} (10^{10} \text{ N/m}^2)$ $e_{33} (c/m^2)$ $e_{31} (c/m^2)$ $e_{33}^{E} (10^{-9} \text{ F/m}^1)$ $\rho (\text{kg/m}^3)$	$ \begin{array}{r} 12.1 \\ 7.54 \\ 7.52 \\ 11.1 \\ 15.8 \\ -5.4 \\ 7.349 \\ 7700 \end{array} $	
(b) Araldite $c_{11} (10^{10} \text{ N/m}^2)$ $c_{12} (10^{10} \text{ N/m}^2)$ $\varepsilon_{11} (10^{-9} \text{ F/m}^1)$ $\rho (\text{kg/m}^3)$	0.8 0.44 0.0354 1150	



Figure 2. Variations of electrical impedance as a function of frequency of a lossless piezocomposite plate with 40% ceramic volume fraction. — Experimental spectra;  $-\cdot - \cdot$  Theoretical spectra obtained using equations (1) and (2), which do not account for losses.

However, the magnitude of impedance differs in the vicinity of resonance because equation (1) does not account for the losses.

Internal losses were introduced to the model by taking into consideration the complex material coefficients. In addition to the initial guess of the material parameters as obtained above, the initial value of the loss factor (tan  $\delta$ ) was obtained from the width at the half-maximum of the real part of impedance versus frequency (R-f) plot [17]. These approximate values were used as the input data to calculate the theoretical spectra of the magnitude of impedance using equation (7a). Experimentally measured impedance data were least-squares fitted to the theoretical model and refined values of the material parameters were obtained by the non-linear regression method described in section 2.3. The new set of values thus obtained was taken as the input for the next cycle and the procedure was repeated a few times to obtain more refined values. These values of the material parameters converge very rapidly and a decade of accuracy is achieved in every cycle. In other words, if the cut-off factor (C) is set to be  $10^{-3}$  then the process requires three steps. It has been observed that it always converges to the same values despite variations in the approximate input values. About 1500 data points were taken for this analysis. The initial and the estimated final values of material parameters of the piezocomposites with 40% and 25% ceramic volume fractions are presented in Table 3. It can be seen from this table that the values of material parameters estimated by this method are comparable with those obtained from constituent relations of the piezocomposites. A small difference in the values of

#### TABLE 3

Material parameters of 1–3 piezocomposites. Initial values of  $\bar{\varepsilon}_{33}^S$ ,  $\bar{k}_t$  and  $\bar{c}_{33}^D$  were calculated using equation (2) and tan  $\delta$  calculated from the width at half-maximum of R-f plots

Parameters	Initial values	Refined values
(a) 40% ceramic volume fraction	1:	
$\bar{\epsilon}_{33}^{S} (10^{-9}  \mathrm{F/m^{1}})$	3.075	3.409
$\overline{k_t}$	0.634	0.530
$\bar{c}_{33}^{D}$ (10 <sup>10</sup> N/m <sup>2</sup> )	4.947	4.992
$\tan \delta$	0.085	0.084
(b) 25% ceramic volume fraction	1:	
$\bar{\epsilon}_{33}^{S} (10^{-9}  \mathrm{F/m^{1}})$	1.936	2.668
$\overline{k_t}$	0.607	0.516
$\bar{c}_{33}^{D}$ (10 <sup>10</sup> N/m <sup>2</sup> )	3.367	3.357
$\tan \delta$	0.039	0.044

dielectric constant and coupling coefficient could be due to a small difference in dimensions of the ceramic pillars from the theoretical values and a partial depolarization effected by the dicing of ceramic plates during the process of fabrication [15], whereas the values obtained from equation (2) are for the ideal case [5].

The estimated complex coefficients are:  $\bar{c}_{33}^{D} = 4.992 \times 10^{10} + j0.419 \times 10^{10} \text{ N/m}^2$ and  $3.357 \times 10^{10} + j0.147 \times 10^{10} \text{ N/m}^2$ , and  $\bar{k}_t = 0.529 - j0.022$  and 0.515 - j0.011for the piezocomposites with 40% and 25% ceramic volume fractions respectively. The real parts of the stiffness and the coupling coefficients were also calculated using the measured values of frequencies at maximum conductance ( $f_s$ ) and maximum resistance ( $f_p$ ), according to the method described in IEEE Standard [12]. For instance, these coefficients for a piezocomposite with 40% ceramic volume fraction are calculated to be  $4.391 \times 10^{10} \text{ N/m}^2$  and 0.416, respectively, and for a 25% ceramic volume fraction calculated to be  $2.832 \times 10^{10} \text{ N/m}^2$  and 0.417respectively. It can be noted that these values are under-estimated because this method is applicable only to lossless resonators and the characteristic frequencies cannot be determined accurately when the resonator's Figure of Merit (M) is less than 5 [12], as in the present case. Hence, the present method makes use of the measured data over a range of frequency rather than the data at a few pre-determined frequencies.

Using the estimated values of the material parameters, theoretical spectra of the magnitude of impedance and the corresponding phase angle as a function of frequency were calculated using equation (7) and are shown in Figure 3, along with the experimentally measured data for the piezocomposite with 40% ceramic volume fraction. Similar results were observed for a piezocomposite plate with 25% volume fraction as shown in Figure 4. As can be seen from these figures, the theoretical impedance spectra agree well with the measured spectra. This suggests



Figure 3. Variations of (a) magnitude of impedance and (b) phase angle as a function of frequency of a lossy 1–3 piezocomposite plate with 40% ceramic volume fraction. — Experimental spectra;  $- \cdot - \cdot$  Theoretical spectra obtained from equations (7), which account for losses.

that the approximations made in the present analysis are valid in the frequency range studied.

In addition to the thickness-mode resonance, the 1–3 piezocomposites exhibit (i) "radial-mode" resonance, in the low-frequency region, corresponding to the lateral dimensions of the composite plate as a whole and (ii) lateral-mode resonance, in the high-frequency region, corresponding to the lateral size and periodic spacing of the piezoelectric rods in the polymer matrix [4, 18, 19]. In



Figure 4. Variations of (a) magnitude of impedance and (b) phase angle as a function of frequency of a lossy 1–3 piezocomposite plate with 25% ceramic volume fraciton. — Experimental spectra;  $- \cdot - \cdot$  Theoretical spectra obtained from equations (7), which account for losses.

Figures 3 and 4, the large peaks around 250 and 800 kHz correspond to the fundamental and third harmonics of thickness-mode resonance respectively. Small resonance peaks seen in the experimental impedance spectra (Figure 4) below the thickness-mode resonance are due to the "radial-mode" resonance. Since these peaks are very small for the piezocomposite with 40% ceramic volume fraction, they are not seen in Figure 3. The lateral-mode resonance peaks appear at a much higher frequencies and are not shown in the figures. However, only the

thickness-mode resonance is observed in the theoretical spectra as shown in Figures 3 and 4, since the present model takes into account only the thickness-mode vibrations of the composite plates, which is considered useful for any practical application.

# 5. CONCLUSIONS

A non-linear regression method has been proposed to estimate the material parameters of 1–3 piezocomposite plates. Electrical impedance equation of a lossy 1–3 piezocomposite plate vibrating in the thickness-mode was arrived at by incorporating the complex material coefficients and was used for least-squares fitting the experimental data. The material parameters evaluated by the present analysis are comparable with those obtained from the constituent relations. In addition to dielectric constant, coupling coefficient and stiffness coefficient, the loss factor can also be determined by the present method. The refined values of the parameters are not affected by the variations in the initial guess. Agreement between the theoretical impedance spectra, obtained using the refined material parameters, and the measured spectra suggests that the approximations made in the present analysis are valid in the frequency range studied. This method has the advantage that it does not depend on the measured values of a few characteristic frequencies, as they cannot be determined accurately in the case of lossy resonators.

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