# Evaluation of the Material Parameters of Piezoelectric Materials by Various Methods

Kin Wing Kwok, Helen Lai Wah Chan, and Chung Loong Choy

*Abstract*—**The elastic, dielectric and piezoelectric constants of four piezoelectric materials, including polyvinylidene fluoride, vinylidene fluoride-trifluoroethylene copolymer, PZT/epoxy 1-3 composite, and lead metaniobate ceramic, have been evaluated from the impedance data using five different methods. A method described in ANSI/IEEE Std. 176-1987, though based on formulae derived for lossless materials, is found to be applicable to materials with moderate loss. However, for high-loss materials such as polyvinylidene fluoride, the electromechanical coupling constant ($k_t$) obtained by the method of Std. 176 is substantially higher than the actual value. Calculations based on a piezoelectric resonance analysis program (PRAP) combine the best features of two earlier methods. In addition to the impedance at the parallel resonance frequency, impedances at two other frequencies are required for calculation. The PRAP method gives quite accurate material parameters regardless of the magnitude of the loss, but the parameters (including $k_t$) vary by as much as 15% depending on the choice of data. In the nonlinear regression method described in the present work, all the impedance data points around the resonance are least-squares fitted to the theoretical expression for the impedance. Besides the advantage of requiring no arbitrary choice of data, the nonlinear regression method can readily take account of the frequency dependence of the dielectric constant.**

## I. Introduction

ULTRASONIC APPLICATIONS of lossy piezoelectric materials such as polyvinylidene fluoride (PVDF), vinylidene-trifluoroethylene copolymers (P(VDF-TrFE)) and ceramic-polymer composites have attracted considerable interest [1]–[8], so a full characterization of these materials becomes very important. However, the procedures recommended in IEEE standards [9], [10] were designed for measuring material constants of low-loss materials, such as lead zirconate titanate (PZT), and thus may not be applicable to lossy piezoelectric materials. Several workers, including Smits [11], [12], Sherrit *et al.* [13], Xu *et al.* [14], Ohigashi [15], and Koga and Ohigashi [16], have introduced methods to determine the mechanical, dielectric, and/or piezoelectric loss. Smits [11], [12] and Sherrit *et al.* [13], considered all the material parameters as complex, and the losses are given by the corresponding imaginary parts. The method of Xu *et al.* [14] is similar to that of Smits [11], [12]. Ohigashi [15] treated the elastic and dielectric constants as complex but the piezoelectric constant as real. In this paper, the material parameters of PVDF, P(VDF-TrFE), PZT/epoxy 1-3 composite, and lead metaniobate are calculated from impedance data using five methods: the IEEE Std. 176 method, the methods of Smits [11], [12] and Sherrit *et al.* [13], a software package "Piezoelectric Resonance Analysis Program (PRAP)," and a nonlinear regression method proposed by the authors. By comparing the results, it is hoped that the best method can be found for evaluating the elastic, dielectric, and piezoelectric constants of lossy piezoelectric materials.

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## II. Experiments

The samples characterized include PVDF, P(VDF-TrFE), PZT7A/Araldite D 1-3 composite, and lead metaniobate ceramic. The PVDF sample (Kynar Piezo Film) was supplied by Atochem North America (now Amp Sensors, Valley Forge, PA). The P(VDF-TrFE) sample, containing 25 mole % of TrFE, was poled at the Institut Franco-Allemand de Recherches de Saint-Louis (ISL) in Saint-Louis, France. The 1-3 composite, containing 43 volume % of PZT-7A, was prepared in the Materials Physics Research Laboratory of Macquarie University in Sydney, Australia. Lead metaniobate ceramic was supplied by American Piezo Ceramics, Inc., Mackeyville, PA. The density, diameter, and thickness of the samples, as well as the type and thickness of the electrodes, are given in Table I. One end of a thin wire was bonded to the center of the sample with silver paste and the other end was connected to a HP4194A impedance analyzer. A personal computer (486DX-50) was used to collect the impedance data and to calculate the material parameters following various methods.

## III. Review of the Methods Used for Evaluating Material Parameters

Consider a lossless transducer plate of thickness $L$ and electrode area $A$. If it is poled in the thickness direction and its lateral dimensions are large compared with its thickness, the electrical impedance at thickness-extensional resonance is given by (14) in [10], and (105) in [17]:

$$Z(f) = \frac{L}{i2\pi f \varepsilon_{33}^{S} A}\left[1 - k_t^2 \frac{\tan\left(\pi f L \sqrt{\frac{\rho}{c_{33}^{D}}}\right)}{\pi f L \sqrt{\frac{\rho}{c_{33}^{D}}}}\right] \quad (1)$$

TABLE I
DENSITIES AND DIMENSIONS OF THE SAMPLES AND THE TYPE AND THICKNESS OF THE ELECTRODES.

| | PVDF | P(VDF-TrFE) | 1-3 PZT/Epoxy Composite | Lead Metaniobate |
|---|---|---|---|---|
| Density (kg/m$^3$) | 1.78 | 1.88 | 3.90 | 6.0 |
| Thickness (mm) | 0.270 | 0.408 | 1.06 | 1.55 |
| Diameter (mm) | 14 | 14 | 14 | 25.2 |
| Type of Electrode | Aluminum | Gold | Gold | Silver |
| Thickness of Electrode ($\mu$m) | $< 0.1$ | $< 0.1$ | $< 0.1$ | $< 10$ |

where

$$k_t^2 = \frac{e_{33}^2}{\varepsilon_{33}^S c_{33}^D} \tag{2}$$

$$c_{33}^D = c_{33}^E/(1 - k_t^2), \tag{3}$$

$\varepsilon_{33}^S$ is the clamped dielectric permittivity, $k_t$ is the electromechanical coupling constant for the thickness mode [given by the positive square root of (2)], $c_{33}^D$ is the elastic stiffness constant at constant electric displacement, $c_{33}^E$ is the elastic stiffness constant at constant electric field, $e_{33}$ is the piezoelectric constant, $\rho$ is the density, and $f$ is the frequency. To account for the losses, we will need to treat the material parameters $c_{33}^D$, $\varepsilon_{33}^S$, and $k_t$ as complex quantities by adding an asterisk as the superscript, e.g., $c_{33}^{D*}$, $\varepsilon_{33}^{S*}$, and $k_t^*$. The complex material parameters are assumed to be independent of frequency. It is clear from (1) that impedances $Z(f)$ (both the magnitude and phase) at only three different frequencies are required for calculating the three complex material parameters.

### A. The Method of IEEE Std. 176-1987

According to IEEE Std. 176 [(161) and (163) in [9]] and (3), $c_{33}^D$ and $k_t$ can be determined using the following equations:

$$c_{33}^D = 4\rho L^2 f_p^2 \tag{4}$$

$$k_t^2 = \frac{\pi}{2}\frac{f_s}{f_p}\tan\left(\frac{\pi}{2}\frac{f_p - f_s}{f_p}\right) \tag{5}$$

where the parallel and series resonance frequencies, $f_p$ and $f_s$, are the frequencies at which the real parts of the impedance $Z$ and the admittance $Y$ have a maximum, respectively. For low loss materials, it was noted [9] that $f_p$ is very close to the frequency, $f_n$, at which the impedance magnitude has a maximum, while $f_s$ is very close to the frequency, $f_m$, at which the impedance magnitude has a minimum (i.e., the admittance magnitude, $|Y|$, has a maximum). Since the measurements of $f_n$ and $f_m$ were simpler than those of $f_p$ and $f_s$, it was proposed [9] to make the approximations $f_n = f_p$ and $f_m = f_s$ when considering low loss materials, e.g., PZT.

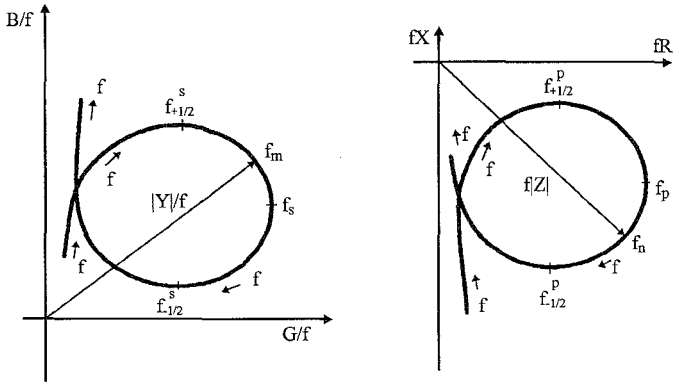


Fig. 1. Schematic diagram showing the definition of the critical frequencies. $Y$ is admittance, $G$ is conductance, $B$ is susceptance, $Z$ is impedance, $R$ is resistance, and $X$ is reactance.

### B. Method by Sherrit et al. [13]

Sherrit *et al.* [13] defined $f_p$ and $f_s$ as the frequencies which correspond to the maxima in the real parts of $fZ(f)$ and $Y(f)/f$, respectively, instead of in the real parts of $Z(f)$ and $Y(f)$ as in IEEE Std. 176. These definitions are more appropriate since $f_p$, for example, is the frequency at which the expression in the square bracket in (1) has a maximum in its real part. Land *et al.* [18] have shown that the bandwidth at resonance, defined as the difference between the frequency at maximum reactance and the frequency at minimum reactance, is related to the mechanical $Q$ factor (i.e., the mechanical loss) of a resonator. Therefore, to account for losses, Sherrit *et al.* [13] defined the complex parallel and series resonance frequencies as:

$$f_p^* = f_p\left[1 - i\frac{f_{-1/2}^p - f_{+1/2}^p}{f_p}\right]^{-1/2} \tag{6}$$

$$f_s^* = f_s\left[1 - i\frac{f_{-1/2}^s - f_{+1/2}^s}{f_s}\right]^{-1/2} \tag{7}$$

where the various frequencies are shown in Fig. 1. After calculating $f_p^*$ and $f_s^*$ from the observed frequencies ($f_p$, $f_{-1/2}^p$, $f_{+1/2}^p$, $f_s$, $f_{-1/2}^s$, $f_{+1/2}^s$), $c_{33}^{D*}$, and $k_t^*$ can be evaluated using (4) and (5) by replacing $f_p$ and $f_s$ by $f_p^*$ and $f_s^*$. Then $\varepsilon_{33}^{s*}$ is calculated by substituting the calculated $c_{33}^{D*}$ and $k_t^*$, and the observed $Z$ value at frequency $2f_p$ in (1).

For materials with low $k_t$ ($k_t < 0.3$), the difference between $f_p$ and $f_s$ is small, so the shifting of the critical frequencies due to a complex $\varepsilon_{33}^*$ may introduce significant errors in the calculated values of $c_{33}^{D*}$ and $k_t^*$ [13]. Therefore, for materials with low $k_t$ or large dielectric loss, they eliminated this effect by obtaining the critical frequencies from $\varepsilon_{33}^{s*} f Z(f)$ and $Y(f)/(\varepsilon_{33}^{s*} f)$ instead of $fZ(f)$ and $Y(f)/f$ after calculating $\varepsilon_{33}^{s*}$ by the above procedure.

### C. Smits' Method

In the framework of the Smits' method [11], [12], an initial value of $c_{33}^{D*}$ is estimated. This value and the observed $Z(f)$ at any two frequencies near the resonance are substituted into (1) to obtain $\varepsilon_{33}^{s*}$ and $k_t^*$. By using these calculated $\varepsilon_{33}^{s*}$ and $k_t^*$ values and an observed $Z(f)$ value at a third frequency, an improved $c_{33}^{D*}$ value is obtained from (1). This procedure is repeated (with the same three chosen $Z(f)$ values) until the calculated $c_{33}^{D*}$ differs from that obtained in the previous step by less than a cut-off value.

However, it was found [19] that the calculated results are very sensitive to the initial value of $c_{33}^{D*}$ and the $Z(f)$ values chosen for the calculation. First, not any three $Z(f)$ data points can give convergent results. Second, even if there is convergence, the resulting parameters depend on the initial $c_{33}^{D*}$ value and the chosen set of $Z(f)$. Although a reasonably accurate initial value of $c_{33}^{D*}$ can be easily calculated [11], it is still necessary to make the correct choice of $Z(f)$ data to obtain convergent results, so the method is not suitable for routine use by an inexperienced operator. In the present work, Smits' method [11], [12] is modified. Instead of using any three $Z(f)$ values, we search for three values in the frequency range of $3(f_n - f_m)$ centered at $f_o$ ($f_o = (f_m + f_n)/2$) in order to obtain best-fit results. This is the guideline used for any material and thickness. The width of this frequency range is different for different materials: about 790 kHz for PVDF, 450 kHz for P(VDF-TrFE), 900 kHz for the 1-3 composite, and 240 kHz for lead metaniobate, but it covers the entire resonance peak. Based on experience, we choose one data point at $f_o$, and the other two at arbitrary frequencies $f_1$ and $f_2$ where $f_1 < f_o < f_2$. The complex material parameters as well as the difference between the calculated and measured impedance in the above frequency range are then calculated. About 100 data points are chosen for each $f_1$ and $f_2$, and then $10^4$ iterations are carried out. The computing time is about 8 minutes. However, if we search for any three frequencies instead of two, the computing time is about 200 times longer and is thus not practical.

### D. Piezoelectric Resonance Analysis Program (PRAP)

Recently, the methods of Smits [11], [12] and Sherrit *et al.* [13] have been combined in the PRAP (from TASI Technical Software, Ontario, Canada). In this program the frequency $f_p$ corresponding to the maximum in the real part of $fZ(f)$ is determined automatically from the experimental impedance spectra, and the impedance value at $f_p$ is used as one of the three data points for the calculation. The user is then required to choose arbitrarily the other two impedance values, $Z(f_1')$ and $Z(f_2')$, such that $f_1' < f_p < f_2'$. Following Sherrit *et al.* [13], an initial value for $c_{33}^{D*}$ is calculated from (6) and (4). Using this $c_{33}^{D*}$ value together with $Z(f_1')$ and $Z(f_2')$ , $\varepsilon_{33}^{S*}$ and $k_t^*$ are calculated from (1). Using $Z(f_p)$ and these calculated values of $\varepsilon_{33}^{S*}$ and $k_t^*$, a new $c_{33}^{D*}$ is calculated from (1), and the process is repeated until the values of $c_{33}^{D*}$, $\varepsilon_{33}^{S*}$ and $k_t^*$ converge. Generally, it just takes one to two seconds for the calculation.

### E. The Nonlinear Regression Procedure (Gauss-Newton Method)

The Gauss-Newton method is a regression procedure [20] widely used to best fit experimental data to a nonlinear equation. We first write $c_{33}^{D*}$, $\varepsilon_{33}^{S*}$, $k_t^*$ as

$$c_{33}^{D*} = c_{33}^{D}(1 + i\tan\delta_m) \tag{8}$$

$$\varepsilon_{33}^{S*} = \varepsilon_{33}^{S}(1 - i\tan\delta_e) \tag{9}$$

$$k_t^* = k_t(1 + i\tan\delta_k), \tag{10}$$

where $\tan\delta_m$, $\tan\delta_e$, and $\tan\delta_k$ are the elastic, dielectric, and electromechanical coupling factor loss tangent, respectively. All the material parameters are assumed to be independent of frequency in the vicinity of the resonance. The impedance of the thin plate (1) then becomes:

$$Z(f) = R(f) + iX(f) \tag{11}$$

where the resistance, $R$, is

$$R(f) = \frac{L\tan\delta_e}{2\pi f\varepsilon_{33}^{S}A(1+\tan^2\delta_e)} + \frac{Lk_t^2}{2\pi f\varepsilon_{33}^{S}A}\frac{T-U}{S} \tag{12}$$

and the reactance, $X$, is

$$X(f) = \frac{-L}{2\pi f\varepsilon_{33}^{S}A(1+\tan^2\delta_e)} + \frac{Lk_t^2}{2\pi f\varepsilon_{33}^{S}A}\frac{F+G}{S}, \tag{13}$$

and

$$T = [\tanh N\sec^2 M(1-\tan^2\delta_k) - 2\tan\delta_k\tan M\,\mathrm{sech}^2 N](M - N\tan\delta_e) \tag{14}$$

$$U = [\tan M\,\mathrm{sec}\,h^2 N(1-\tan^2\delta_k) + 2\tan\delta_k\tanh N\sec^2 M](N + M\tan\delta_e) \tag{15}$$

$$F = [\tanh N\sec^2 M(1-\tan^2\delta_k) - 2\tan\delta_k\tan M\,\mathrm{sech}^2 N](N + M\tan\delta_e) \tag{16}$$

$$G = [\tan M\,\mathrm{sech}^2 N(1-\tan^2\delta_k) + 2\tan\delta_k\tanh N\sec^2 M](M - N\tan\delta_e) \tag{17}$$

$$S = (M^2+N^2)(1+\tan^2\delta_e)(1+\tan^2 M\tanh^2 N) \tag{18}$$

$$M = \pi f L\sqrt{\frac{\rho}{c_{33}^{D}}}\left(1 - \frac{3}{8}\tan^2\delta_m\right) \tag{19}$$

$$N = \frac{\pi f L\tan\delta_m}{2}\sqrt{\frac{\rho}{c_{33}^{D}}}\left(1 - \frac{5}{16}\tan^2\delta_m\right) \tag{20}$$

Since the mechanical loss is not too high, terms of $\tan\delta_m$ of order higher than three are neglected in (19) and (20). The magnitude of impedance ($|Z|$) and the phase angle ($\phi$) are given by:

$$|Z(f)| = \sqrt{R^2(f) + X^2(f)} \tag{21}$$

$$\phi(f) = \tan^{-1}\left(\frac{X(f)}{R(f)}\right). \tag{22}$$

It is seen from (1) that the impedance consists of two components: the baseline (first term) and the resonance (second term). The baseline term, $Z_o$, is:

$$Z_o(f) = \frac{L}{i2\pi f\varepsilon_{33}^S A(1 - i\tan\delta_e)}, \tag{23}$$

and the corresponding magnitude, $|Z_o|$, and phase angle, $\phi_o$, are given by:

$$|Z_o(f)| = \frac{L}{2\pi f\varepsilon_{33}^S A\sqrt{1 + \tan^2\delta_e}} \tag{24}$$

$$\tan\phi_o(f) = -\frac{1}{\tan\delta_e}. \tag{25}$$

The $\phi_o$ depends only on $\tan\delta_e$ while $|Z_o|$ is weakly dependent on $\tan\delta_e$. Equations (1) and (4) show that the resonance term equals zero at $f = 2f_p$, so the procedure uses an estimated initial value of $\tan\delta_e$ calculated from the observed $\phi$ value at $f = 2f_p$. Then the nonlinear regression procedure (Gauss-Newton method) [20] is applied to fit the observed $|Z(f)|$ to (21) to obtain the other material parameters: $c_{33}^D$, $\varepsilon_{33}^S$, $k_t$, $\tan\delta_m$, and $\tan\delta_k$. With the use of the calculated $c_{33}^D$, $\varepsilon_{33}^S$, $k_t$, $\tan\delta_m$, and $\tan\delta_k$, the observed $\phi(f)$ are fitted to (22) to obtain an improved $\tan\delta_e$ value. The process is repeated until converging values are obtained. A program has been written to perform the above procedure using 200 data points. After the acquisition of data, it takes about 1 minute to complete the calculation.

In general, all of the six parameters depend on frequency. Therefore, each calculated material parameter represents an average within the frequency range where data points are used for the calculation. The narrower the fitting range, the closer are the calculated average values to the actual values. In the present work, data in the frequency range of $3(f_n - f_m)$ centered at $f_o$ are used for calculation.

## IV. Results and Discussion

The material parameters of the samples evaluated using the previously described methods and the piezoelectric constant $e_{33}$ calculated from (2) are given in Table II. In taking the square root of (2), the minus sign of $e_{33}$ is taken for PVDF and P(VDF-TrFE) because of the 180° phase difference between the excitation and response in the piezoelectric effect, while the positive sign is taken for PZT/epoxy 1-3 composite and lead metaniobate.

It should be noted that the impedance and resonance frequency are not sensitive to variation in $\tan\delta_k$ (and hence $\tan\delta_p$) in the range $-0.1$ to 0.1, so the different $\tan\delta_k$ and $\tan\delta_p$ values obtained using different methods are not meaningful. In fact, using PVDF as an example, a good fit to the impedance data can be obtained using the nonlinear regression method and taking $\tan\delta_k = 0$.

Except for the method of IEEE Std. 176, the other four sets of measured parameters were used to generate the impedance magnitude and phase angle as functions of frequency using (1). The experimental data and the generated curves are shown in Figs. 2 to 5. The results are discussed in the following sections.

### A. Method of IEEE Std. 176

Since only the real parameters $k_t$ and $c_{33}^D$ can be determined using the IEEE Std. 176 method, it is not possible to calculate the impedance spectra for comparison with experimental data. As the parallel and series resonance frequencies, $f_p$ and $f_s$, can be directly measured (as the frequencies at maximum resistance and conductance, respectively) using an impedance analyzer, it is not necessary to make the approximations $f_n = f_p$ and $f_m = f_s$. Since (4) and (5) used for calculating $c_{33}^D$ and $k_t$ from $f_p$ and $f_s$ are derived from (1) for lossless materials, they are not valid for lossy materials. For lossy materials, $f_p$ should correspond to the maximum value of (12), and $f_s$ should correspond to the maximum value of the real part of the reciprocal of (11). It is then obvious that the two equations involving $f_p$ and $f_s$ would include all six material parameters, instead of only $c_{33}^D$ and $k_t$. These equations reduce to (4) and (5) only when the losses are negligible. It is seen from Table II that the calculated $k_t$ and $c_{33}^D$ of P(VDF-TrFE), PZT/epoxy 1-3 composite, and lead metaniobate are very close to the real parts of $k_t^*$ and $c_{33}^{D*}$ evaluated by our method. This is not surprising because these three materials do not exhibit very strong mechanical and dielectric loss in the frequency range under study. For PVDF which shows a significant loss, the IEEE Std. 176 value of $c_{33}^D$ is close to that obtained by our method, but the $k_t$ value is substantially higher. Therefore, if the approximations $f_n = f_p$ and $f_m = f_s$ are not used, the IEEE Std. 176 method is applicable to piezoelectric materials with moderate loss as has been well-known and accepted.

In practice, the resonance frequencies $f_p$ and $f_s$ are often approximated by $f_n$ and $f_m$, respectively, and Table III shows the results of this approximation. The values of $k_t$ would be overestimated, especially for materials with high $\tan\delta_m$ (low $Q_m$), such as PVDF, P(VDF-TrFE), and lead metaniobate. For the material with very low $\tan\delta_m$ (high $Q_m$), PZT/epoxy 1-3 composite, the overestimation is not significant. The discrepancy in $k_t$ arises largely from the difference between $(f_p - f_s)$ and $(f_n - f_m)$. The higher the mechanical loss, the larger the difference between $(f_p - f_s)$ and $(f_n - f_m)$ and the larger the error in the electromechanical coupling constant $k_t$ [see (5)]. For example, the values of $f_p$, $f_s$, $f_n$, $f_m$ are 4.125, 4.075, 4.225, 3.962 MHz

TABLE II

MATERIAL CONSTANTS OF FOUR SAMPLES DETERMINED BY DIFFERENT METHODS. THE RESULTS OF IEEE STD. 176 METHOD ARE OBTAINED USING $f_p$ AND $f_S$. THE PIEZOELECTRIC CONSTANT $e_{33}^*(= e_{33}(1 + i \tan \delta_p))$ WAS CALCULATED ACCORDING TO (2).

| | Method | $k_t$ | $\tan \delta_k$ | $\varepsilon_{33}^S/\varepsilon_o$ | $\tan \delta_e$ | $c_{33}^D$ (GPa) | $\tan \delta_m$ | $e_{33}$ (C/m$^2$) | $\tan \delta_p$ |
|---|---|---|---|---|---|---|---|---|---|
| PVDF | Present work | 0.146 | 0.0362 | 6.3 | 0.256 | 8.7 | 0.133 | −0.104 | −0.023 |
| | IEEE Std. 176 | 0.171 | – | – | – | 8.9 | – | – | – |
| | Sherrit | 0.087 | −0.0325 | 5.5 | 0.309 | 8.5 | 0.134 | −0.057 | −0.116 |
| | Smits | 0.147 | 0.079 | 6.3 | 0.255 | 8.6 | 0.137 | −0.104 | 0.022 |
| | PRAP | 0.145 | 0.091 | 6.3 | 0.256 | 8.6 | 0.135 | −0.103 | 0.033 |
| P(VDF-TrFE) | Present work | 0.262 | 0.0143 | 4.38 | 0.106 | 10.1 | 0.051 | −0.165 | −0.013 |
| | IEEE Std. 176 | 0.266 | – | – | – | 10.1 | – | – | – |
| | Sherrit | 0.260 | 0.0184 | 4.27 | 0.090 | 10.1 | 0.049 | −0.161 | −0.002 |
| | Smits | 0.253 | 0.0261 | 4.27 | 0.090 | 10.1 | 0.051 | −0.157 | 0.007 |
| | PRAP | 0.258 | 0.0413 | 4.27 | 0.102 | 10.1 | 0.050 | −0.160 | 0.015 |
| 1-3 | Present work | 0.58 | −0.0074 | 116 | 0.0420 | 61.3 | 0.0237 | 4.57 | −0.016 |
| PZT-Epoxy | IEEE Std. 176 | 0.58 | – | – | – | 61.4 | – | – | – |
| composite | Sherrit | 0.58 | 0.0087 | 116 | 0.0309 | 61.4 | 0.0233 | 4.57 | 0.005 |
| | Smits | 0.57 | −0.0084 | 116 | 0.0078 | 61.4 | 0.0224 | 4.53 | −0.001 |
| | PRAP | 0.59 | 0.0076 | 120 | 0.0319 | 61.3 | 0.0241 | 4.74 | 0.004 |
| Lead | Present work | 0.334 | 0.0008 | 258 | 0.0089 | 65.8 | 0.063 | 4.10 | 0.028 |
| metaniobate | IEEE Std. 176 | 0.332 | – | – | – | 65.8 | – | – | – |
| | Sherrit | 0.330 | 0.0169 | 259 | 0.0052 | 65.5 | 0.065 | 4.04 | 0.047 |
| | Smits | 0.334 | −0.0023 | 259 | 0.0105 | 65.8 | 0.063 | 4.10 | 0.024 |
| | PRAP | 0.330 | 0.0056 | 255 | 0.0229 | 65.7 | 0.063 | 4.02 | 0.026 |

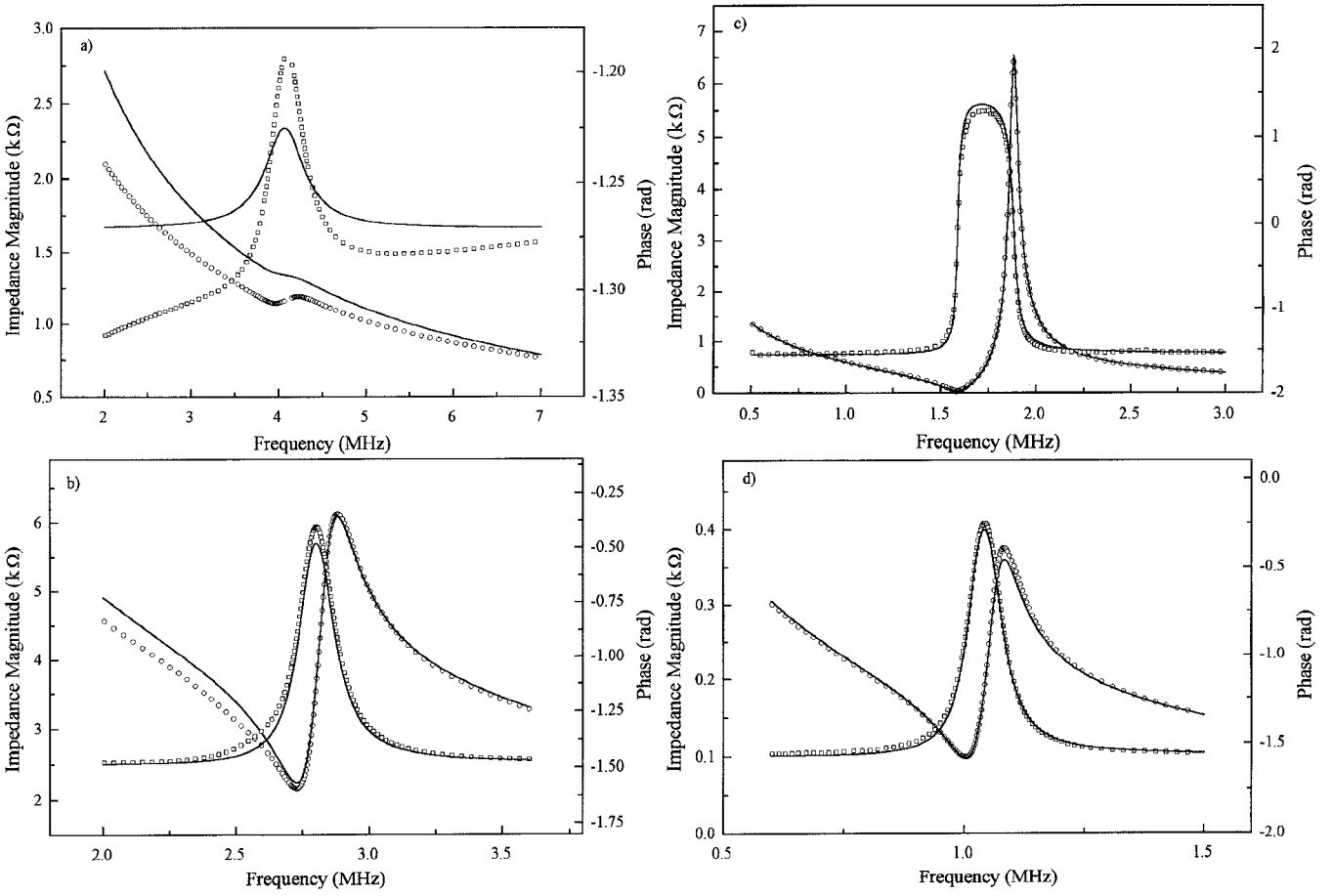


Fig. 2. Comparison of calculated impedance and phase angle (solid lines) with experimental data ($|Z|$, ○; phase angle □): a) PVDF, b) P(VDF-TrFE), c) PZT/epoxy 1-3 composite, d) lead metaniobate. The material parameters used for generating the theoretical spectra are calculated using the method of Sherrit *et al.* [13].

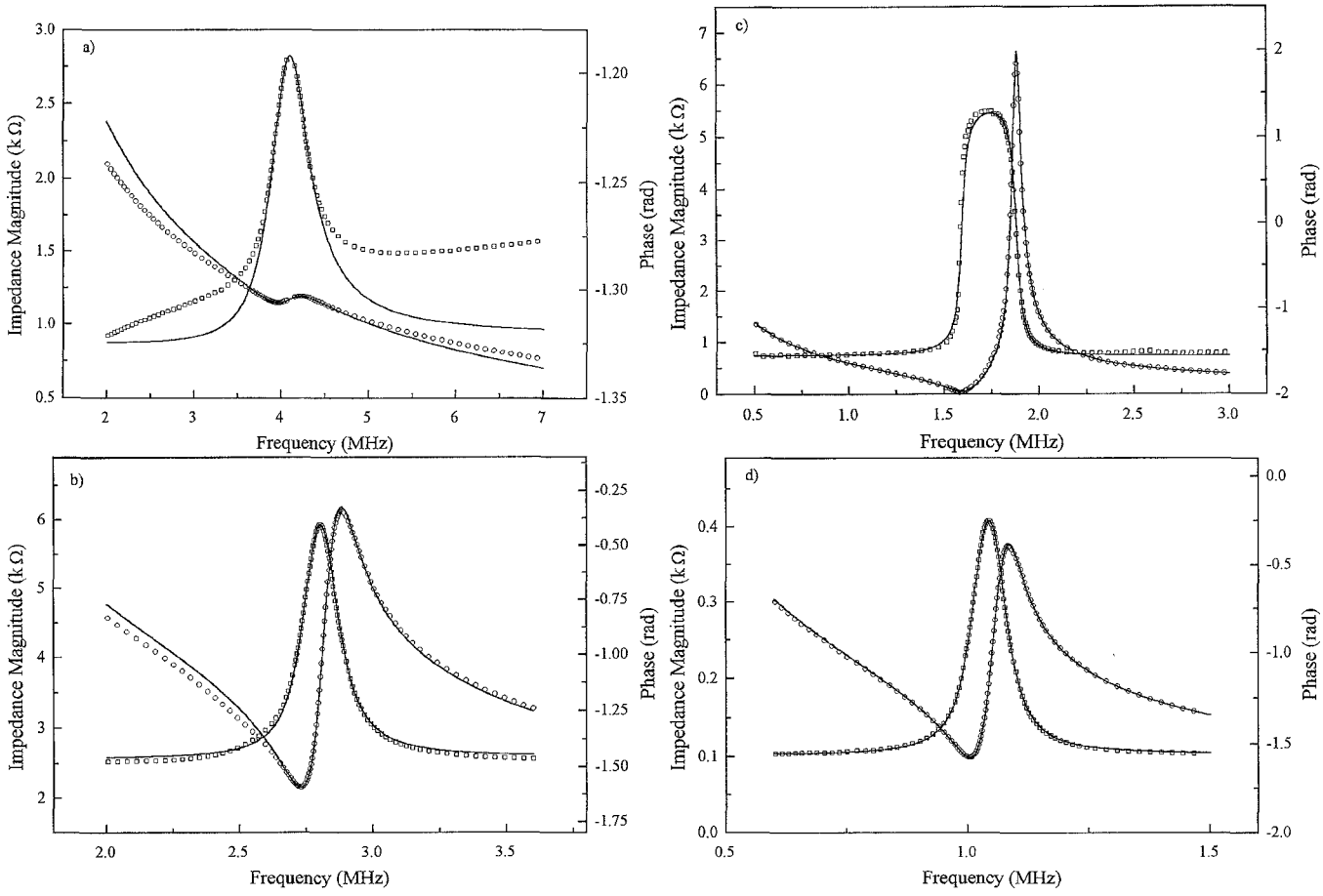


Fig. 3. Comparison of calculated impedance and phase angle (solid lines) with experimental data ($|Z|$, o; phase angle □): a) PVDF, b) P(VDF-TrFE), c) PZT/epoxy 1-3 composite, d) lead metaniobate. The material parameters used for generating the theoretical spectra are calculated using the modified Smits' method.

TABLE III
MATERIAL CONSTANTS OF FOUR SAMPLES EVALUATED BY THE IEEE STD. 176 METHOD USING TWO DIFFERENT PAIRS OF RESONANCE FREQUENCIES.

| | $k_t$ | | $c_{33}^D$ (GPa) | |
|---|---|---|---|---|
| | $f_p$ & $f_s$ | $f_n$ & $f_m$ | $f_p$ & $f_s$ | $f_n$ & $f_m$ |
| PVDF | 0.171 | 0.380 | 8.92 | 9.37 |
| P(VDF-TrFE) | 0.266 | 0.351 | 10.1 | 10.4 |
| 1-3 PZT/Epoxy Composite | 0.575 | 0.581 | 61.4 | 61.4 |
| Lead Metaniobate | 0.332 | 0.414 | 65.8 | 67.8 |

for our PVDF sample, so $(f_p - f_s)$ and $(f_n - f_m)$ are 50 kHz and 263 kHz, respectively.

### *B. Method by Sherrit et al. [13]*

For all samples, the resulting $\varepsilon_{33}^{s*}$ is found to be dependent on which impedance data point is chosen for the calculation. Therefore, we have followed the recommendation of Sherrit *et al.* [13] by calculating $\varepsilon_{33}^{S*}$ from $Z(2f_p)$. For P(VDF-TrFE), PZT/epoxy 1-3 composite, and lead metaniobate, the resulting material parameters agree with those obtained by our method, and there is also good agreement between the calculated and observed impedance spectra [Figs. 2(b) to (d)]. However, for PVDF, the calculated $k_t$, and hence $e_{33}$, deviate appreciably from the value obtained by our method. It seems that the introduction of complex resonance frequencies into (4) and (5) is not sufficient for obtaining accurate material parameters for materials with high loss.

### *C. Smits' Method*

Table II shows that the modified Smits' method gives more accurate $k_t$ and $e_{33}$ values for PVDF than the IEEE Std. 176 method. As shown in Figs. 3(b) to (d) there is good agreement between the calculated and observed spec-

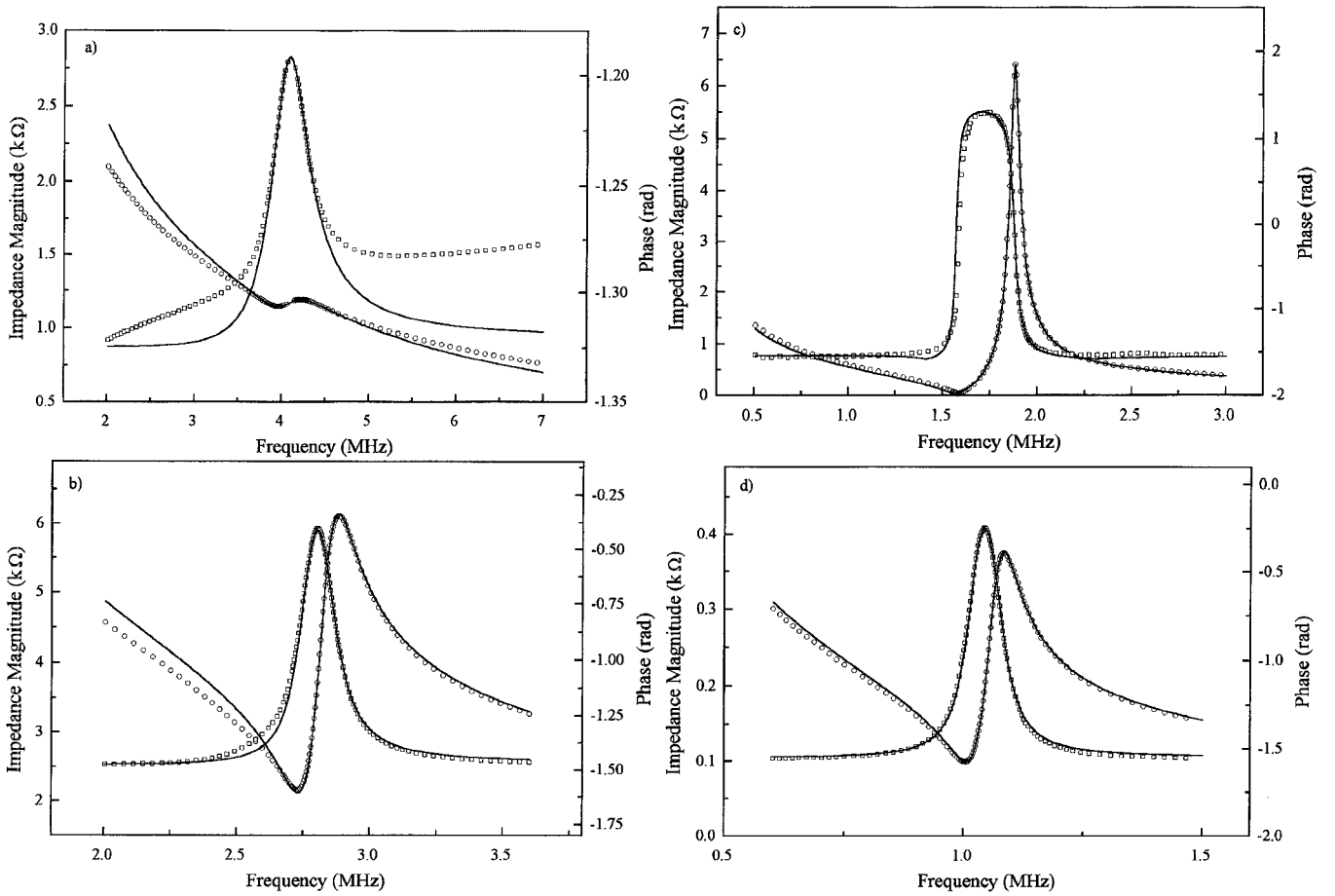


Fig. 4. Comparison of calculated impedance and phase angle (solid lines) with experimental data ($|Z|$, ○; phase angle □): a) PVDF, b) P(VDF-TrFE), c) PZT/epoxy 1-3 composite, d) lead metaniobate. The material parameters used for generating the theoretical spectra are calculated using PRAP.

tra for P(VDF-TrFE), PZT/epoxy 1-3 composite, and lead metaniobate. For PVDF, the agreement is poor outside the resonance region [see Fig. 3(a)], and this is due to the variation of $\varepsilon_{33}^{S*}$ with frequency which will be discussed in a later section.

### *D. Piezoelectric Resonance Analysis Program (PRAP)*

Since this approach is basically the same as that of Smits [11], [12] except that the initial $c_{33}^{D*}$ is estimated by following the method of Sherrit *et al.* [13], it is not surprising that the results [Table II and Figs. 4(a) to (d)] are very close to those obtained from Smits' method. Since the initial value of $c_{33}^{D*}$ is more accurate, the choice of the two impedance data points ($Z(f_1')$ and $Z(f_2')$, $f_1' < f_p < f_2'$) is no longer critical for obtaining convergent results. However, the calculated results are still dependent on the choice of impedance data. If the frequencies $f_1'$ and $f_2'$ are too close to or too far from $f_p$, the resulting $k_t$, $\tan\delta_k$, $\tan\delta_e$, and $\tan\delta_m$ could have an uncertainty of 10% or more. The material parameters listed in Table II were obtained using the impedance data in the optimum frequency range such that $0.2fR(f_p) < fR(f_1'), fR(f_2') < 0.5fR(f_p)$, where $R$ is the resistance.

### *E. Nonlinear Regression Method*

Good agreement is obtained between the calculated and observed impedance spectra for all samples except PVDF [Figs. 5(a) to (d)]. For PVDF, the poor agreement outside the resonance region is due to the variation of $\varepsilon_{33}^{S*}$ with frequency which will be discussed below.

## V. Frequency Dependence of Material Parameters

In the above calculation, all the material parameters are assumed to be independent of frequency. For P(VDF-TrFE), PZT/epoxy 1-3 composite, and lead metaniobate, the good agreement between the calculated and observed impedance spectra indicates that this is a reasonable approximation. For PVDF, however, the discrepancy between the calculated and observed values, particularly outside the resonance region, implies that the frequency dependence of the material parameters has a significant effect on the impedance. In general, all of the six parameters depend on frequency. However, (1) shows that the impedance outside the resonance region is mainly contributed by the

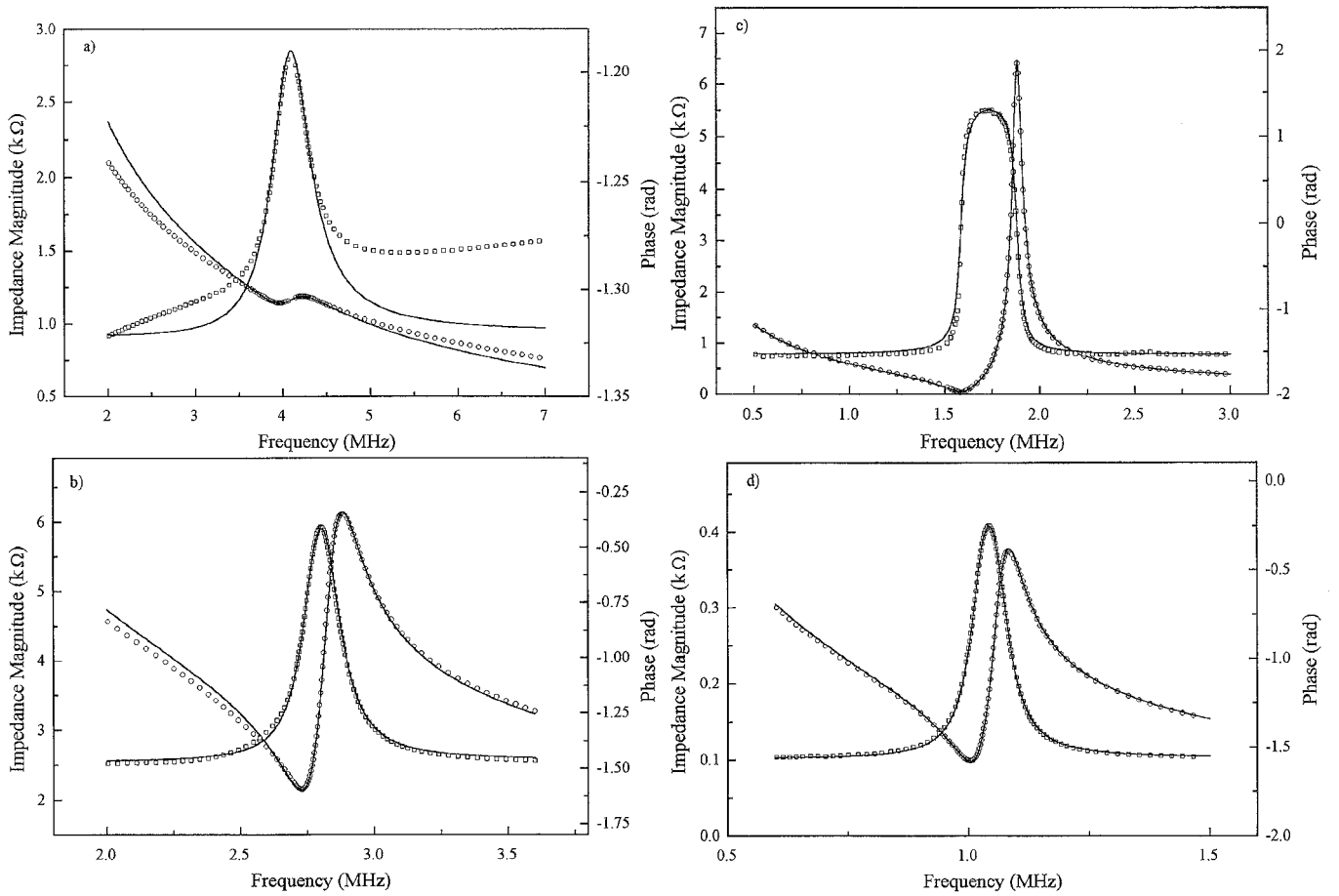


Fig. 5. Comparison of calculated impedance and phase angle (solid lines) with experimental data ($|Z|$, ○; phase angle □): a) PVDF, b) P(VDF-TrFE), c) PZT/epoxy 1-3 composite, d) lead metaniobate. The material parameters used for generating the theoretical spectra are calculated using the nonlinear regression method.

first term so, as a first approximation, we consider only the effect of the frequency dependences of $\varepsilon_{33}^S$ and $\tan\delta_e$.

Using an approach similar to that of Brown and Carlson [21], we insert the values of $c_{33}^D$, $k_t$, $\tan\delta_m$, and $\tan\delta_k$ obtained by the nonlinear regression method assuming frequency-independent parameters (Table II) and the observed $|Z|$ and $\phi$ values into (21) and (22) to calculate $\varepsilon_{33}^S$ and $\tan\delta_e$ at each frequency. Since the input values of $c_{33}^D$, $k_t$, $\tan\delta_m$, and $\tan\delta_k$ are not exactly correct, the calculated $\varepsilon_{33}^S$ and $\tan\delta_e$ values show appreciable scatter in the resonance region. Therefore, we obtain smooth curves by fitting the $\varepsilon_{33}^S$ and $\tan\delta_e$ values outside the resonance region to polynomials of order 9. Fig. 6 shows the resulting $\varepsilon_{33}^S$ and $\tan\delta_e$ as functions of frequency. Using these results, the other four parameters are calculated by fitting the observed impedance data in the frequency range of $3(f_n - f_m)$ centered at $f_o$ to (21) and (22). We have mentioned that it takes 1 minute to calculate the parameters if they are assumed to be frequency-independent. An additional period of 2 minutes is required to perform the steps treating the frequency dependences of $\varepsilon_{33}^S$ and $\tan\delta_e$.

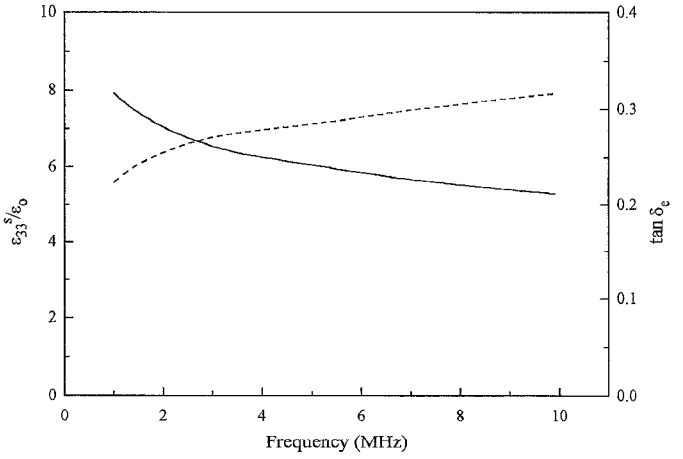


Fig. 6. Variation of $\varepsilon_{33}^S$ (——) and $\tan\delta_e$ (- - - - -) with frequency for PVDF.

Table IV shows that there is 10 to 20% change in the calculated parameters for PVDF (except $c_{33}^D$) when the frequency dependences of $\varepsilon_{33}^S$ and $\tan\delta_e$ are taken into account. With the use of this new set of parameters the calculated impedance spectra are found to agree closely with the observed spectra (Fig. 7). It is necessary to take

TABLE IV

MATERIAL PARAMETERS OF PVDF CALCULATED BY TWO PROCEDURES. IN PROCEDURE 1, ALL THE PARAMETERS ARE ASSUMED TO BE INDEPENDENT OF FREQUENCY. IN PROCEDURE 2, $c_{33}^D$, $k_t$, $\tan\delta_k$, AND $\tan\delta_m$ ARE INDEPENDENT OF FREQUENCY, WHILE $\varepsilon_{33}^S$ AND $\tan\delta_e$ DEPEND ON FREQUENCY. FOR PROCEDURE 2, THE $\varepsilon_{33}^S$ AND $\tan\delta_e$ VALUES GIVEN IN THIS TABLE CORRESPOND TO THOSE AT THE RESONANCE FREQUENCY $f_o$ (= 4.15 MHz). THE PIEZOELECTRIC CONSTANT $e_{33}^*(= e_{33}(1 + i\tan\delta_p))$ WAS CALCULATED ACCORDING TO (2).

| | $k_t$ | $\tan\delta_k$ | $\varepsilon_{33}^S/\varepsilon_o$ | $\tan\delta_e$ | $c_{33}^D$ (GPa) | $\tan\delta_m$ | $e_{33}$ (C/m$^2$) | $\tan\delta_p$ |
|---|---|---|---|---|---|---|---|---|
| Procedure 1 | 0.146 | 0.0362 | 6.3 | 0.256 | 8.7 | 0.133 | −0.104 | −0.023 |
| Procedure 2 | 0.127 | 0.0435 | 6.3 | 0.280 | 8.7 | 0.117 | −0.091 | −0.035 |

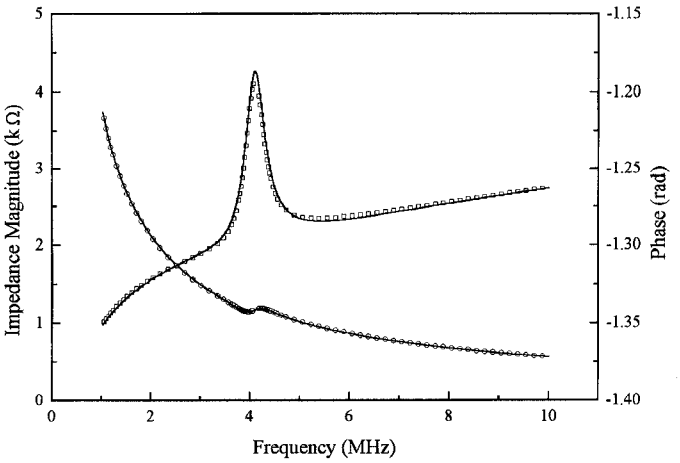


Fig. 7. Comparison of calculated impedance and phase angle (solid lines) with experimental data ($|Z|$, ○; phase angle □)for PVDF. The material parameters used for generating the theoretical spectra are calculated by the nonlinear regression method taking into account the frequency dependence of $\varepsilon_{33}^S$ and $\tan\delta_e$.

account of the frequency dependence of the material parameters because PVDF has a strong relaxation associated with the glass transition within the frequency range of our measurements.

## VI. CONCLUSION

Five different methods have been used to evaluate the material parameters of four piezoelectric materials, including PVDF, P(VDF-TrFE), PZT-7A/Araldite D 1-3 composite, and lead metaniobate ceramic. Although the IEEE Std. 176 method is strictly valid only for lossless materials, we have found that it is applicable to materials with moderate loss, in agreement with generally accepted practice. However, for high-loss materials such as PVDF, the $k_t$ value evaluated using this method is much higher than the value determined by the nonlinear regression method. Sherrit *et al.* [13] accounted for the losses by treating the critical frequencies in (4) and (5) as complex quantities. Consequently, the loss factors for the material parameters can also be deduced. This method is quite successful for moderately lossy materials, but the resulting $k_t$ and $e_{33}$ for high-loss PVDF are too low.

Smits' method suffers from the fact that the results are strongly dependent on the three $Z(f)$ values chosen for the calculation. The PRAP improves on Smits' method by using a more accurate initial $c_{33}^{D*}$ value calculated by the procedure of Sherrit *et al.* [13]. Consequently, the resulting parameters are less dependent on the chosen $Z(f_1')$ and $Z(f_2')$. However, if frequencies $f_1'$ and $f_2'$ are too close or too far from $f_p$, the resulting material parameters could have an uncertainty of 10% or more. The nonlinear regression method has the advantages that an arbitrary choice of data is not necessary and that all the impedance data points within a certain frequency range are given a best fit to the theoretical expression for the impedance to give the material parameters. Moreover, it is quite convenient to take account of the frequency dependences of $\varepsilon_{33}^S$ and $\tan\delta_e$.

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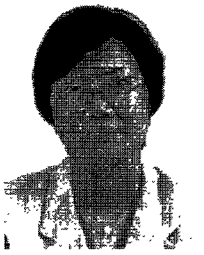

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