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Noniterative method for evaluation of the complex material constants of piezoelectric ceramics in the radial vibration mode

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Abstract

A new noniterative method, for determining the dielectric, piezoelectric and elastic constants, in complex form, for piezoceramic materials, in the radial mode, was proposed.

This method uses the standard procedure to determine the elastic compliance and Poisson factor and the measurement of admittance at two frequencies to calculate the dielectric and piezoelectric constants, by solving a system of two equations.

The accuracy of the new method was determined for materials with different planar coupling coefficients (k_p = 2.5–57%) and mechanical quality factors (Q_m = 20–3000). This method proved to be very accurate for all materials especially for those with large coupling factors. The accuracy of standard method was also evaluated for the same materials.

Keywords: Complex material constants; Piezoelectric properties; Ferroelectric properties; Impedance; Perovskites

1. Introduction

The standard method[1] used to determine the dielectric, piezoelectric and elastic constants of piezoelectric ceramics, in the radial mode, ignores their losses, considering these constants as real quantities. However, the development of impedance spectroscopy performed with impedance analyzers, provides the spectra of the real and imaginary parts of electrical admittance, which can only be explained by taking into account the losses and treating the material constants as complex quantities. The methods used to determine material constants, in complex form, can be classified as iterative[2–4] and noniterative.[5] The iterative ones use the frequency spectra of the electrical admittance, within the range of the resonance–antiresonance of the fundamental mode only, to determine all material constants, while the noniterative method requires some more measurements, away from fundamental resonance, and a set of specific frequencies, in the range of fundamental mode and the first overtone. For radial mode, the noniterative method is valid only for materials with intermediate mechanical quality factors.[5] Iterative methods can be used even for materials with high losses, but they need a judicious choice of the frequencies where admittance is measured. These frequencies have to be calculated before[2,4] or during the iteration process[3] which is a drawback of these methods. In this later case, measurement of new admittance data at calculated frequencies is required, hence data acquisition and iteration process cannot be separated. Despite their high accuracy,[4] iterative methods have another drawback. They require the measurement of the admittance at series resonance frequency, which is performed with large errors for the imaginary part, mostly for materials with high coupling factors.

Recently, a new method for determining material constants in complex form by curve fitting approach has been reported.[6] The fitting approach of the whole admittance spectra was performed by a nonlinear iteration method, in the series resonance band, and the resulting constants were compared to those provided by standard method. A significant mismatch between the two methods was observed for materials with high coupling factors. This method is rather difficult to be applied since it uses some complicated expressions for linearization of the admittance and for solving the systems of equations for the initial guess of the dielectric and piezoelectric constants.

In the present paper, we developed a new noniterative method to determine the complex material constants. It only uses two

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formulas and a system of two equations to calculate the complex material constants. The accuracy of the new method was determined for materials with different planar coupling coefficients (k_p = 2.5–57%) and mechanical quality factors (Q_m = 20–3000).

2. Measurements

The measurement technique consists in generating a radial mode of vibration in a disc shaped piezoceramic resonator, by sinusoidal electrical stimulation and frequency sweep, by means of an HP-4294A impedance analyzer, controlled by a computer.

The real (conductance G) and imaginary (susceptance B) parts of the complex electrical admittance Y are measured as a function of frequency within the resonance band of the fundamental radial mode and these data are stored as input resonance spectra, in order to check only the agreement with output data calculated with the constants provided by this method.

The series resonance frequency f_s corresponding to the maximum of G and the frequencies $f_{B\max}$ and $f_{B\min}$ of maximum and minimum of B around f_s, respectively, are determined from these spectra and stored. Electrical admittance, is also measured at two frequencies f_{1sis} and f_{2sis}, outside the resonance band and the corresponding values Y_{1sis} and Y_{2sis} are stored. The criteria used to select these frequencies will be further discussed. Then, the frequency range is changed to sweep the first overtone band of the radial mode, in order to determine its series resonance frequency f_{s1}.

3. Description of the method

This new method is based on the following expression for the electrical admittance Y, of a disk resonator oscillating in the radial mode under a sinusoidal signal, as a function of frequency f:

$$Y = i\frac{2\pi^2 fa^2}{t}\left[\varepsilon_{33}^T + \frac{2d_{31}^2}{s_{11}^E(1-\sigma)} \times \frac{2 - j_1(z)}{j_1(z)+\sigma-1}\right] \quad (1)$$

The symbols a, t and ρ represent the radius, thickness and density of the sample, respectively and $i = \sqrt{-1}$. The dielectric permittivity at constant stress ε_{33}^T, the piezoelectric constant d_{31}, and the elastic compliance at constant electric field s_{11}^E, are complex parameters. The Poisson's ratio σ is defined as the ratio of the elastic compliances s_{11}^E and s_{12}^E and is considered as a real parameter.

$$\sigma = -\frac{s_{12}^E}{s_{11}^E} \quad (2)$$

The complex function $j_1(z)$ of complex variable z is given by:

$$j_1(z) = \frac{zJ_0(z)}{J_1(z)} \quad (3)$$

J_0 and J_1 are Bessel functions of first kind and zeroth and first order, respectively. The argument z of the function $j_1(z)$ is given by:

$$z = 2\pi af\sqrt{\rho s_{11}^E(1-\sigma^2)} \quad (4)$$

The relationship (1) is derived from Eq. (117) of the IEEE Std.,[1] by substituting the planar coupling factor k_p with the following formula, in order to evidence the explicit contributions of the dielectric, piezoelectric and elastic constants:

$$k_p = \frac{\sqrt{2}d_{31}}{\sqrt{\varepsilon_{33}^T(s_{11}^E + s_{12}^E)}} \quad (5)$$

First, one has to determine two parameters: the Poisson's ratio σ and η which is the first positive root of the equation:

$$j_1(z) + \sigma - 1 = 0 \quad (6)$$

representing the condition of radial resonance in a lossless piezoceramic resonator with all material constants as real quantities, including the function $j_1(z)$. These parameters were calculated by a polynomial fit[5] of the data given in Table 12 of IEEE Std.[1]:

$$\sigma = \sum_{i=0}^{4} a_i r^i \quad (7)$$

$$\eta = \sum_{i=0}^{3} b_i r^i \quad (8)$$

with a_i and b_i given by Table I of Ref. 5 and $r = f_{s1}/f_s$.

The real part of s_{11}^E was calculated according to standard method by the relationship[1]:

$$(s_{11}^E)' = \frac{\eta^2}{\rho(2\pi af_s)^2(1-\sigma^2)} \quad (9)$$

and the imaginary part with:

$$(s_{11}^E)'' = -\frac{\Delta f}{f_s}(s_{11}^E)' \quad (10)$$

where $\Delta f = f_{B\min} - f_{B\max}$.

By substituting the above calculated constants, the admittance values Y_{1sis} and Y_{2sis} with their corresponding frequencies $f_{1\mathrm{sis}}$ and $f_{2\mathrm{sis}}$ into Eq. (1), a system of two equations linear with respect to the constants ε_{33}^T and d_{31}^2 was obtained. By solving it, ε_{33}^T and d_{31} were determined. The frequencies $f_{1,2sis}$, used to calculate the dielectric and piezoelectric constants are symmetrically situated outside the resonance–antiresonance band ($f_{1sis} < f_m < f_n < f_{2sis}$), where f_m and f_n are the frequencies corresponding to maximum and minimum of absolute admittance, respectively, in the fundamental resonance band of the radial mode. The frequencies $f_{1,2sis}$ should be chosen such the absolute impedance/admittance has about the same values at these frequencies. For materials with very low k_p and Q_m, the difference between them ($f_{2sis} - f_{1sis}$) is recommended to be about ten times the difference ($f_n - f_m$), since otherwise they would be to close to f_m and f_n, where the accuracy of the dielectric constant would be decreased. At series resonance, which corresponds to mechanical resonance of the disk, a large amount of input energy is converted to elastic energy, thus allowing an accurate determination of the elastic constant around f_s, to the detriment of dielectric constant. Therefore, it is necessary to determine the dielectric constant far from f_s, where the dielectric energy becomes dominant. This is the reason of choosing the

frequencies $f_{1,2sis}$, near but outside the resonance–antiresonance band.

The algorithm of this method was written in Mathematica 3.0.

4. Results and discussion

The new method was tested on materials with different planar coupling factors (k_p = 2.5–57%) and mechanical quality factors (Q_m = 20–3000). Their experimental resonance spectra were simulated by giving complex values to material constants ε_{33}^T, d_{31}, s_{11}^E and σ, considered as input constants into Eq. (1). The new method was then applied to the generated admittance data in order to check the agreement between calculated and input material constants.

The errors for the real and imaginary parts of material constants were calculated by the following relationships:

$$\varepsilon_{const'} = \frac{\left|const_i' - const_c'\right|}{\left|const_i'\right|} \tag{11}$$

and

$$\varepsilon_{const''} = \frac{\left|const_i'' - const_c''\right|}{\left|const_i''\right|} \tag{12}$$

where subscripts "*i*" and "*c*" designate the input and calculated constants, respectively and single and double primes signify the real and imaginary parts, respectively.

Table 1 shows the input constants for eleven materials and their input k_p and Q_m calculated with input constants by formula (5) and the following one, respectively:

$$Q_m = \left|\frac{(s_{11}^E)'}{(s_{11}^E)''}\right| \tag{13}$$

In order to evaluate the accuracy of our method, it is required to check for the agreement between experimental and calculated resonance profiles, but also to determine the errors of all material constants provided by the method.

In this regard, Table 2 shows the errors of the four material constants, of the radial mode, determined by the new method, for investigated materials. It also shows the errors of other material constants related to them: the coupling factors k_p and k_{31}, the piezoelectric constant g_{31} and the elastic compliances s_{11}^D and $s_{12}^{E,D}$ at constant E (electric field) and constant D (dielectric displacement), which were subsequently calculated by the previous constants.

Table 2 proves that our new method is very accurate for materials 1–3 with large coupling factors (the errors for material constants are below 0.1%). For materials 4–7 with low k_p and Q_m, the errors were lower then 0.5% and 2% for the real and imaginary parts of material constants, respectively. Similar results were obtained for materials 10 and 11 with very low k_p and high Q_m. For materials 8 and 9 with very low k_p and Q_m errors less then 1–3% were obtained for the real parts of the constants and large errors up to 60% for their imaginary parts. However, by substituting Δf in formula (10) with Δf_{hb} (the half band width of the conductance spectrum around series resonance), the errors of the imaginary parts significantly decreased to 1–3%, except for d_{31}, for which they reduced to 5–20%. Same errors resulted, for these two materials, by fitting approach of the experimental data with formula (1) in the same conditions ($(s_{11}^E)''$ calculated with Δf_{hb}). For the rest of the constants, indirectly determined by this method, similar errors were obtained. For material 11, with very high Q_m, larger errors of 7% resulted for the imaginary parts of the coupling factors, since they are more than three orders of magnitude lower than the real parts and therefore can be ignored.

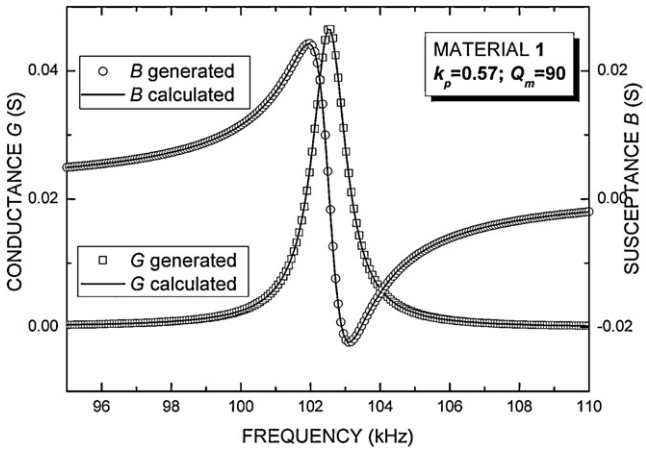

Fig. 1. Generated and calculated G and B versus frequency around the series resonance of the radial mode of material 1.

Figs. 1–4 show the real and imaginary parts of the generated and calculated admittance data versus frequency, in the series resonance band of the fundamental radial mode, for materials 1, 6, 8 and 11, representing the four groups of materials mentioned in Table 2. The resonance spectra were generated for disc shaped resonators of 20 mm in diameter and 1 mm thickness.

One can see the very good agreement between the generated and calculated admittance spectra, for all materials except for material 8 with $(s_{11}^E)''$ calculated with Δf (Fig. 3a). In this case the mismatch between input and output spectra is the result of large errors of the imaginary parts of the material constants

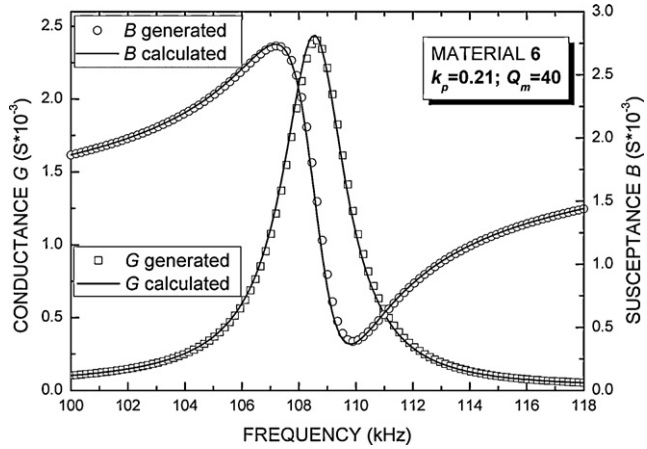

Fig. 2. Generated and calculated G and B versus frequency around the series resonance of the radial mode of material 6.

Table 1
Input constants of materials 1–11.

Materials	Input constants							
	$\varepsilon_{33}^T/\varepsilon_0$	d_{31} (10^{-12} C/N)	s_{11}^E (10^{-12} m^2/N)	σ	k_p	Q_m	$f_s/\Delta f$	ρ (kg/m^3)
1. $k_p = 0.56$ $Q_m = 90$	$1200 - i17$	$-130 + i3$	$15 - i0.17$	0.32	$0.56 - i0.0058$	88	88	7600
2. $k_p = 0.60$ $Q_m = 600$	$1400 - i3$	$-138 + i0.3$	$12.7 - i0.02$	0.30	$0.59 - i0.0002$	635	635	7600
3. $k_p = 0.57$ $Q_m = 1200$	$1100 - i2.7$	$-120 + i0.2$	$13 - i0.011$	0.31	$0.57 - i9 \times 10^{-6}$	1182	1182	7600
4. $k_p = 0.10$ $Q_m = 50$	$800 - i15$	$-20.1 + i1.8$	$14 - i0.28$	0.32	$0.10 - i0.0077$	50	51	7550
5. $k_p = 0.18$ $Q_m = 34$	$915 - i6.3$	$-32.1 + i2.3$	$12 - i0.35$	0.35	$0.18 - i0.0097$	34	35	7550
6. $k_p = 0.21$ $Q_m = 40$	$900 - i16$	$-40.1 + i2.3$	$14 - i0.35$	0.35	$0.21 - i0.0076$	40	40	7550
7. $k_p = 0.22$ $Q_m = 50$	$800 - i12$	$-40.1 + i2.3$	$14 - i0.28$	0.35	$0.22 - i0.0089$	50	50	7550
8. $k_p = 0.033$ $Q_m = 30$	$420 - i10$	$-4.6 + i0.02$	$15 - i0.5$	0.33	$0.033 + i0.0008$	30	74	4650
9. $k_p = 0.097$ $Q_m = 18$	$240 - i1.5$	$-12.7 + i0.1$	$20 - i1.1$	0.2	$0.097 + i0.0022$	18	25	5700
10. $k_p = 0.028$ $Q_m = 400$	$140 - i2$	$-2.2 + i0.011$	$12.9 - i0.0322$	0.21	$0.028 + i9 \times 10^{-5}$	400	395	7500
11. $k_p = 0.025$ $Q_m = 2800$	$168 - i0.5$	$-1.72 + i0.002$	$8 - i0.00287$	0.25	$0.026 + i10^{-5}$	2787	2825	7200

previously discussed. The frequency bands Δf and Δf_{hb} are significantly narrower (which means lower mechanical losses) for calculated than for generated spectra. When $(s_{11}^E)''$ is calculated with Δf_{hb}, the errors decrease resulting in a very good agreement between generated and calculated spectra, shown in Fig. 3b.

For materials 8 and 9, with high losses, $(s_{11}^E)''$ as well as the imaginary parts of other elastic constants, is given with large errors, because of the large difference, of about 40–140%, between input Q_m and the ratio $f_s/\Delta f$, as can be seen in Table 1. For the rest of materials, the difference between the two quantities is less than 3%. Sherit et al.[5] explained this by giving a

Table 2
The errors of complex material constants determined by the noniterative method.

Constant		Errors (%)			
		Materials with $k_p > 0.5$ and $Q_m \geq 90$	Materials with $0.1 < k_p < 0.25$ and $Q_m \leq 50$	Materials with $k_p < 0.1$ and $Q_m \leq 30$[a]	Materials with $k_p < 0.1$ and $Q_m \geq 400$
$\varepsilon_{33}^T/\varepsilon_0$	Real	0.02	0.06	0.01	0.01
	Imag.	0.05	0.6	5–28 (1)	0.1
d_{31}	Real	0.005	0.3	1–3.5 (0.05)	0.15
	Imag.	0.025	0.3–1	16–40(5–20)	0.5–3
s_{11}^E	Real	0.02	0.2	0.3	0.002
	Imag.	0.01	2	30–60(1)	1.5
σ	Real	0.01	0.006	0.6	0.08
	Imag.				
k_p	Real	0.004	0.3	3.4(0.03)	0.15
	Imag.	0.8	0.3–2	30(3)	0.1–7
s_{12}^E	Real	0.03	0.2	0.9	0.08
	Imag.	0.02	0.5–2.5	30–60(1)	1.5
s_{11}^D	Real	0.02	0.2	0.3	0.002
	Imag.	0.02	0.6–2.5	30–60(1)	1.5
s_{12}^D	Real	0.03	0.2	1	0.08
	Imag.	0.02	0.5–2	30–60(1)	1.5
k_{31}	Real	0.004	0.3	3(0.2)	0.1
	Imag.	1	0.4–2	30(3)	0.1–7
g_{31}	Real	0.02	0.4	3(0.1)	0.1
	Imag.	0.5	0.5–2	10–100(5–20)	0.2–2

[a] The values in brackets correspond to $(s_{11}^E)''$ calculated with Δf_{hb}.

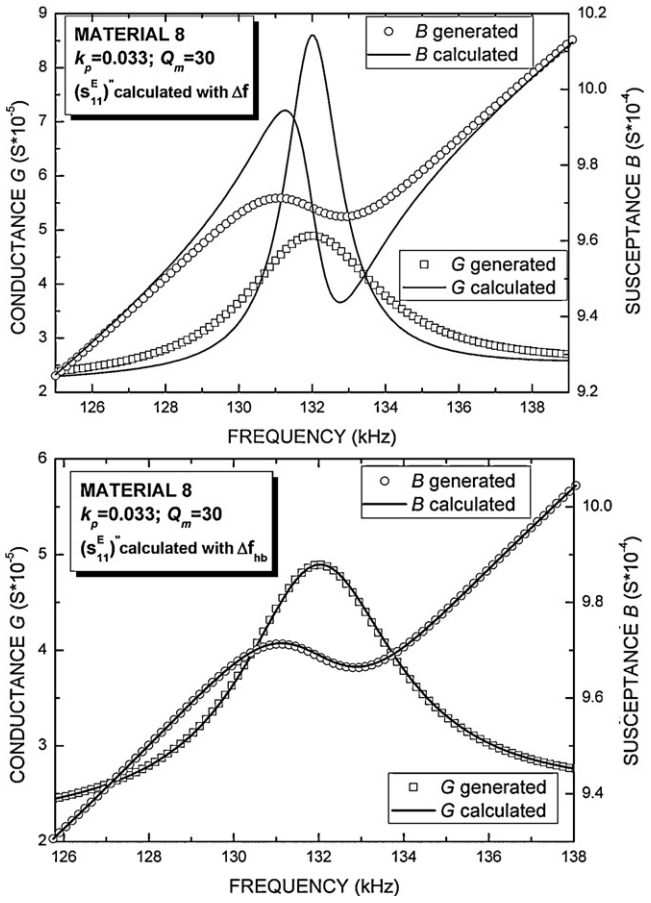

Fig. 3. Generated and calculated G and B versus frequency around the series resonance of the radial mode of material 8 with $(s_{11}^E)''$ calculated with (a) Δf and (b) Δf_{hb}.

new definition of these frequencies. They defined f_s, $f_{B\max}$ and $f_{B\min}$ as the frequencies corresponding to extrema of $G(f)/f$ and $B(f)/f$, respectively. The difference between the two definitions is only significant for materials with very low Q_m. According to Sherrit definition, Q_m and the ratio $f_s/\Delta f$ are almost identical, for

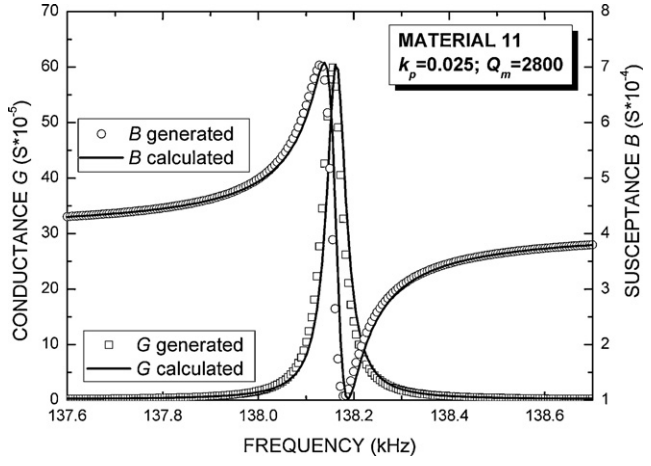

Fig. 4. Generated and calculated G and B versus frequency around the series resonance of the radial mode of material 11.

materials 8 and 9. Moreover, this means that the frequency band width ($f_{B\min} - f_{B\max}$), given by Sherrit definition, corresponds to Δf_{hb} of conductance G, given by the usual definition.

Following the above discussion and taking into account that experimental admittance is measured with errors of about 1%, one may conclude that this method is very accurate provided a good estimation of $(s_{11}^E)''$ is achieved.

The accuracy of standard method used to determine the lossless material constants of the radial mode was also investigated and the errors, calculated with respect to the real parts of input constants, are given in Table 3. Standard method is based on the measurement of the characteristic frequencies f_s, f_p (corresponding to maximum of resistance R) of the fundamental mode and f_{s1} of the first overtone. Constants σ, s_{11}^E, Q_{mst} and k_{pst} were directly calculated with these frequencies, by formulas (7) and (9) and the following ones, respectively:

$$Q_{mst} = \frac{f_s}{\Delta f} \tag{14}$$

$$k_{pst} = \sqrt{\frac{j_1(\eta \cdot f_p/f_s) + \sigma - 1}{j_1(\eta \cdot f_p/f_s) - 2}} \tag{15}$$

The other constants were calculated with the previous ones.

Table 3 shows that standard method is very accurate, with errors less then 1%, for all materials, except for those with very low k_p and Q_m ($k_p < 0.1$ and $Q_m \leq 30$). For such materials, errors of 12–18% resulted for the radial and transversal coupling factors and very large errors of 40–140% for Q_{mst}, but they were drastically reduced to 0.1%, by determining Q_{mst} with Δf_{hb}. As it was already mentioned, constants s_{11}^E and σ determined with high accuracy by standard method were used as the real parts of these constants in our method, that provides materials constants in complex form. The dielectric constant $\varepsilon_{33}^T/\varepsilon_0$ and the piezoelectric constants d_{31} and g_{31} are not included in Table 3, since following standard method, the dielectric constant is determined by measuring the capacitance at low frequencies, far from the radial resonance band, where the admittance spectra (1) are no longer valid, and the piezoelectric constants are calculated with the dielectric constant.

The accuracy of standard method is very useful to be known in order to compare complex constants provided by other methods with those obtained by standard method, since it is not enough to evaluate whether the results are reliable or not, by only checking for the agreement between calculated and experimental spectra.

For example, in Tables 1 and 2 of Ref. 6, complex material constants determined by nonlinear fitting curve approach were compared with those calculated by standard method for two PZT materials with high k_p (0.56 and 0.5) and medium Q_m (700 and 100). Large differences between the constants provided by the two methods were obtained: 14–300% for k_p, 5–16% for Q_m, 0.2–9% for $(s_{11}^E)'$, and 40–500% for $(s_{11}^E)''$, despite the concordance of calculated with experimental resonance profiles. As it is shown in Table 3, the accuracy of standard method is very good for materials with high coupling factors, by consequence

Table 3
The errors of material constants determined by standard method.

Materials	Errors (%)							
	Q_{mst}[a]	s_{11}^E	σ	k_{pst}	s_{12}^E	s_{11}^D	s_{12}^D	k_{31}
$k_p > 0.5$ and $Q_m \geq 90$	0.01	0.02	0.01	0.01	0.03	0.02	0.04	0.01
$0.1 < k_p < 0.25$ and $Q_m \leq 50$	3	0.2	0.006	0.8	0.2	0.2	0.2	0.8
$k_p < 0.1$ and $Q_m \leq 30$	40–140 (0.1)	0.3	0.6	18	0.9	0.3	0.9	18
$k_p < 0.1$ and $Q_m \geq 400$	1.5	0.002	0.08	12	0.08	0.009	0.05	12

[a] The values in brackets correspond to Q_{mst} calculated with $\Delta f_{hb.}$

the large differences, beyond the limit of experimental errors, between the results of these methods imply a possible lower accuracy of the fitting method for materials with large coupling factors.

5. Conclusions

A new noniterative method, for determining the dielectric, piezoelectric and elastic constants, in complex form, for piezoceramic materials, in the radial mode, was proposed.

The method is very simple and could be easily applied, since it is based on the standard procedure to determine the elastic compliance and Poisson factor and the measurement of admittance at two frequencies to calculate the dielectric and piezoelectric constants, by solving a system of two equations.

The accuracy of the new method was determined for materials with different planar coupling factors and mechanical quality factors. This method proved to be very accurate and reliable for all materials especially for those with large coupling factors. The accuracy of standard method was also evaluated for the same materials.

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