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Determination of temperature dependence of full matrix material constants of PZT-8 piezoceramics using only one sample

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Abstract

Resonant ultrasound spectroscopy (RUS) was used to determine the temperature dependence of full matrix material constants of PZT-8 piezoceramics from room temperature to 100 °C. Property variations from sample to samples can be eliminated by using only one sample, so that data self-consistency can be guaranteed. The RUS measurement system error was estimated to be lower than 2.35%. The obtained full matrix material constants at different temperatures all have excellent self-consistency, which can help accurately predict device performance at high temperatures using finite element simulations.

Keywords

Resonant ultrasound spectroscopy; PZT-8; Material constants; Temperature

1. Introduction

High power ultrasonic devices are widely used in many fields, such as ultrasonic motors, high intensity focused ultrasound (HIFU) and underwater sonar projectors, which require 'hard' piezoelectric materials with a high mechanical quality factor and a low dielectric loss to deliver a high acoustic power without excessive heat generation and thermal runaway[1].

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Currently, 'hard' $\text{Pb}(\text{Zr,Ti})\text{O}_3$ (PZT) piezoelectric ceramics, including PZT-4 and PZT-8, are the mainstay for high power electromechanical device applications[1, 2].

The mechanical properties of piezoelectric materials always determine the device performance and life[3]. Piezoelectric ceramics are responsible for the mutual transformation of electrical energy and mechanical energy, thus we need dielectric, piezoelectric parameters as well as elastic coefficients in designing those piezoelectric devices using finite element packages[4]. During the operation of high power electromechanical devices, there are generally temperature increase due to the generation of heat by the electric and mechanical losses of piezoelectric materials, which would cause performance degradation of devices[5]. Therefore, knowledge about temperature dependence of the full set material constants of piezoelectric materials is very important for device performance simulations. Unfortunately, it is not easy to perform such property characterization at high temperatures, particularly when the material are strongly anisotropic and sensitive to temperature changes, like piezoelectric materials. Therefore, self-consistent data on the temperature dependence of full matrix material properties for piezoelectric materials were seldom measured[6]. There are some reports on the elastic and piezoelectric constants of PZT-8 as well as lead-free piezoceramics using the resonance and anti-resonance method.[7, 8] However, there is no report in the literature on the temperature dependent full matrix material properties of PZT8 ceramics up to date. Such full matrix material properties of PZT8 ceramics at high temperatures, especially self-consistency data, are very useful and urgently needed for accurately predicting the high temperature performance of high power electromechanical devices using finite element simulations.

In this study, the resonant ultrasound spectroscopy (RUS) technique has been used to determine the complete set of material constants of PZT-8 piezoceramics. This technique can determine the elastic constants of anisotropic elastic materials with very high accuracy[9–11]. One advantage of the RUS over other characterization methods is that all constants are determined from a single specimen, which saves time and eliminated errors from sample to sample variation when multiple samples are used[12].

The identified resonance frequencies will directly determine the inversion results. Note that the measurement of resonance frequencies are somewhat affected by how the sample is placed between the transducers[13]. Although generally speaking, the sample is held by its opposite corners between the two transducers, the resonance frequencies may shift to a higher values if the sample is not held properly[14]. Therefore, we measured the temperature dependent RUS spectra of the same sample three times to estimate the system error, and the sample was placed at room temperature for more than 24 hours after each temperature run. The temperature dependence of elastic and piezoelectric constants were obtained from identified resonance frequencies by the Levenberg-Marquardt (LM) algorithm.

2. Experimental

A piezoceramic PZT-8 ring was cut with a diamond saw into a rectangular parallelepiped. Then, the sample was ground to guarantee flatness of each face to ensure that the sample has proper contacts in the RUS measurement. Loose mechanical contacts between the sample

and transducers resulted in minimal energy transmission between the vibrating sample and the chamber[15]. The final size of the sample was $4.986 \times 6.132 \times 5.720 \text{ mm}^3$. The density determined from the volume and mass of the sample was 7639 kg/cm^3 . The processed PZT-8 sample was poled with a voltage of 10 kV/cm along the z axis, and used for all the measurements in our study.

The low-frequency (100 Hz to 10 kHz) and high-frequency (10 MHz to 40 MHz) capacitances from 20°C to 100°C were measured by an HP4294A impedance analyzer connected to a Delta 9023 temperature chamber. In our case, we used the capacitances at 1 kHz and 30 MHz to calculate the free and clamped dielectric constants. The clamped dielectric constants $\varepsilon_{11}^S/\varepsilon_0$ and $\varepsilon_{33}^S/\varepsilon_0$ versus temperature are shown in Fig. 1. The data are from the average value of three independent measurements, so error bars are also shown in the figure. The red solid and blue dashed lines are fitted $\varepsilon_{11}^S/\varepsilon_0$ and $\varepsilon_{33}^S/\varepsilon_0$ curves:

$$\varepsilon_{11}^S/\varepsilon_0 = 809.300 + 0.452T + 1.365 \times 10^{-2}T^2 \quad \text{with } R^2 = 0.9997 \quad (1)$$

$$\varepsilon_{33}^S/\varepsilon_0 = 561.780 + 0.771T + 7.650 \times 10^{-3}T^2 \quad \text{with } R^2 = 0.9996 \quad (2)$$

The temperature dependence of free dielectric constants $\varepsilon_{11}^T/\varepsilon_0$ and $\varepsilon_{33}^T/\varepsilon_0$ were also calculated, which are used for self-consistency checking of the obtained full set data.

The elastic stiffness constants c_{11}^E , c_{44}^E and c_{66}^E were directly measured on the same sample using ultrasonic pulse-echo method. c_{11}^E , c_{44}^E and c_{66}^E were subsequently calculated from the velocities and the measured density. The RUS setup and the related details for experiments had been reported in our previous study[16]. The DRS 9000 system (Dynamic Resonance Systems, Inc.) was used to measure the resonance frequency spectrum in a furnace (KSL 1100X) from 25°C to 100°C with a thermocouple placed near the sample to detect temperature. The self-written software utilizes the algorithm developed by Levenberg and Mauquardt to obtain the unknown constants by fitting the measured resonance frequencies[17, 18]. A library of FORTRAN subroutines based on LM algorithm, MINPACK, was used in our computation codes[19].

3. Results and discussion

3.1. Resonant ultrasound spectra and mode shape simulation

The mode shapes of the first thirteen frequencies were performed using finite element modal analysis in ANSYS with the RUS inversion results as input parameters. Fig. 2 shows contour plots of the mode shapes calculated with ANSYS, which is well identified with the resonance peaks observed from 180–300 kHz in the RUS. The resonance spectra of three independent measurements from 180 kHz to 300 kHz at 25°C and 85°C are also shown in Fig. 2, one can see that all resonance frequencies increase with temperature for the PZT-8 piezoceramic sample. The first thirteen modes (the sixth mode was missing in the

measurements) between 140 kHz and 300 kHz of measured, fitted and calculated resonance frequencies at $T = 25\text{ }^{\circ}\text{C}$ are shown in Table 1. One can see that the differences between the calculated and fitted frequencies are all very small.

3.2. Inversion results

In the RUS technique, mode identification is the most difficult task during the inversion process when trying to extract the material constants. The dimensions and density of the sample, clamping dielectric constants, elastic and piezoelectric constants, were used as input parameters to calculate the resonance frequencies. In the calculation process, we adjusted the guessed parameters until the calculated frequencies match the measured frequencies. Then these parameters were used to determine the elastic and piezoelectric constants at $25\text{ }^{\circ}\text{C}$. Next, the full tensor properties at $25\text{ }^{\circ}\text{C}$ were used as the initial parameters for the next temperature $T=30\text{ }^{\circ}\text{C}$. By this analogy, the inversion process was conducted every $5\text{ }^{\circ}\text{C}$ until $100\text{ }^{\circ}\text{C}$.

The inversed elastic constants versus temperature are shown in Fig. 3. The error bars are due to three independent measurements using the same sample (placing the sample on different corners between the transducers). In the investigated temperature interval, the elastic and piezoelectric constants varied nonlinearly with temperature, and were fitted to quadratic functions of temperature. The fitted temperature dependence curves for these constants are given by

$$c_{11}^E = 14.515 + 8.350 \times 10^{-3}T - 3.870 \times 10^{-5}T^2 \text{ with } R^2 = 0.9899 \quad (3)$$

$$c_{12}^E = 7.314 + 6.410 \times 10^{-3}T - 3.122 \times 10^{-5}T^2 \text{ with } R^2 = 0.9799 \quad (4)$$

$$c_{13}^E = 7.512 + 4.540 \times 10^{-3}T - 1.878 \times 10^{-5}T^2 \text{ with } R^2 = 0.9874 \quad (5)$$

$$c_{33}^E = 12.565 + 6.180 \times 10^{-3}T - 2.865 \times 10^{-5}T^2 \text{ with } R^2 = 0.9911 \quad (6)$$

$$c_{44}^E = 2.807 + 1.320 \times 10^{-3}T - 3.122 \times 10^{-6}T^2 \text{ with } R^2 = 0.9971 \quad (7)$$

$$e_{15} = 11.777 + 2.100 \times 10^{-3}T + 8.151 \times 10^{-5}T^2 \text{ with } R^2 = 0.9983 \quad (8)$$

$$e_{31}=4.614-2.300 \times 10^{-3}T+5.436 \times 10^{-5}T^2 \text{ with } R^2=0.9919 \quad (9)$$

$$e_{33}=13.570+4.520 \times 10^{-3}T+1.230 \times 10^{-4}T^2 \text{ with } R^2=0.9869 \quad (10)$$

The longitudinal stiffness constants (c_{11}^E and c_{33}^E) depend strongly on temperature, however, the shear stiffness constants (c_{12}^E , c_{13}^E and c_{44}^E) are only weakly temperature dependent in this temperature range. The maximum error for (c_{12}^E , c_{13}^E and c_{44}^E) is 2.35% appearing at 100 °C for c_{12}^E , and the maximum error for (c_{11}^E and c_{33}^E) is lower than 1.35%, which are quite accurate in practical measurements.

Temperature dependence of the piezoelectric constants e_{15} , $-e_{31}$ and e_{33} are also extracted from the observed resonant spectra and are shown in Fig. 4. All piezoelectric constants increase with temperature. The errors of e_{15} and e_{33} due to three independent measurements are very small. The biggest error of $-e_{31}$ is within 1.5% at 60 °C. The complete set of elastic and piezoelectric constants of three independent measurements are in excellent agreement, indicating the high precision of our RUS setup. The inversed piezoelectric and elastic constants, the free and clamped dielectric constants together with mechanical coupling factor k_{33} at 25 °C and 85 °C are compared with other reported data measured by the resonance anti-resonance method as shown in Table 2.

Fig. 5 shows the temperature dependence of coupling factors k_t , k_{15} , k_{31} and k_{33} calculated from the inversion results. An error bar is applied to all the coupling factors from three independent inversion results. k_{15} , k_{31} and k_{33} decrease slightly with the increasing temperature, however, k_t is not sensitive to the variation of temperature. The literature data for k_{33} are also listed in Table 2, and they are highly consistent.

3.3. Self-consistency check

A poled rectangular parallelepiped PZT-8 sample has a macroscopic symmetry of C_{2h} , and there exist four kinds of mode group for the parallelepiped, Ag, Au, Bg and Bu[22]. The experimental and calculated frequencies of four mode frequencies Au-1, Bu-2, Ag-11 and Bg-10 versus temperature are shown in Fig. 6. The points are measured results and the lines are the calculated results using our obtained material constants. An error bar is applied to each mode based on three independent experimental and inversion results. One can see that the calculated and measured results are in excellent agreement.

In order to check the self-consistency of the inversion results, we first calculated the free dielectric constants ε_{11}^T and ε_{33}^T from the inversion results using equations (11) and (12), then compared them with those measured as shown in Fig. 7. We also calculated piezoelectric constants d_{15} and d_{33} using different equations (13–16) and the results are shown in Fig. 8. The difference are 3.0% for d_{15} and 2.2% for d_{33} , demonstrating good agreement. In addition, the direct measurement result of the piezoelectric constant d_{33} at room temperature

using a d_{33} PiezoMeter is about 232 pm/V, which also corresponds well with the calculated results. The equations used in self-consistency checking are as follows:

$$\varepsilon_{11}^S = \varepsilon_{11}^S + e_{15}^2 s_{44}^E \quad (11)$$

$$\varepsilon_{33}^S = \varepsilon_{33}^S + 2d_{31}e_{31} + d_{33}e_{33} \quad (12)$$

$$d_{15} = \sqrt{1 - c_{44}^E / c_{44}^D} \sqrt{\varepsilon_{11}^T s_{44}^E} \quad (13)$$

$$d_{15} = e_{15} s_{44}^E \quad (14)$$

$$d_{33} = 2e_{31} s_{13}^E + e_{33} s_{33}^E \quad (15)$$

$$d_{33} = \sqrt{(s_{33}^E - s_{33}^D)} \varepsilon_{33}^T \quad (16)$$

4. Conclusion

Temperature evolutions of full matrix material constants for PZT-8 ceramics are obtained from room temperature to 100 °C using only one sample. RUS method can eliminate the variation problem from sample to sample as well as substantially reduce the experimental workload. The self-consistency of obtained data was checked by comparing the measured and calculated free dielectric constants, calculated piezoelectric constants d_{15} and d_{33} using different equations, all showing excellent agreement. In addition, the RUS setup system error was also estimated by comparing the inversion results from three independent measurements using the same sample, and the maximum error for all the elastic and piezoelectric constants is only 2.35%. The temperature dependent data of PZT-8 ceramic can help us design and optimize high power electromechanical devices and predict their performance during operation, which usually will induce temperature increase.

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Highlights

1. Full matrix material constants of PZT-8 were determined from 25 °C to 100 °C.
2. Data self-consistency is guaranteed in the whole temperature range.
3. The RUS system error is less than 2.35% much smaller than other methods.

Fig. 1.

Measured clamped dielectric constants $\epsilon_{11}^S/\epsilon_0$ and $\epsilon_{33}^S/\epsilon_0$, and the fitting curves.

Fig. 2.
Resonant ultrasound spectra between 180 kHz–300 kHz measured at 25 °C and 85 °C, and mode shapes calculated by ANSYS

Fig. 3.

Temperature dependence of elastic stiffness constants c_{11}^E , c_{12}^E , c_{13}^E , c_{33}^E and c_{44}^E . The left (red) and right (green) y-axes are for (c_{11}^E and c_{33}^E) and (c_{12}^E , c_{13}^E and c_{44}^E), respectively.

Fig. 4. Temperature dependence of piezoelectric constants of e_{15} , $-e_{31}$ and e_{33} . The left (red) and right (green) y-axes are for (e_{31} and e_{33}) and $-e_{31}$, respectively.

Fig. 5. Temperature dependence of coupling factors k_t , k_{15} , k_{31} and k_{33} calculated from the inversion results.

Fig. 6. Temperature dependence of resonance modes Au-1, Bu-2, Ag-11 and Bg-10, where the points and lines correspond to measured and calculated results, respectively.

Fig. 7. Comparison between measured and calculated free dielectric constants. Solid and dashed lines are the fitting results.

Fig. 8. Temperature dependence of d_{15} calculated from Equations (13) (red square) and (14) (red solid line), and d_{33} calculated from Equations (15) (blue rhombus) and (16) (blue dashed line).

Table 1

Comparison of measured, fitted and calculated resonance frequencies at $T = 25\text{ }^{\circ}\text{C}$, and calculated frequencies by ANSYS for the first 13 modes.

No.	Mode	$f_{meas}(\text{kHz})$	$f_{fit}(\text{kHz})$	$f_{cal} \text{ by RUS}(\text{kHz})$	Diff. ($\times 10^{-3}\mu$)	$f_{cal} \text{ by ANSYS}(\text{kHz})$
1	Au-1	143.340	143.461	143.642	1.09	143.595
2	Au-2	194.555	194.631	195.462	-1.96	195.780
3	Bu-1	201.452	201.528	202.366	-0.11	202.266
4	Bu-2	224.771	224.893	225.622	-0.13	225.619
5	Au-3	234.110	234.191	234.862	0.004	235.045
6	Bg-1	243.145	--	244.308	--	244.576
7	Ag-1	243.877	243.967	244.480	0.50	244.674
8	Bu-3	249.188	249.265	249.645	0.51	249.618
9	Bg-2	257.062	257.159	258.220	-1.32	258.411
10	Ag-2	259.382	259.482	259.743	2.87	259.821
11	Ag-3	269.332	269.413	270.786	-2.91	270.782
12	Ag-4	271.407	271.515	271.841	1.51	272.041
13	Bg-3	284.348	284.440	285.446	-0.59	285.728

$$^a \text{Diff} = (f_{cal} - f_{fit})/f_{fit}$$

Table 2

Inversed elastic constants c_{ij}^E (10^{10} N/m²) and piezoelectric constants e_j (C/m²) at 25 °C and 85 °C, the free and clamping relative dielectric constants and mechanical coupling factor k_{33} , as well as the reported data in the literatures.

c_{11}^E	c_{12}^E	c_{13}^E	c_{33}^E	c_{44}^E	Reference
14.70	7.45	7.61	12.70	2.84	this work(25 °C)
14.94	7.63	7.75	12.88	2.90	this work(85 °C)
13.7	7.20	7.50	12.30	3.10	[20](RT)
14.9	8.11	8.11	13.2	3.13	[21](RT)

e_{15}	e_{31}	e_{33}	k_{33}	
11.88	-4.59	13.83	0.65	this work(25 °C)
12.55	-4.76	14.85	0.64	this work(85 °C)
10.40	-4.00	13.20	0.64	[20](RT)
10.30	-4.10	14.00	0.64	[21](RT)

$\epsilon_{11}^T/\epsilon_0$	$\epsilon_{33}^T/\epsilon_0$	$\epsilon_{11}^S/\epsilon_0$	$\epsilon_{33}^S/\epsilon_0$	
1429	1087	830	587	this work(25 °C)
1563	1209	946	680	this work(85 °C)
1290	1000	900	580	[20](RT)
1290	1000	900	600	[21](RT)