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FAST TRACK COMMUNICATION

Evolution of transverse piezoelectric response of lead zirconate titanate ceramics under hydrostatic pressure

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Abstract
The piezoelectric properties of 31-mode resonators of lead zirconate titanate ceramics under hydrostatic pressure from 0.1 to 325 MPa were evaluated by a fitting method, in which mechanical loss was taken into account. Our results based on the fitting method showed a hydrostatic pressure independent tendency of the piezoelectric coefficient and the electromechanical coupling factor because the adopted PZT ceramic can be considered as a linear system in our experiment, while two misleading tendencies of piezoelectric coefficient were obtained based on the resonance method when ignoring the contribution of the mechanical loss.

Knowledge of the piezoelectric properties of ferroelectric materials under hydrostatic pressure is necessary in the design of high power underwater acoustic devices, since high pressures could change the materials' properties significantly that influence the devices performance [1–5]. In particular, piezoelectric coefficients and electromechanical coupling factors attract more attention due to their importance in the design of devices. Usually these parameters are determined by the resonance method through the characteristic frequencies extracted from the impedance spectrum, for example, f_m (frequency of minimum impedance) and f_n (frequency of maximum impedance), f_s (frequency of maximum conductance) and f_p (frequency of maximum resistance) or f_r (resonance frequency (zero reactance)) and f_a (antiresonance frequency (zero susceptance)) [6].

On the other hand, as pointed out by Holland and EerNisse [7], the resonance method destroyed the all phase information. In other words, this method can only be applied to the materials if the losses are small, which means that the maximum phase angle must approach 90° in the phase angle spectrum (lossless condition). However, the maximum phase angle usually deviates considerably from 90° upon application of hydrostatic pressure. In this case, using the resonance method to calculate the piezoelectric coefficients is unfounded and would introduce a considerable error [8, 9]. Although several more accurate methods have been proposed to determine the piezoelectric coefficients at lossy condition (the common ground of these methods is to establish the impedance expression for the specified vibration mode and then determine various physical parameters by analysing the observed data with a certain theoretical relationship around the resonance frequency) [8–10], discussion on the piezoelectric properties under hydrostatic pressure, in which lossless conditions are often not fulfilled, is still expected. In this communication, taking into account the mechanical loss, we evaluated the piezoelectric response of PZT5 ceramics by a fitting method and showed that the piezoelectric coefficient d_{31} was almost independent of the hydrostatic pressure increased from 0.1 to 325 MPa at room temperature.

In this communication, a commercial soft PZT5 ceramic was cut into 16 mm length, 3 mm width, 0.8 mm thickness and was then well poled along the thickness direction in order to excite the length-extensional vibration mode (31-mode). The hydrostatic pressure, generated by an in-house built setup [11], was applied from 0.1 to 325 MPa. An impedance analyzer (HP4294A, Hewlett Packard) was employed to measure the characteristic frequencies (f_m, f_n, f_s and f_p), frequency dependent impedance and the phase angle spectrum of the sample.

For the 31-mode, the admittance expression can be written as follows [12]:

$$Y = j\omega\frac{lw}{t}\varepsilon_{33}^T\left[\frac{d_{31}^2}{\varepsilon_{33}^T s_{11}^E}\frac{\tan(\omega l\sqrt{s_{11}^E\rho}/2)}{\omega l\sqrt{s_{11}^E\rho}/2} + 1 - \frac{d_{31}^2}{\varepsilon_{33}^T s_{11}^E}\right], \quad (1)$$

where ω is the angle frequency, ε_{33}^T is the dielectric permittivity, s_{11}^E is the elastic constant, d_{31} is the piezoelectric coefficient, ρ is the density, l, w and t are the length, the width and the thickness of the piezoelectric vibrator, respectively.

At lossless condition, k_{31} ($k_{31}^2 = d_{31}^2/\varepsilon_{33}^T s_{11}^E$) and d_{31} can be calculated using equations (2) and (3), respectively [6]:

$$k_{31}^2/(1-k_{31}^2) = \frac{\pi}{2}\frac{f_2}{f_1}\tan\frac{\pi}{2}\frac{\Delta f}{f_1}, \quad (2)$$

$$d_{31} = k_{31}\sqrt{s_{11}^E\varepsilon_{33}^T}, \quad (3)$$

where $s_{11}^E = 1/4\rho f_1^2 l^2$, $f_m = f_s = f_1$ and $f_n = f_p = f_2$.

At lossy condition, three different types of losses in the piezoelectric vibrator are presented. They are electrical loss, mechanical loss and piezoelectric loss, which can be represented by assuming the parameters of equation (1) to be complex:

$$\begin{aligned}\varepsilon_{33}^T &= \varepsilon_{33}^{T'}(1 - j\tan\delta_E),\\ s_{11}^E &= s_{11}^{E'}(1 - j\tan\delta_M),\\ d_{31} &= d_{31}'(1 - j\tan\delta_P),\end{aligned} \quad (4)$$

where $\tan\delta_E$, $\tan\delta_M$ and $\tan\delta_P$ are electric, mechanical and piezoelectric loss factors, respectively. In this case, the real and imaginary parts of the dielectric permittivity can be determined by an impedance analyzer at a low frequency (1 kHz). Then the fitting method is used to determine d_{31}', $s_{11}^{E'}$ and $\tan\delta_M$. The initial values of the imaginary part of the piezoelectric and elastic coefficients are set to zero, and the initial values of the real part of the piezoelectric and elastic coefficients are calculated through the conventional resonance method by equations (2) and (3). Using the nonlinear fitting method (Gauss–Newton method) we fit the obtained impedance and phase angle to equation (1) to extract the value of d_{31}', $s_{11}^{E'}$ and $\tan\delta_M$.

Theoretically, $\tan\delta_P$ can be taken into account in the fitting process. However, from a practical point of view, it is difficult to determine $\tan\delta_P$ due to its relatively small value in PZT ceramics [9]. Furthermore, the influence of $\tan\delta_P$ on the impedance and the phase angle spectrum is also very limited compared with the influence of other coefficients (d_{31}', $s_{11}^{E'}$, $\tan\delta_M$). Therefore, $\tan\delta_P$ was ignored, resulting in only the

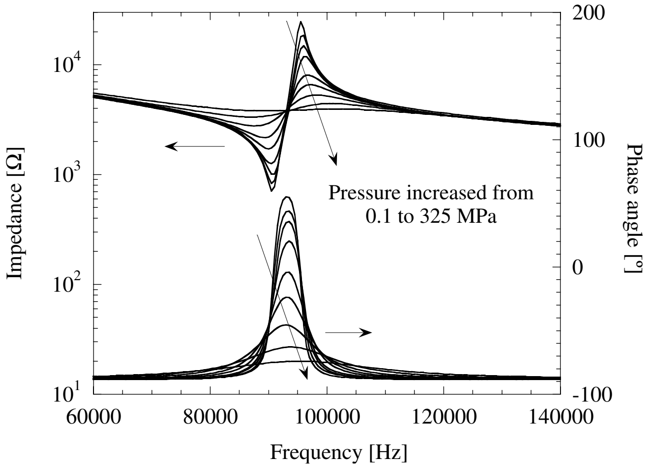

Figure 1. The impedance and the phase angle spectrum of PZT5 piezoelectric vibrator in k_{31} mode under different hydrostatic pressures.

real part of d_{31} in equation (1). After determination of those parameters, the coupling factor k_{31} can be denoted as

$$k_{31}^2 = \frac{(d_{31}')^2}{s_{11}^{E'}\varepsilon_{33}^{T'}}. \quad (5)$$

It is clear that there is no imaginary part for the coupling factor k_{31} in equation (5). On the contrary, many papers reported the imaginary part of k_{31}, which was calculated from the complex physical parameters [8, 9]. Although we could readily get k_{31} in the complex format using the parameters extracted from the fitting method, we preferred k_{31} as a real number taking into account the following: (1) according to the IEEE standard, k_{31} is defined as a real number and obtain it at a static/quasistatic state; (2) k_{31} represents the ability of conversion of mechanical to electric energy or vice versa. If we define k_{31} as a complex number, the imaginary part of it is meaningless; (3) in order to make a comparison with k_{31} determined by other methods, it is convenient to use the uniform format, i.e. the real number format.

By increasing the hydrostatic pressure from 0.1 to 325 MPa, the difference between Z_m (minimum impedance) and Z_n (maximum impedance) was decreased gradually and the shape of the impedance spectrum being changed from sharp to broad was also observed simultaneously, as shown in figure 1. It is observed that the maximum phase angle is also decreased drastically down to $-75°$ with increasing hydrostatic pressure, while the corresponding frequency is almost independent of the pressure. Similar responses in other systems are also reported, but sometimes the frequency of the maximum phase angle shifts with increasing pressure [4, 5]. Table 1 displays the elastic coefficient (s_{11}^E, $\tan\delta_M$) determined by the fitting method, the dielectric permittivity (ε_{33}^T, $\tan\delta_E$) determined by the impedance analyzer and k_{31}^{fit} calculated from equation (5) under different hydrostatic pressures. These data, except for the mechanical loss $\tan\delta_M$, which is significantly increased from 0.017 to 0.352 under the pressure from 0.1 to 325 MPa (increased by a factor of 20), are almost independent of the pressure (the maximum variation is less than 5%). Unlike the analysis in [3], we believe that

Table 1. Fitting and experimental data of piezoelectric vibrator under different hydrostatic pressures.

Pressure (MPa)	k_{31}^{fit}	tgδ_M	s_{11}^{E} ($\text{m}^2\,\text{N}^{-1}$)	$\varepsilon_{33}^{T'}/\varepsilon_0$	tgδ_E
0.1	35.80%	0.017	1.60E-11	1682	0.018
60	36.60%	0.023	1.60E-11	1650	0.018
90	36.90%	0.029	1.60E-11	1653	0.019
120	37.20%	0.038	1.60E-11	1680	0.017
170	36.60%	0.058	1.60E-11	1693	0.018
210	36.00%	0.080	1.60E-11	1675	0.020
250	35.50%	0.125	1.60E-11	1695	0.019
290	35.10%	0.220	1.57E-11	1687	0.020
325	34.90%	0.352	1.52E-11	1662	0.019

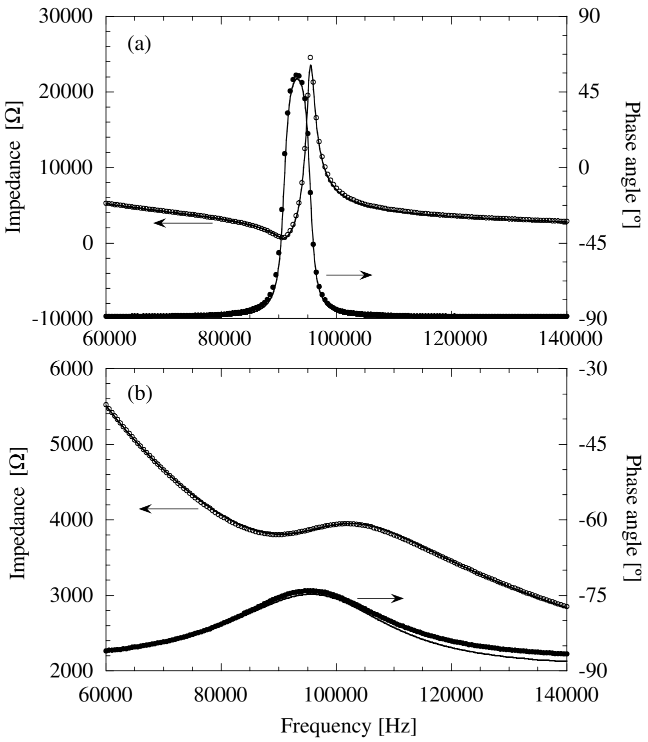

Figure 2. Comparison between the calculated impedance and the phase angle spectrum (solid lines) with the experimental ones (open and solid circles) at (*a*) 0.1 MPa and (*b*) 325 MPa hydrostatic pressure.

the drastic increase in the mechanical loss is not only due to the domain wall movement but also due to the friction between the vibrator and the pressure transmitting liquid. In principle, domain wall movement could also result in the increase in the dielectric and piezoelectric losses. However, according to the result shown in table 1, there is no notable increase in the dielectric loss. At present, there is no suitable method to evaluate the response of the piezoelectric loss under hydrostatic pressure. On the other hand, the friction between the vibrator and the pressure transmitting liquid could only increase the mechanical loss because this friction is a kind of mechanical friction. Figures 2(*a*) and (*b*) show the fitting results at 0.1 MPa and 325 MPa pressure, respectively. It can be seen that in both cases, the fitting results show good agreement with the experimental data, even the phase angle approaches −75° under 325 MPa pressure.

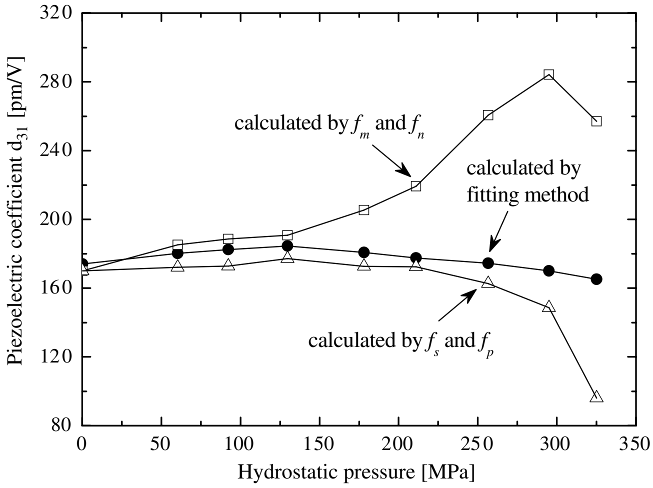

Figure 3. Comparison of d_{31} determined by the fitting and resonance methods under different hydrostatic pressures.

Figure 3 displays a comparison of d_{31} determined by the fitting and resonance methods with respect to the pressure. It is clear that d_{31} determined by the fitting method suggests a flat response to the pressure, while the calculations based on the resonance method show a first increasing and then a decreasing tendency (calculated from equations (2) and (3) using f_m and f_n), and a decreasing tendency (calculated from equations (2) and (3) using f_s and f_p). Here, we have to answer a question as to which variation tendency of d_{31} is right. According to linear theory of thermodynamics [13], the piezoelectric response will be unchanged under different hydrostatic pressures if the material can be considered as a linear system (or can be described by linear equations of state). To the best of our knowledge, the pressure used in this experiment is not high enough to induce any depoling effect (because the piezoelectric response has not changed when the pressure was taken off) or phase transition [14]. Furthermore, the dielectric permittivity ε_{33}^{T} of PZT5 is almost independent of the hydrostatic pressure (presented in table 1). Therefore, PZT5 ceramics can be considered as a linear system under the hydrostatic pressure increasing to 325 MPa for a small signal measurement and a flat piezoelectric response under hydrostatic pressure is also reasonable, while the two misleading tendencies of d_{31} are caused by the influence of the mechanical loss on shifting the characteristic frequencies (f_m, f_n, f_s and f_p) [15].

As discussed above, the vibration of the piezoelectric vibrator under hydrostatic pressure can be recognized as damped vibration, in which the mechanical loss cannot be ignored for studying the piezoelectric response of the vibrator. In addition, because of the increase in the mechanical loss factor with increasing pressure, the difference between Z_m and Z_n and the maximum phase angle usually decrease with increasing pressure. These decrease phenomena are common characteristics for the piezoelectric vibrator under hydrostatic pressure [1–5]. On the other hand, it should be noted that the amplitude of the mechanical loss factor is not only related to the hydrostatic pressure but also related to the type of vibration mode and pressure transmitting fluid, because the mechanical loss comes partly from the friction between the vibrator and the pressure transmitting liquid.

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