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The Complex Piezoelectric Measurement Using Analysis of Complex Immittance of PZT Ceramics

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The material coefficients, such as piezoelectric constant, elastic constant and dielectric permittivity, were determined as complex values by the non-linear least-squares-fitting of immittance data measured for length-extensional bar resonators of "soft"- and "hard"-PZTs. The piezoelectric *d*-constant should be a complex value to obtain a best fitting between observed and calculated results. As the elastic, dielectric and piezoelectric losses determined in this process were not "intrinsic" losses, the procedure to estimate intrinsic elastic, dielectric and piezoelectric losses were proposed. The piezoelectric loss was dominantly derived from the elastic and dielectric losses but imaginary part remained even in the intrinsic piezoelectric *h* constant. The domain-wall-clamping in hard PZT markedly reduced the elastic loss rather than dielectric or piezoelectric loss. Most notable difference between the soft- and hard-PZTs were observed in their elastic losses. The displacement in resonant mode was measured using a newly developed double beam laser Doppler interferometer. The displacement of hard PZT is larger than the soft PZT around the resonance frequency because the displacement was determined not only by the piezoelectric *d* constant by also by the elastic loss.

Key words: PZT, piezoelectricity, least-squares-fitting, ceramic resonator, resonance method

1. INTRODUCTION

The resonance method is commonly used to determine material coefficients of piezoelectric ceramics, such as the elastic compliance (s), the dielectric permittivity (ε) and the piezoelectric d-constant (d).[1] This method is practically adequate for measurements of materials with high quality and a high electromechanical coupling factor, but the losses cannot be found easily and the piezoelectric constant is usually regarded as a real number. Smits[2] and Alemany *et al.*[3] proposed the "iterative method" for accurate determination of real and imaginary material coefficients from complex admittance measured at three or four different frequencies. The similar method was employed by Alberta and Bhalla[4] to determine the complex material coefficients of lead zirconate-based ceramics. However, it is obvious that the accuracy of the analysis increases if the whole immittance spectrum is incorporated into the calculation to determine the complex material coefficients. The complex piezoelectric d-constant can be determined by other methods, such as the measurements of the phase delay of the strain to the applied field using an interferometer[5] or a modified measuring method of the "direct" piezoelectric effect.[6,7] However, these methods are adequate for a quasi-static or off-resonance state and cannot be applied to piezoelectric resonators. It is important to develop analyzing technique to accurately determine complex material coefficients in the conventional resonance method.

The material coefficients determined by the resonance method for a length-extensional bar resonator are s^E, ε^T and d. It should be noted that the losses of these coefficients are not "intrinsic" losses of a material because of the coupling between material coefficients. Recently, Uchino and Hirose[8] reported the phenomenological theory to deal with losses of these coefficients and proposed a method to measure the intrinsic losses separately in the off-resonance state. They concluded that the intrinsic piezoelectric loss of PZT ceramics was relatively large. As for the resonance state, Sasaki *et al.*[9] tried to determine the intrinsic losses of the material coefficients by measuring elastic losses at both resonance and anti-resonance frequencies. They concluded that the intrinsic piezoelectric loss was negligible. There has been a discussion whether the intrinsic piezoelectric loss exists or does not exist.

In the present study, we have developed a method to accurately determine the complex material coefficients using non-linear least-squares-fitting of immittance data, and establish the process to determine the intrinsic losses of material coefficients. Moreover, a double beam laser Doppler interferometer was developed to measure the displacement of the PZT resonators around the resonance frequency.

2. EXPERIMENTAL PROCEDURE

Ceramics resonators used for the analysis were made of "soft"-PZT ceramics with the composition of ($Pb(Zr_{0.52}Ti_{0.48})O_3$+0.5mol%$Nb_2O_5$) and "hard"-PZT ceramics with the composition of ($0.95Pb(Zr_{0.45}Ti_{0.55})O_3$ +$0.05Pb(Sb_{0.5}Nb_{0.5})O_3$+1.5mol%MnO). These ceramics were prepared from raw materials of PbO, ZrO_2, TiO_2, Nb_2O_5, Sb_2O_5 and $MnCO_3$. The sintering temperatures of the soft- and hard-PZTs were 1270℃ and 1210℃, respectively. These ceramics were cut into a rectangular shape of 12 x 3 x 1 mm^3. Electrodes were formed on the 12 x 3 mm^2 surfaces by Au-sputtering. The poling condition was 3 kV/mm at 120℃ for 10 min.

Complex admittance was measured as a function of frequency using an impedance analyzer (HP4192A) at room temperature. The oscillation level of the ac-signal was 0.1V. Complex impedance was calculated from the complex admittance data. The elastic compliance, dielectric permittivity and piezoelectric constant of PZT ceramics were determined as complex values using the "immittance-fitting method" as described in the next section.

The displacement of ceramic resonators was measured to know the vibration amplitude at the resonance frequency. A schematic diagram of double beam laser Doppler interferometer (DB-LDI) is shown in Fig.1. Two LDIs measured the velocities of two opposite surfaces of PZT plate at the same time. Signals from the interferometers were amplified with lock-in amplifiers and analyzed by a computer.

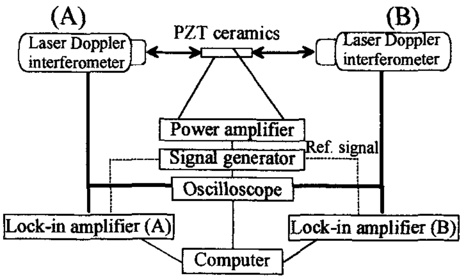

Fig.1 Schematic diagram of double beam laser Doppler interferometer.

3. IMMITTANCE FITTING METHOD

The admittance of a Z cut X plate of piezoelectric ceramic poled along Z-axis is given by the following equation,

$$Y = \frac{i\omega a\ell}{t}\left(\varepsilon_{33}^{T} - \frac{d_{31}^{2}}{s_{11}^{E}} + \frac{d_{31}^{2}}{s_{11}^{E}}\frac{\tan(\omega l/2v)}{\omega l/2v}\right) \quad (1)$$

where ω is an angular frequency, a, l and t are respectively the width, length and thickness of the resonator, d_{31} is a piezoelectric constant, $\varepsilon_{33}{}^{T}$ is a dielectric permittivity under a constant stress, $s_{11}{}^{E}$ is a elastic compliance under a constant field, and $v = \sqrt{1/\rho s_{11}^{E}}$ (ρ: density) is a sound velocity. The d_{31}, $\varepsilon_{33}{}^{T}$, $s_{11}{}^{E}$ are represented as complex values,

$$\begin{aligned}(d_{31})^* &= (d_{31})' - i(d_{31})'',\\ (\varepsilon_{33}{}^{T})^* &= (\varepsilon_{33}{}^{T})' - i(\varepsilon_{33}{}^{T})'',\\ (s_{11}{}^{E})^* &= (s_{11}{}^{E})' - i(s_{11}{}^{E})''\end{aligned} \quad (2)$$

In the least-squares calculation, these complex parameters were substituted into the eq.(1) and they were refined to minimize the following function F,

$$F = |G_o-G_c| / |G_o| + |B_o-B_c| / |B_o| + |R_o-R_c| / |R_o| + |X_o-X_c| / |X_o| \quad (3)$$

where G, B, R and X are respectively conductance, susceptance, resistance and reactance as a function of frequency, and suffixes "c" and "o" represent calculated and observed values. The complex impedance term was indispensable to accurately determine the imaginary part of d- constant. We employed the "simplex method"[10] for the algorism of the least-squares calculation.

From the complex values of elastic compliance, dielectric permittivity and piezoelectric constant in eq.(2), the elastic, dielectric and piezoelectric losses are defined as follows,

$$(s_{11}^{E})^* = (s_{11}^{E})' - i(s_{11}^{E})'' = (s_{11}^{E})' \times [1 - i\psi(s^{E})] \quad (4)$$

$$(\varepsilon_{33}^{T})^* = (\varepsilon_{33}^{T})' - i(\varepsilon_{33}^{T})'' = (\varepsilon_{33}^{T})' \times [1 - i\varphi(\varepsilon^{T})] \quad (5)$$

$$(d_{31})^* = (d_{31})' - i(d_{31})'' = (d_{31})' \times [1 - i\eta(d)] \quad (6)$$

where $\psi(s^{E})$ is the elastic loss, $\varphi(\varepsilon^{T})$ is the dielectric loss and $\eta(d)$ is the piezoelectric loss. However, it is very important to point out that the $\psi(s^{E})$, $\varphi(\varepsilon^{T})$ and $\eta(d)$ defined above are not "intrinsic" losses because they are coupled with each other. These losses are called as "extrinsic" losses. The theory to define the intrinsic losses was first established by Ikeda.[11] In order to define intrinsic losses, we have to start from piezoelectric equations in terms of extensive valuables of strain (S) and electric displacement (D),

$$T = (1/s^{D})S - hD \quad (7)$$

$$E = (1/\varepsilon^{S})D - hS \quad (8)$$

where T is stress, E is field, s^{D} is elastic compliance under a constant electric displacement, ε^{S} is dielectric permittivity under a constant strain and h is piezoelectric constant. The intrinsic losses can be defined as the losses of material constants in the above piezoelectric equations as follows:

$$(s_{11}^{D})^* = (s_{11}^{D})' \times (1 - i\psi) \quad (9)$$

$$(\varepsilon_{33}^{S})^* = (\varepsilon_{33}^{S})' \times (1 - i\varphi) \quad (10)$$

$$(h_{31})^* = (h_{31})' \times (1 + i\eta) \quad (11)$$

where ψ is the intrinsic elastic loss, ϕ is the intrinsic dielectric loss, and η is the intrinsic piezoelectric loss. The relations between the extrinsic and the intrinsic losses are given by,[11]

$$\eta(d) = \psi(s^{E}) + \varphi - \eta \quad (12)$$

$$\psi(s^{E}) = [\psi + k^{2}(\varphi - 2\eta)]/(1 - k^{2}) \quad (13)$$

$$\varphi(\varepsilon^{T}) = [\varphi + k^{2}(\psi - 2\eta)]/(1 - k^{2}) \quad (14)$$

$$\eta(d) = [\psi + \varphi - (1 + k^{2})\eta]/(1 - k^{2}) \quad (15)$$

where k is the electromechanical coupling factor and its square is given by $k^2 = d^2/(s_{11}{}^{E}\cdot\varepsilon_{33}{}^{T})$.

The $s_{11}{}^{D}$ was first calculated as a complex value using the relation, $s_{11}{}^{D} = (1-k^2)s_{11}{}^{E}$, where k^2 should be a complex value. The intrinsic elastic loss (ψ) was subsequently determined from $\psi = (s_{11}{}^{D})''/(s_{11}{}^{D})'$. The quasi-intrinsic dielectric loss $\varphi(\varepsilon^{LS})$, where LS means the length of the specimen is fixed, was calculated using the relations, $\varepsilon_{33}{}^{LS} = \varepsilon_{33}{}^{T}(1-k^2)$ and $\varphi(\varepsilon^{LS}) = (\varepsilon_{33}{}^{LS})''/(\varepsilon_{33}{}^{LS})'$. In the strict sense, $\varphi(\varepsilon^{LS})$ is not equal to the intrinsic dielectric loss (φ). Both losses have the following interrelation,[9,11]

$$\phi(\varepsilon^{LS}) = [\varphi + f(\psi - 2\eta)]/(1 - f) \quad (16)$$

where f is a constant which is not equal to k^2 because the type of coupling is different. As f is unknown, we regarded $\varphi(\varepsilon^{LS})$ to be the intrinsic dielectric loss (φ) as reported by Sasaki *et al.*[9] The loss of h_{31} was evaluated from eq.(12). Equations (13)-(15) were used to verify the relations between parameters.

4. RESULTS AND DISCUSSION

Figure 2 shows a result of the immittance-fitting of the soft-PZT. At first, the imaginary part of the *d*-constant (d_{31}") was fixed at zero and the immittance-fitting process was performed. Large deviations were observed in the reactance (X) as shown in Fig.2. This deviation was markedly improved by assuming $d_{31}" \neq 0$, indicating that the piezoelectric *d*-constant of the soft-PZT is a complex number. It seems to be necessary to modify the analysis in the resonance method in order to determine the parameters accurately. In the case of the hard-PZT, a good agreement between the observation and calculation was obtained but the effect of the d_{31}" was small in comparison with the soft-PZT.

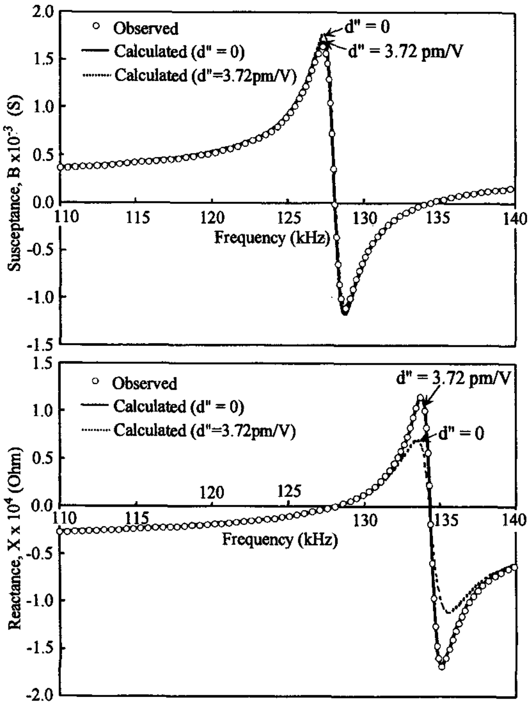

Fig.2 Calculated and observed susceptance and reactance of soft-PZT around the resonance frequency.

Material constants determined by the immittance-fitting method are listed in Table 1. Real parts of all material coefficients [$(s_{11}^E)'$, $(\varepsilon_{33}^T)'$ and $(d_{31})'$] are larger in the soft-PZT than in the hard-PZT. These coefficients determine the response of a material to the electric field and/or the stress. The difference between the soft- and hard-PZTs can be interpreted in terms of domain wall motion. The domain walls in acceptor-doped perovskite ferroelectrics are clamped by the internal bias field caused by defect dipoles consisting of acceptors and oxygen vacancies.[12] In the hard-PZT, Mn ions act as acceptors. As the domain wall motion is clamped in the hard-PZT, the contribution to the real part of the material coefficients of the hard-PZT is smaller than that in the soft-PZT. It is known that domain wall motion also increases the losses.[13]

The elastic, dielectric and piezoelectric losses are listed in Table 2. All intrinsic losses (ψ, φ, η) are smaller than the corresponding extrinsic losses ($\psi(s^E)$,

Table 1 Material coefficients of the soft- and hard-PZTs

Material coefficients	Soft-PZT	Hard-PZT
$(s_{11}^E)'$ x10^{-12} [m^2/N]	13.5	9.72
$(s_{11}^E)''$ x10^{-12} [m^2/N]	0.15	0.011
$(\varepsilon_{33}^T)'/\varepsilon_0$	1340	815
$(\varepsilon_{33}^T)''/\varepsilon_0$	43.7	9.2
$-(d_{31})'$ x10^{-12} [m/V]	135	65.2
$-(d_{31})''$ x10^{-12} [m/V]	3.72	0.53
Density [kg/m^3]	7.85x10^3	7.81x10^3

Table 2 Elastic, dielectric and piezoelectric losses of the soft- and hard-PZTs

Elastic, dielectric and piezoelectric losses	Soft-PZT	Hard-PZT
$\psi(s^E)$	1.11x10^{-2}	1.16x10^{-3}
$[\psi + k^2(\varphi - 2\eta)]/(1 - k^2)$	1.11x10^{-2}	1.16x10^{-3}
ψ	9.67x10^{-3}	9.19x10^{-4}
$\psi / \psi(\sigma^E)$	87 %	79 %
$\varphi(\varepsilon^T)$	3.27x10^{-2}	1.13x10^{-2}
$[\varphi + k^2(\psi - 2\eta)]/(1 - k^2)$	3.27x10^{-2}	1.13x10^{-2}
$\varphi(\varepsilon^{LS}) \approx \varphi$	3.12x10^{-2}	1.11x10^{-2}
$\varphi / \varphi(\varepsilon^T)$	95 %	98 %
$\eta(d)$	2.75x10^{-2}	8.05x10^{-3}
$[\psi + \varphi - (1 + k^2)\eta)]/(1 - k^2)$	2.75x10^{-2}	8.05x10^{-3}
η	1.48x10^{-2}	4.16x10^{-3}
$\eta/\eta(\delta)$	54 %	52 %

$\varphi(\varepsilon^T)$, and $\eta(d)$), which is fairly reasonable. In Table 2 are also listed the values calculated from the right hand side of eqs.(13)-(15). These values are completely consistent with the extrinsic losses (left had side of the eqs.(13)-(15)), indicating validity of these equations as well as the calculation process used in the present analysis. It should be first noted that the intrinsic loss of piezoelectric constant is not zero. The origin of the intrinsic piezoelectric loss is not clear at present. Uchino and Hirose[9] reported that the intrinsic piezoelectric loss of a PZT base actuator (soft-PZT) in the off-resonance state was between 0.05 and 0.1 depending on the electric field and stress applied on the specimen. This value is a little larger than that determined in the present study (1.48x10^{-2}), but their result seems to be fairly reasonable because the contribution of domain wall motion is enhanced at high fields and at low frequencies.[14]

The intrinsic to extrinsic ratio of each loss was also listed in Table 2. In the case of the elastic loss, the ratio of the intrinsic loss is 87 % in the soft-PZT and it is 79 % for the hard-PZT. However, above 95 % of the dielectric loss is due to the intrinsic loss in both soft- and hard-PZTs. As for the piezoelectric loss, the ratio is much smaller than those in the elastic and dielectric losses. This means that the extrinsic piezoelectric loss ($\eta(d)$) is dominantly derived from the elastic and dielectric losses. Most notable difference between the soft- and hard-PZTs are observed in the elastic loss. The elastic loss of the hard-PZT is one order of magnitude smaller than that of the soft-PZT. The difference in the soft- and hard-PZT is considered to be the degree of clamping of the domain wall motion. This result in indicates that the clamping of domain wall motion markedly reduces the elastic loss but the effects on the dielectric and piezoelectric losses are relatively small.

Figure 3 shows the displacements (k_{31} mode) of the soft- and the hard-PZTs. The displacements show maximum around the resonance frequency. Total displacement of ceramics plate can be obtained by adding the displacement of surface (A) to that of surface (B). It should be noted that the displacement of hard PZT is larger than that of soft PZT in the resonant mode in spite that the piezoelectric d constant of hard PZT is smaller than soft PZT (Table 1). This is because the sharpness of the resonance is mainly determined by the elastic loss. A small elastic loss gives sharp resonance to give a large displacement. In the off-resonant mode, the displacement of soft PZT is larger than that of hard PZT because of the domain contribution. However, in the resonance mode, the domain contribution increased the elastic loss to reduce the displacement.

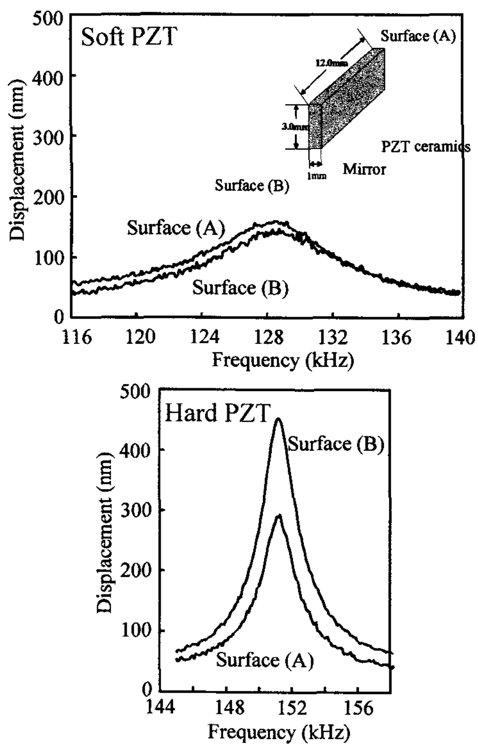

Fig.3 Displacements (k_{31} mode) of soft and hard PZT. Driving voltage was 10V for 1mm thick sample.

4.CONCLUSION

The complex material coefficients of PZT ceramic resonators were determined by the immittance-fitting method. The piezoelectric d-constant should be a complex value to obtain a best fitting between observed and calculated results. A calculation process to determine the intrinsic elastic, dielectric and piezoelectric losses was established in the resonance state. It was confirmed that the intrinsic losses were smaller than the corresponding extrinsic losses. The intrinsic piezoelectric loss had a certain value, indicating that the intrinsic piezoelectric h-constant was a complex value. About 50 % of the extrinsic piezoelectric loss was derived from the elastic and dielectric losses. Most notable difference between the soft- and hard-PZTs were observed in their elastic losses.

The displacement in resonant mode was measured using a newly developed double beam laser Doppler interferometer. The displacement of the hard-PZT is larger than the soft-PZT around the resonance frequency because the small elastic loss.

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