Thermal Conductivities of PZT Piezoelectric Ceramics under Different Electrical Boundary Conditions[i]

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Abstract: Physical properties of polycrystalline lead-zirconate-titanate (PZT) changes according to electrical boundary conditions and poling. This paper reports the thermal properties of poled and unpoled PZT's in the poling direction for open circuit and short circuit conditions. The authors found that the short-circuit condition exhibited the largest thermal conductivity than the open-circuit condition. In the relationship between these two thermal properties, the authors propose the "electrothermal" coupling factor k^{κ}_{33}, which is similar to the electromechanical coupling factor k_{33} relating the elastic compliances under short- and open-circuit conditions. On the other hand, the thermal conductivity of the unpoled specimen exhibits the lowest thermal conductivity, in comparison with the poled specimens, which suggests the importance of phonon mode scattering on the thermal conductivity with respect to elastic compliance.

Keywords: Ferroelectric; Piezoelectric; Thermal Conductivity; Thermal Diffusivity

1. Background

Piezoelectric materials, in which the electrical and mechanical properties are coupled, exhibit significant difference of physical performances according to the boundary conditions: the permittivity changes according to the mechanical clamped or free conditions, and the elastic compliance differs according to the electrical short- or open-circuit conditions. One of the most popular piezoelectric materials is lead-zirconate-titanate (PZT), well-commercialized due to its large electromechanical properties. It is well known that the elastic compliance under open-circuit condition is stiffened by the factor of $(1 - k^2)$ in comparison with the compliance under short-circuit condition, where k stands for the "electromechanical coupling factor"[1]. Thus, it may be expected that the thermal properties, such as thermal conductivity or diffusivity, may also be affected by electrical boundary conditions and poling due to the fact that both the thermal property and elastic compliance arise primarily from phonon mode phenomena.

The research group of this paper at the Pennsylvania State University has been concentrating on the development of high power piezoelectrics, which can handle the power density as high as 40 W/cm^3[2]. One of the Figures-of-Merit for the 'high power density' is the 'maximum vibration velocity', which is determined

by the vibration velocity generated at the sample edge under operation, when the maximum temperature rise (*i.e.*, at the nodal point) reaches 20°C above the room temperature[3]. When we operate the piezo-sample under higher vibration levels, most of the additionally-input electric energy is converted to heat (*i.e.*, PZT becomes a 'ceramic heater'!). Actually, it was found that the maximum vibration velocity is strongly dependent on the thermal conductivity. **Figure 1** demonstrates the simulation results of the saturated temperature distribution profile difference between two compositions, PZT-5H and PZT-19 under the same vibration velocity operating condition. These two compositions exhibit significant difference in thermal conductivity: 0.14 W/m K versus 2 W/m K (more than 10 times difference)[4,5].

The profile curve of PZT-5H (low thermal conductivity) fits a sinusoidal line beautifully, while PZT-19 shows considerable edge temperature rise. You can understand easily that this profile difference is originated primarily from the thermal conductivity or diffusivity difference. Taking the total thermal energy dissipated from the specimen by integrating the temperature rise with respect to the position coordinate, a similar mechanical quality factor Q_m value for these two samples can be concluded. However, the peak temperature at the nodal point is significantly lower for the larger thermal conductivity material, which means there will be more vibration under higher voltage for the PZT-19, since the maximum vibration velocity is defined by the highest temperature rise 20°C above room temperature.
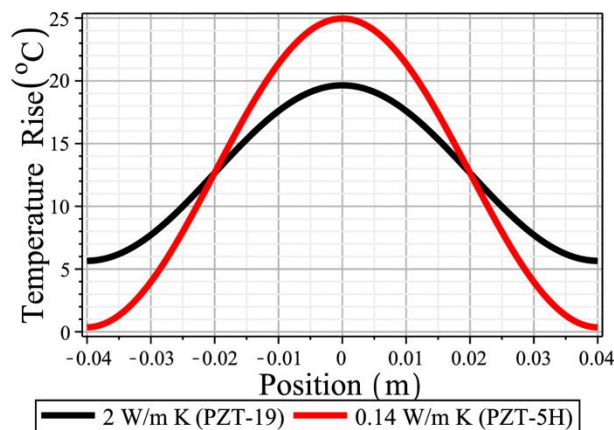
Figure 1. Saturated temperature distribution profile difference between PZT-5H and PZT-19.

This is the motivation on the thermal conductivity/diffusivity measurements on piezoelectric ceramics from the practical application viewpoints.

2. Thermal diffusivity measurements

Previous papers[6,7] which report on the thermal properties of PZT ceramics at low temperature (20K-300K), indicate a transition temperature between 50K and 80K, and at high temperature (300K-800K), characterizing the effect of phase transition on the thermal properties. However, in the authors' knowledge, the relationship of thermal properties with electrical boundary conditions and poling in PZT and other ferroelectric piezoelectric ceramics has not been systematically studied yet.

There have been several methods reported for measuring thermal diffusivity using either "steady" state or "transient" techniques. Steady state methods in general provide larger experimental error and inaccuracies, while "transient" techniques, namely the laser flash method, are expensive and require specialized equipment and advanced data analysis. In this paper a new experimental setup has been used for measuring the thermal diffusivity of PZT's. Though Ref. [8] reported the details of the experimental setup, it has been summarized briefly in this paper for the reader's sake (see **Figure 2**). Hot isothermal and insulating boundary conditions are imposed on a flat disk sample in our experiments. The transient temperature profile of the insulated side of the sample is analytically similar to an exponential formulation with a classic time constant. The thermal diffusivity is given proportional to the "inverse time constant". This method hosts a variety of advantages

over other methods such as high accuracy, low cost, elimination of interface effects, and small sample size.

The accuracy of the setup used in this paper has been already demonstrated in several materials with low to medium thermal diffusivity (0.1→3 mm²/s). The thermal diffusivity measurements in this experiment have an accuracy of 5% or better for some standard materials such as Fused Quartz and Pyrex 7740, in comparison to the literature values[8].
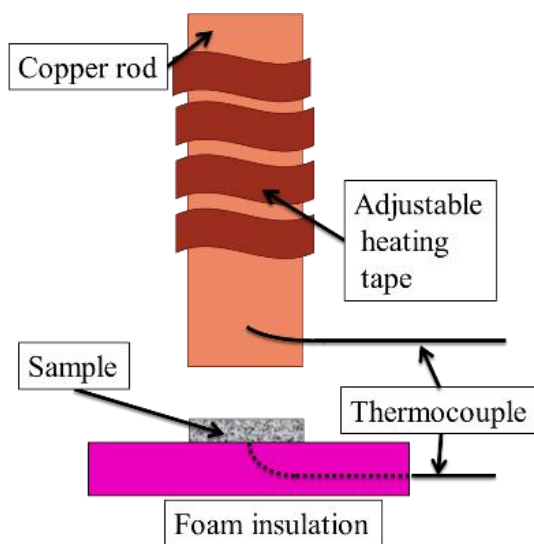
Figure 2. Experimental Setup for determining directional thermal diffusivity.

Using this method, the thermal diffusivity α was measured on "poled" and "unpoled" commercially-available hard PZT ceramic discs, APC 841 (APC Int., USA), of a diameter of 51 mm. The 'unpoled' sample was depoled by heating them, annealing, and then checking their piezoelectric response on a conventional d_{33} meter. The thermal diffusivity was measured in the direction of polarization for the poled samples. In parallel, the absolute value heat capacity of a small sample was measured using the DSC Q2000 (TA Instruments), by comparing it to a sapphire reference sample. The heat capacity and density are respectively c_p = 340 J/kgK and ρ = 7600 kg/m³. Interestingly, the heat capacity is not affected by electrical boundary conditions and it is a "scalar" property; therefore, the thermal conductivity κ can be determined from the relation given by thermal diffusivity α and the heat capacity c_p and mass density ρ as

$$\kappa = \alpha \, c_p \, \rho. \qquad (1)$$

Taking into account a standard deviation of less than 5% of the experimental setup, the results of the PZT specimen experiments summarized in **Table 1** seem to show larger experimental deviations, probably due to the fact that the diameters of the PZT specimens were a few millimeters smaller than the sample holder. The error found is different for different boundary conditions and poling states, but this deviation is believed to be random. Nevertheless, it is essential to note that the thermal diffusivity/conductivity difference among the three PZT specimens (open-, closed-circuit poled, and unpoled samples) is much larger than the experimental deviation.

Hard PZT	Thermal diffusivity $\alpha_{experiment}$ (10 mm²/s)	+/-	Thermal conductivity $\kappa_{experiment}$ (W/m K)	+/-
Open Circuit	5.02	0.23	1.4	0.06
Closed Circuit	8.25	0.78	2.3	0.23
Unpoled	4.32	0.34	1.2	0.10

Table 1. Thermal diffusivity and thermal conductivity of a Hard PZT, for unpoled and poled under different electrical boundary conditions

Table 2 summarized the preliminary data on the thermal conductivity in the NKN-based material with the PZT's value (both unpoled samples). Pb-free piezoceramics such as $(Na,K)NbO_3$- and $(Bi,Na)TiO_3$- based materials show much higher maximum vibration velocity than the PZT's[9,10]. Much larger thermal conductivity in the NKN-based material than the PZT's, as shown in **Table 2**, may also contribute to this good high-power performance in NKN-ceramics.

Thermal Properties	c_p (J/g/K)	κ (W/m/K)
Hard-PZT	0.42	1.25
NKN-Cu	0.58	3.10

Table 2. Thermal properties of NKN-Cu in comparison with Hard PZT

3. 'Electrothermal coupling' factor

The difference among the measured thermal diffusivity and thermal conductivity values for poled open-circuit, poled short-circuit, and unpoled specimens (**Table 1**) is discussed as follows. First, the short-circuit thermal conductivity κ^E_{33} is 1.6 times larger than the open circuit one κ^D_{33}. Denote the superscripts E and D of the conductivity κ, supposing that the sample is roughly maintained "electric field constant" and "electric displacement constant" along the thermal conduction direction during a rather short experimental time period less than 60 seconds. Second, the unpoled thermal conductivity κ^u showed the smallest value, 15% less than that of the open circuit case.

The authors would like to define a new "electrothermal coupling coefficient" k^κ_{33} below, in the relationship between the open-circuit κ^D_{33} and short-circuit thermal conductivity κ^E_{33}:

$$\kappa^E_{33} (1-(k^\kappa_{33})^2) = \kappa^D_{33} \qquad (2)$$

This definition is analogously related to the relationship between short-circuit and open-circuit elastic compliance in a piezoelectric with the electromechanical coupling coefficient k_{33}[11]:

$$s^E_{33}(1- k_{33}^2) = s^D_{33} \qquad (3)$$

The electromechanical coupling factor of this Hard PZT ceramic found from electrical impedance spectroscopy is k_{33}= 0.68, which is equivalent to Eq. (3). Using Eq. (2), the "electrothermal" coupling factor can be calculated to be k^κ_{33} = 0.63. The authors would like to point out rather close numbers between k_{33} and k^κ_{33}, even taking into account the error between the two coupling factors due to error in the thermal measurements and possibly other microscopic features which do not correlate between the k value determined from electrical and thermal measurements.

4. Discussion

Thermal conductivities of PZT piezoelectric ceramics were investigated under different electrical boundary conditions (poled under short-circuit, poled under open-circuit, and unpoloed specimens) and obtained the relation, $\kappa^E_{33}> \kappa^D_{33}> \kappa^u$.

It may be speculated the reasons from the aspects of phonon mode scattering, orientation of domains, and elastic compliance. Remember the fact that ferroelectric-originated piezoelectric materials exhibit the phase transition associated with phonon mode "softening"[11]. First, because of the random orientation of domains in the unpoled sample, it is expected that there will be the most phonon scattering at the domain boundaries in this specimen, which is related with acoustic mismatch at the domain boundaries. Therefore, it will have the lowest thermal conductivity. Second, with increasing the degree of domain orientation, less scattering is expected in the poled specimens and thermal conductivity will be larger for the poled material (κ^E_{33} and κ^D_{33}). Third, because the elastic compliance under the short-circuit condition (s^E_{33}) is softer than the elastic compliance under open-circuit conditions (s^D_{33}) via the relation $s^E_{33}(1- k_{33}^2) = s^D_{33}$, the lattice and domain wall dynamic motion are expected to be larger in the polarization direction under the short-circuit condition, probably originated from no-"depolarization field". To the contrary, the depolarization field (*i.e.* reverse field

against the spontaneous polarization direction) under the open-circuit condition stiffens the elasticity, and reduces phonon mode excitation. You may also consider that the thermal 'pyroelectric' charges on the surface create the depolarization field under open-circuit condition,

$$E_{dep} = -\frac{\Delta P}{\varepsilon_0 \varepsilon}$$

The larger lattice vibration and domain wall dynamics in the short-circuit condition may also introduce a larger thermal conductivity in short-circuit condition (κ^E_{33}) due to increased phonon mode transport. This speculation can also suggest the analogous relations among two coupling factors, electromechanical k_{33} and electrothermal k^{κ}_{33}.

It can be concluded in the experiments that thermal conductivity in ferroelectric ceramics depends on electrical boundary conditions. It is highly expected that the electromechanical coupling factor in these materials is related to electrothermal properties as well, namely thermal conductivity and thermal diffusivity. This paper provided a new concept on electrothermal coupling coefficient k^{κ}_{33}, and discussed the origin using phonon scattering and domain orientation concepts.

Electrothermal coupling may be categorized as primary and secondary effects, in general. The primary effect includes "pyroelectric" and "electro-caloric" phenomena, and the secondary effect includes the space-gradient (*i.e.*, first-derivative) phenomena, such as "thermal conductivity". The authors are expecting further theoretical expansion in this area.

Acknowledgments

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