**The Effect of Electrical Boundary Conditions on the Thermal Properties of Ferroelectric Piezoelectric Ceramics**

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**Abstract**

*The thermal conductivity of polycrystalline bulk PZT (lead-zirconate-titanate) has been investigated according to electrical boundary conditions and poling. The thermal conductivity of poled PZT was measured in the poling direction for open circuit and short circuit conditions. The short circuit thermal conductivity had the largest thermal conductivity. The relationship between these two thermal properties, the “electrothermal” coupling factor k κ33, was found to be similar to the electromechanical coupling factor k33 relating elastic compliance under short circuit and open circuit conditions. The thermal conductivity of the unpoled sample was found to have the lowest thermal conductivity. The significance of the thermal conductivity with regards to phonon mode scattering and elastic compliance was discussed.*

**Background**

Piezoelectric materials are a unique class of materials in which the electrical and mechanical properties are coupled. The most popular piezoelectric material is lead-zirconate-titanate (PZT) due to its large electromechanical properties and the adaptability of its properties with dopants.

It is well known that the electrical boundary conditions and poling affect the elastic compliance of piezoelectric materials.1 The thermal properties, namely thermal conductivity, may also be expected to be affected by electrical boundary conditions and poling due to the fact that this property and elastic compliance both arise primarily from phonon mode phenomena.

Our research group 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/cm3.2 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 (at the nodal point) reaches 20°C above the room temperature.3 When we operate the piezo-sample under higher vibration level, most of the additionally-input electric energy is converted to heat (PZT becomes the ‘ceramic heater’!). We 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 The profile curve of PZT-5H 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, we can expect a similar mechanical quality factor *Qm* value for these two samples. However, you can notice that the peak temperature at the nodal point is significantly lower for the larger thermal conductivity material, which means we can excite the vibration more 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 our motivation on the thermal conductivity/diffusivity measurements on piezoelectric ceramics.

**Thermal Diffusivity Measurements**

The thermal properties of PZT ceramics have been studied at low temperature (20K-300K), and a transition temperature was found between 50K and 80K.5 It has also been studied at high temperature, between 300K-800K, which characterizes the effect of phase transition on the thermal properties.6 However, the relationship of thermal properties with electrical boundary conditions and poling in PZT and other ferroelectric piezoelectric ceramics has not been studied.

Many methods exist to measure thermal diffusivity using either steady state or transient techniques. Steady state methods yield large experimental error and inaccuracies. Transient techniques, namely the laser flash method, are expensive and require specialized equipment and advanced data analysis. In this paper a novel experimental setup was used to evaluate thermal diffusivity. Figure 2 shows our thermal diffusivity measurement setup. In this experiment hot isothermal and insulating boundary conditions are imposed on a flat disk sample. The transient temperature profile of the insulated side of the sample is analytically similar to a classic time constant formulation. The thermal diffusivity is 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.

Several materials with low to medium thermal diffusivity (0.1→3 mm2/s) have been measured. The diameter of the sample is 32 mm and its thickness ranges from 2 to 6.5 mm. 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.7



Figure 2: Experimental Setup for determining directional thermal diffusivity

Using this method, the thermal diffusivity α of a poled and depoled commercially available hard PZT ceramic discs, APC 841 (APC Int., USA), of a diameter of 51 mm was measured. The thermal diffusivity was measured in the direction of polarization for the poled samples. Using the DSC Q2000 (TA Instruments), the absolute value heat capacity of a small sample was measured by comparing it to a sapphire reference sample. The heat capacity and density are respectively cp = 340 J/kgK and ρ = 7600 kg/ m3. 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 thermal diffusivity α and the heat capacity cp as

κ = α cp ρ . (1)

Though the previous report regarding the thermal diffusivity experiment on some standard materials presented a standard deviation of less than 5%, the results of this experiment had larger deviation as shown in Table 1, probably due to the fact that the diameters of the samples were a few millimeters smaller than the sample holder. The error found is different for different boundary conditions and poling states, but this is believed to be random.

Table 2 summarized the thermal conductivity in the NKN-based material with the PZT’s value (both unpoled samples). Pb-free piezoceramics such as (Na,K)NbO3- and (Bi,Na)TiO3- based materials show much higher maximum vibration velocity than the PZT’s.8,9 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.

Table 1: Thermal diffusivity and thermal conductivity depending on electrical boundary conditions

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| **Hard PZT** | Thermal diffusivity$$α\_{experiment}$$(10 mm2/s) | +/- | Thermal conductivity$$κ\_{experiment}$$(W/m K) | +/- |
| Open Circuit | 5.02 | 0.23 | 1.4 | 0.06 |
| Closed Circuit | 8.25 | 0.78 | 2.3 | 0.23 |
| Depoled | 4.32 | 0.34 | 1.2 | 0.10 |

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

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

**‘Electrothermal Coupling’ Factor**

The measured thermal diffusivity and thermal conductivity values for poled open circuit, poled short circuit, and depoled samples are described in Table 1 as an average of two measurements on three samples each. The samples were depoled by heating them and then checking their response on a conventional d33 meter. The short-circuit thermal conductivity κE33 is more than 1.5 times larger than the open circuit one κD33. The unpoled thermal conductivity κu showed the smallest value, 15% less than that of the open circuit case. The relationship between the open-circuit κD33 and short-circuit thermal conductivity κE33 can be described by an electrothermal coupling coefficient

κE33 (1-( kκ33)2) = κD33, (2)

which is closely related to the relationship between short-circuit and open-circuit elastic compliance and the electromechanical coupling coefficient k33 10

sE33(1- k332 ) = sD33. (3)

The electromechanical coupling factor of this ceramic found from electrical impedance spectroscopy is k33= 0.68. Using Eq. (2), the “electrothermal” coupling factor can be calculated to be kκ33 = 0.63. The error between the two coupling factors may be 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.

**Discussion**

In summary, κE33> κD33> κu. This may be understood from phonon mode scattering, orientation of domains, and elastic compliance. Because of the random orientation of domains in the depoled sample, it is expected that there will be the most phonon scattering in this material. Therefore, it will have the lowest thermal conductivity. Because the domains of the poled material are oriented, less scattering is expected and thermal conductivity will be larger for the poled material (κE33 and κD33). The elastic compliance under short-circuit conditions sE33 is softer than the elastic compliance under open-circuit conditions sD33. This means that the lattice and domain wall motion are larger in the short-circuit condition, probably originated from no-depolarization field. The larger lattice vibration and domain motion in short-circuit conditions will also correlate to a larger thermal conductivity in short-circuit conditions κE33 due to increased phonon mode transport. This observation can also be used to understand the relation between k33 and kκ33

The clear result of the experiments is that thermal conductivity in ferroelectric ceramics depends on electrical boundary conditions. It is very possible that the electromechanical coupling factor in these materials is related to thermal properties as well, namely thermal conductivity and thermal diffusivity. A discussion was presented to explain the behavior using phonon scattering and domain orientation concepts. Future work includes studying the effect of microstructure on phonon mode transport in these materials and further clarifying the thermal-electrical coupling experimentally demonstrated in the experiments.

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