SAND2007-4751C

Presented at the 15th APS Topical Conference on Shock Compression of Condensed Matter,
June 25 - 29, 2007, Kohala Coast,
To appear in AIP Conference Proceedings, 2008

INITIAL TEMPERATURE EFFECTS ON THE DIELECTRIC PROPERTIES OF PZT 95/5 DURING SHOCK COMPRESSION

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Abstract. A strong electric field can be generated when the shock-induced depoling current from a normally poled PZT 95/5 sample is passed through a large resistive load. The portion of total depoling current that is retained on the sample electrodes to account for capacitance is governed by the dynamic dielectric properties of both unshocked and shocked material. Early studies used measured load currents from single samples to assess models for dielectric response. In more recent studies, we used shock-driven circuits in which multiple PZT 95/5 elements were displaced both parallel and perpendicular to the shock motion. This allowed both load and charging currents to be measured for individual elements that are subjected to shock compression and release at different times. In the present study, these techniques have been utilized to examine dielectric properties in PZT 95/5 samples at initial temperatures from -56 to 74 °C. Significant changes in permittivity with temperature are observed in both unshocked and shocked samples. Measured currents show a complex dielectric response which can only be partially predicted using a simple dielectric relaxation model.

Keywords: ferroelectrics, phase transitions, dielectric properties, temperature effects
PACS: 62.50.+p, 77.84.Dy, 77.22.Ej

INTRODUCTION

When poled by an applied electric field, a ferroelectric ceramic retains bound charge associated with remanent polarization. A particular lead zirconate/titanate composition of interest, denoted by PZT 95/5, has a Zr:Ti ratio of 95:5 and is typically modified with 2% niobium. This composition is in a ferroelectric phase at ambient conditions but is near the boundary for an anti-ferroelectric phase. Shock compression into this adjacent phase provides a fast mechanism for releasing the bound charge. In a "normal mode" configuration, a planar shock propagates in a direction perpendicular to the polarization axis. If the resulting depoling current is passed through an external circuit with a large resistive load, strong electric fields can be generated within the PZT 95/5 sample. Under these conditions, a portion of the released charge will be retained on the sample electrodes to account for capacitance. This effect is governed by the dielectric properties of both unshocked and shocked PZT 95/5 during shock transit, and the retained charge is not available for delivery to the external circuit. Earlier studies of shock-driven load currents from normal-mode PZT 95/5 samples (1-5) assumed constant but different values for permittivity on either side of the shock front, or used simple models for dielectric relaxation. In all of these studies, however, the measured load currents did not provide a means to directly determine the separate dielectric response of unshocked and shocked material. In addition, it was not possible to distinguish between changes in dielectric properties and possible field effects on the depoling kinetics.

In more recent studies, new experimental configurations were utilized to better elucidate dielectric properties in both unshocked and shocked PZT 95/5 (6). Experiments examined shock-driven circuits containing multiple, small PZT 95/5 elements that were displaced both parallel and perpendicular to the shock motion. This allowed both load and charging currents to be measured for individual elements that are subjected to shock compression and release at different times. In the present study, similar experiments are conducted with the target assemblies at initial temperatures that vary from -56 to 74 °C. Small-signal permittivity measurements over this temperature range (7) indicate that significant temperature effects could be observed.

EXPERIMENTAL CONFIGURATION

The basic experimental configuration used in the present study is shown in Fig. 1. Two PZT 95/5 samples are mounted within a target assembly consisting of unfilled epoxy elements. The target

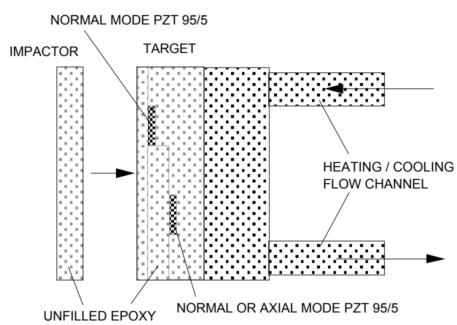

FIGURE 1. Configuration for hot and cold experiments on PZT 95/5 dielectric properties.

temperature is monitored using thermocouples embedded within these elements. A copper flow channel is incorporated into the assembly through which either heated water or chilled ethanol is circulated. Final target temperatures are achieved over a 3-4 hour period.

A particular configuration was used to examine just the dielectric properties of poled, unshocked material. The first PZT 95/5 sample in the target is a normal-mode bar having dimensions 2 mm by 10-20 mm (electroded surfaces) by 4-5 mm. Following shock transit through this bar, some time elapses before the shock reaches a PZT 95/5 disk 2 mm thick, 18-27 mm in diameter, and poled in the direction of shock motion ("axial mode"). Figure 2 shows the corresponding electrical circuit. The

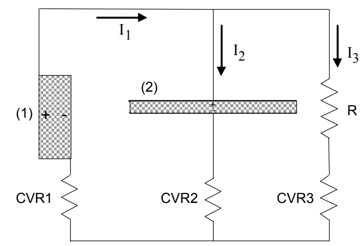

FIGURE 2. Circuit for a target configuration to examine dielectric properties in poled, unshocked PZT 95/5.

PZT bar is connected to the disk and a large load resistor (10 kΩ) in parallel. The dimensions of the disk are chosen so that its initial capacitance is much larger than that of the first bar. Thus, the depoling current from the bar acts like a current source driving the rest of the circuit. Current-viewing resistors (CVRs) are used to measure currents flowing in each circuit segment. Shock-induced conduction typical of axial-mode response (5) occurs within the disk soon after shock arrival, and no additional information on dielectric properties can be obtained after this time.

Figure 3 shows currents measured in a room-temperature experiment using this configuration.

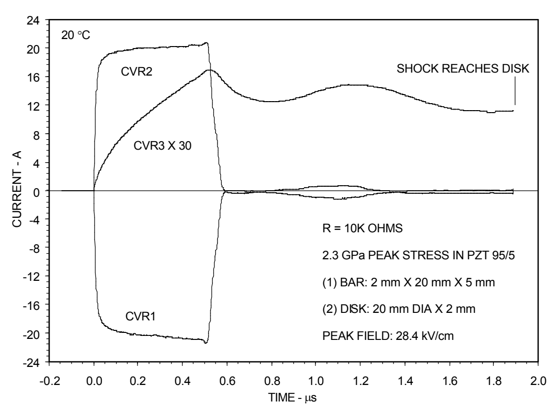

FIGURE 3. Measured currents in a room-temperature experiment using the shock-driven circuit shown in Fig. 2.

The bottom curve (CVR1) is the depoling current from the PZT bar, and shows a small amount of late-time depoling due to wave reverberations. The load current (CVR3) rises nonlinearly and shows a pronounced relaxation after shock transit followed by a disproportionate response to the late-time depoling. In a previous study (6), the circuit shown in Fig. 2 was analyzed by assuming the displacement current (I_2 in Fig. 2) corresponded to a simple relaxing dielectric according to the Debye approximation:

$$\frac{dD_I}{dt} = \varepsilon_i \frac{dE}{dt} - (D_I - \varepsilon_{eq} E)/\tau$$

where D_I is the field-induced electric displacement, ε_i and ε_{eq} are the instantaneous and equilibrium permittivities, E is the electric field, and τ is the relaxation time. Figure 4 shows comparisons between the load current shown in Fig. 3 and a current predicted using the Debye approximation. A different PZT bar configuration in the previous study did not produce late-time depoling, and predicted load currents matched measured currents fairly well with small adjustments of the Debye parameters. The predicted current in Fig. 4 again matches the measured current fairly well until late-time depoling occurs, then fails to reproduce the resulting behavior. The dominant dielectric response is domain reorientation, and the simple relaxation model appears to adequately reproduce that response. When late-time depoling occurs, however, the resulting dielectric response corresponds to a much smaller permittivity, as if additional domain reorientation is not possible and a different, weaker polarization process governs this response. Also shown in Fig. 4 is a current predicted with a preliminary model that accounts for a subsequent small-permittivity response, but clearly further model development will be required.

Figure 5 shows the permittivity behavior using Debye-approximation fits to load currents measured in experiments conducted at initial temperatures of 73 °C, 20 °C (Fig. 4), and -56 °C. The initial permittivity values rise with initial temperature, but the equilibrium values show only a significant reduction in the cold case. Peak fields varied from 28.4 to 29.7 kV/cm in these experiments.

A second configuration was used to examine the dielectric properties of shocked, depoled PZT 95/5.

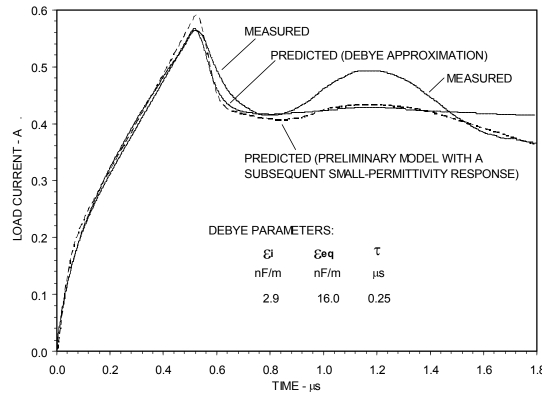

FIGURE 4. Comparison between the measured load current shown in Fig. 3 and a current predicted using the Debye approximation. Also shown is a current predicted with a preliminary model that allows for a subsequent small-permittivity response.

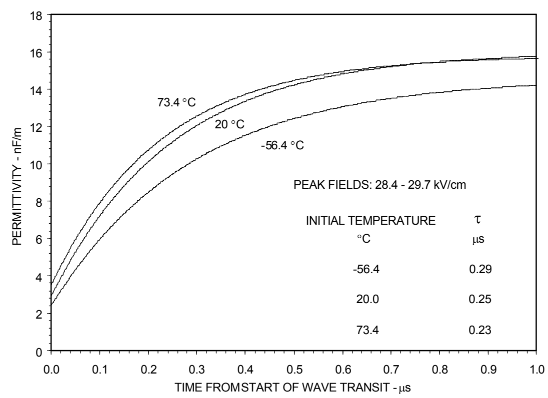

FIGURE 5. Permittivity histories corresponding to Debye-approximation fits to measured load currents in experiments conducted at different initial temperatures.

The axial-mode disk used in the first configuration is replaced by a normal-mode PZT bar identical to the first bar, and circuit currents are recorded past shock transit of the second bar. Both shock transits produce similar depoling currents, but the second results in capacitive charging of a shocked, depoled sample. Figure 6 shows currents measured in a 20 °C experiment using this target configuration. The depoling currents are nearly the same, as are the load currents (CVR3). The displacement current during the second shock transit (CVR1) is much less than during the first transit (CVR2), showing that the dielectric response of shocked material is

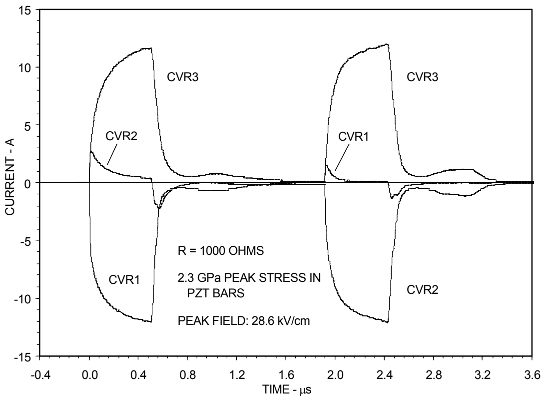

FIGURE 6. Measured currents in a room-temperature experiment using identical, normally poled PZT 95/5 bars for the both the first and second target elements.

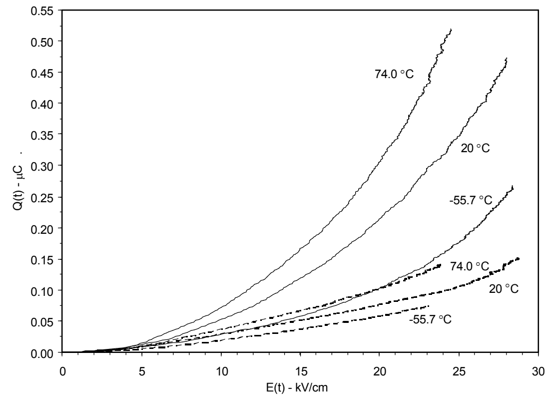

FIGURE 7. Displacement charges plotted versus electric fields present at corresponding times. Room-temperature results are from measured currents shown in Fig. 6. Solid lines correspond to shock transit through the first PZT bar, and dashed lines correspond to shock transit through the second bar.

significantly smaller. These displacement currents can be integrated in time to give the total displacement charge at any time during shock transit. Figure 7 shows displacement charges plotted against the field values that existed at a corresponding time. In addition to the results from Fig. 6, this figure shows results from experiments conducted at initial temperatures of 74 °C and -56 °C. Dielectric response decreases rapidly with decreasing temperature, although this trend appears to be greater in unshocked than in shocked PZT.

SUMMARY

Experimental configurations utilizing multiple PZT 95/5 elements have been used to examine initial temperature effects on the dielectric response of both unshocked and shocked material. In experiments on unshocked PZT, a simple dielectric relaxation model could predict the primary response but not a subsequent secondary response. Fits to the primary response obtained at different temperatures showed a strong effect only in the cold case. Experiments using a different target configuration showed a much weaker dielectric response in shocked material. Decreasing dielectric response with decreasing temperature was more pronounced in unshocked material.

ACKNOWLEDGMENTS

Sandia is a multiprogram laboratory operated by Sandia Corporation, a Lockheed Martin Company, for the United States Department of Energy's National Nuclear Security Administration under Contract DE-AC04-94AL85000.

REFERENCES

1. Lysne, P. C., and Percival, C. M., *J. Appl. Phys.* **46**, 1519-1525 (1975).
2. Lysne, P. C., *J. Appl. Phys.* **48**, 1020-1023 (1977).
3. Lysne, P. C., *J. Appl. Phys.* **48**, 4565-4568 (1977).
4. Lysne, P. C., "Electrical Response of Shock-Wave-Compressed Ferroelectrics," in *High Pressure Science and Technology, Vol. I*, edited by K. D. Timmerhaus and M. S. Barber, Plenum Press, New York, 1978, pp. 202-209.
5. Setchell, R. E., J. Appl. Phys. **97**, 013507 (2005).
6. Setchell, R. E., Montgomery, S. T., Cox, D. E., and Anderson, M. U., "Dielectric Properties of PZT 95/5 During Shock Compression Under High Electric Fields," in Shock Compression of Condensed Matter-2005 (M.D. Furnish et al., eds.), part I, pp. 278-281.
7. Avdeev, M. et al., Phys. Rev. B **73**, 064105 (2006).