RECENT PROGRESS IN UNDERSTANDING THE SHOCK RESPONSE OF FERROELECTRIC CERAMICS

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Abstract. Ferroelectric ceramics exhibit a permanent remanent polarization, and shock depoling of these materials to achieve pulsed sources of electrical power was proposed in the late 1950s. During the following twenty years, extensive studies were conducted to examine the shock response of ferroelectric ceramics primarily based on lead zirconate titanate (PZT). Under limited conditions, relatively simple analytical models were found to adequately describe the observed electrical behavior. A more complex behavior was indicated over broader conditions, however, resulting in the incorporation of shock-induced conductivity and dielectric relaxation into analytical models. Unfortunately, few experimental studies were undertaken over the next twenty years, and the development of more comprehensive models was inhibited. In recent years, a strong interest in advancing numerical simulation capabilities has motivated new experimental studies and corresponding model development. More than seventy gas gun experiments have examined several ferroelectric ceramics, with most experiments on lead zirconate titanate having a Zr:Ti ratio of 95:5 and modified with 2% niobium (PZT 95/5). This material is nominally ferroelectric but is near an antiferroelectric phase boundary, and depoling results from a shock-driven phase transition. Experiments have examined unpoled, normally poled, and axially poled PZT 95/5 over broad ranges of shock pressure and peak electric field. The extensive base of new data provides quantitative insights into both the stress and field dependencies of depoling kinetics, and the significance of pore collapse at higher stresses. The results are being actively utilized to develop and refine material response models used in numerical simulations of pulsed power devices.

INTRODUCTION

A ferroelectric ceramic exhibits a remanent polarization when poled by an electric field. The bound charge associated with this polarization was recognized as a means of achieving pulsed electrical power more than forty years ago (1). Early studies examined how shock waves could release the bound charge in barium titanate and in other materials based on solid solutions of lead zirconate and lead titanate (2-4). These studies used axial mode configurations, where shock propagation occurs along the poling axis. Issues that were considered included shock-induced conduction and whether charge release resulted from domain reorientation or through phase transitions. Extensive studies followed during the 1970s, primarily on lead zirconate titanate ceramics. Axial mode studies continued, primarily with PZT 65/35 (5), but the majority of experiments were done with normal mode configurations in which shock propagation occurs in a direction perpendicular to the poling axis. These studies focused on ceramics having a Zr:Ti ratio of 95:5 and modified with 2% niobium, subsequently referred to as PZT 95/5 (6-8). The nominal state of this material is ferroelectric (FE), but it is near an antiferroelectric (AFE) phase boundary. Bound charge is released through shock compression into the AFE phase. A simple model for the depoling current was used by Lysne (6) to analyze external circuit voltages generated across different resistance loads during shock propagation. This model assumed instantaneous and complete depoling at a discontinuous shock front, and the
released charge was partitioned between passing through the external circuit and being retained on the sample electrodes to account for capacitance. Dielectric constants were assumed to differ between shocked and unshocked material. This model was fairly successful in predicting measured voltages at shock pressures sufficient to completely depole the PZT 95/5 samples. To improve comparisons, particularly with experiments having inductive loads, Lysne added finite resistivity (9) and dielectric relaxation (10) to his analysis.

A period of nearly twenty years followed in which few studies were conducted on the shock response of ferroelectric ceramics. About five years ago, however, a strong interest developed in establishing a capability for numerically simulating the operation of pulsed power devices that utilize PZT 95/5. This interest has motivated significant new experimental and analytical efforts to understand the complex behavior of this material. Some of the resulting experimental work has been previously reported (11-13). The purpose of the present paper is to provide a selected summary of experiments performed to date which have been most useful for gaining insights into the mechanical and electrical response of PZT 95/5 under shock loading.

HUGONIOT STATES

Although a Hugoniot curve based on available transmitted-wave data was reported previously (11), a more accurate determination was desired over the stress range of interest. The reverse-impact configuration used for these measurements is shown in Fig. 1. Samples of unpoled PZT 95/5 having a nominal density of 7.30 g/cm³ were impacted into a fused silica window, and the particle velocity at the impact interface was determined using velocity interferometry (VISAR). PZT 95/5 at this density has a pore volume fraction of approximately 9%. Typical profiles recorded at different impact velocities are shown in Fig. 2. The top profile in this figure shows a transient overshoot, characteristic of impacts resulting in stress states above the threshold for the onset of pore collapse. Figure 3 shows the Hugoniot curve resulting from this series of experiments. The complex curve has multiple reversals in curvature resulting from a FE phase with anomalous behavior, an extended mixed-phase region, and the onset of pore collapse.
NORMALLY POLED EXPERIMENTS

A more complex experimental configuration is required for investigating the response of normally poled PZT 95/5 samples, as shown in Fig. 4.

Although this configuration has been described previously (12), one change used in more recent experiments should be noted. For introduction of a sharp shock jump into the PZT 95/5 sample, a sapphire disc on the projectile face is impacted into a similar disc in the target. The resulting shock discontinuity propagates through the target disc and into the PZT 95/5 sample. If the sapphire elements are replaced by 828/Z ALOX, an alumina-filled epoxy (14), the inelastic response of this material results in an extended wave front having a rise time of several hundred nanoseconds. The resulting input wave into a PZT 95/5 sample is shown in Fig. 5, and will be referred to as a "ramp" input.

Figure 6 shows waveforms recorded after input shocks had propagated through 4.0 mm of normally poled material, under short-circuit conditions. The wave having a final stress of 0.93 GPa shows an extended structure consistent with the lower part of the Hugoniot curve (Fig. 3), which has negative curvature. The 1.8 GPa case has a two-wave structure corresponding to the phase transition. At shock stresses of 2.5 GPa or higher, a distinct plateau is evident at a state corresponding to 2.2 GPa in PZT 95/5. This plateau identifies the onset of pore collapse in the ceramic. At higher stresses the velocity slowly rises from the plateau value to the final state that would be predicted from the Hugoniot curve. Useful VISAR data typically terminates before the final state is reached, due to the transmitted shock wave in the sapphire window reaching the window’s free surface (approximately 1.1 μs after the start of the waveform).

Figure 7 shows waveforms recorded with 2.5 GPa input shocks, with the different curves corresponding to increasing load resistance in the external circuit. A field of 37 kV/cm is reached in the sample for the highest load resistance. The transmitted waveforms show essentially no change in their final state, indicating that electromechanical effects on Hugoniot states are not apparent under these conditions.

Figure 8 shows short-circuit currents generated during the transit of shocks having different strengths. At a shock stress of 2.4 GPa, a very flat current profile is recorded. The level of this current
is accurately predicted by the simple model (6):
\[
I_{sc} = U_s P_r L
\]  

where the short-circuit current $I_{sc}$ simply equals the product of the shock velocity $U_s$, the remanent polarization $P_r$, and the electrode dimension $L$ that is perpendicular to the shock direction. This model represents complete, instantaneous depolarization by a steady shock discontinuity. The overshoots appearing at higher stresses as the shock enters the PZT 95/5 are possibly a transient effect associated with the onset of pore collapse. The fact that higher stresses do not result in higher average currents will be discussed in a subsequent section. As input shock stresses are decreased, complete depoling is inhibited and current levels during shock transit drop rapidly. The high-impedance sapphire window at the back of the PZT 95/5 sample (Fig. 4) results in a reflected shock propagating back into the sample, hence the continuing depoling later in time.

Similar short-circuit currents are shown in Fig. 9, except that shock input cases are compared with ramp inputs. At a peak stress of 2.5 GPa the differences are small, with the ramp case showing a slightly slower rise and a smaller average value during wave transit. At a shock stress of 0.9 GPa the differences are quite large, with depoling rates much lower for the ramp input.

Figure 10 shows currents generated during the transit of 2.5 GPa shocks, with the different curves corresponding to increasing resistance loads in the external circuit. The current through a finite load results in a voltage drop across the electrodes of the PZT 95/5 sample, and the capacitance of the sample dictates that some of the initially bound charge be retained. This results in the "RC" time constant shown by the current rise, and these profiles can be
adequately predicted by a simple depoling model (6). At lower shock pressures, however, such a model cannot account for the inhibited depoling of the material. Figure 11 shows a comparison of currents generated with waves having a peak stress of 0.9 GPa. The current resulting from a shock input is significantly reduced throughout the wave transit when a large load is added. The bottom profile shows a further reduction in current when a large load and a ramp input are combined.

**FIGURE 11.** Currents generated by 0.9 GPa shock and ramp waves with different loads.

**EFFECTIVE SHOCK STATES OBTAINED FROM SHORT-CIRCUIT CURRENTS**

The history of charge release in short-circuit experiments is found by simply integrating the current over time. For experiments conducted at shock pressures of 2.5 GPa or higher, the charge divided by the electrode area rises linearly to a final value corresponding to the initial remanent polarization, as shown in Fig. 12. This provides an accurate measure of the initial polarization. Using the simple model for depoling given by Eq. (1), the slope of this curve (the average current during shock transit) can be used to find an “effective” shock velocity. The product of the initial density and this velocity defines a linear curve in a stress, particle velocity space. Using normal impedance-matching procedures, the intersection can be found between this line and a left-facing Hugoniot curve for sapphire originating at the impact velocity. Note that conditions produced in a sample by the transmission of a wave generated by symmetric impact (in an elastic material) are the same as if the impactor strikes the sample directly at the same impact velocity. Figure 13 shows a comparison between the effective stress-velocity states found in this manner and the Hugoniot states found in the reverse-impact experiments. A curve fitted to the effective states progressively separates from the Hugoniot curve at stresses above the threshold for pore collapse. This curve slowly deviates from a linear curve starting at the origin, indicating that the effective shock velocities are increasing very slowly with increasing stress.

The effective stress-velocity states correspond to the plateau condition shown by transmitted wave profiles at the higher stress conditions (Fig. 6). This part of the wave structure is responsible for depoling the PZT 95/5, effectively decoupling the depoling process from the final states achieved following pore collapse.

**FIGURE 12.** Using the simplest model for depoling, an effective shock velocity can be found by integrating short-circuit currents.

**FIGURE 13.** The effective stress-velocity states shown with the Hugoniot curve found from reverse-impact experiments.
SUMMARY

The experimental results presented in this paper represent an ongoing effort to understand the fundamental behavior of PZT 95/5 under carefully controlled, planar impact conditions. Two aspects of this behavior have been emphasized. The first is the reduction in depoling rates by decreasing shock pressures and increasing electric fields. The results using ramp input waves show that the rate of compression, in addition to the final stress, can strongly influence the depoling rate. In order to develop a predictive capability for the shock response of PZT 95/5, a description for depoling kinetics must account for these dependencies. This is under active investigation (14). The second aspect is the apparent decoupling at higher stresses between shock-driven depoling and the pore collapse process that occurs in the material at these stresses. The threshold stress for pore collapse in our samples is 2.2 GPa, and a wave structure with this amplitude persists regardless of the final state. Under short-circuit conditions, the depoling kinetics at this stress must be fast compared to the time scale of the pore collapse process. The presence of a strong electric field could reduce this difference. In addition, recent studies have shown that the threshold stress for the onset of pore collapse is very sensitive to the initial material density (15). For shock stresses above the threshold in materials having a lower density, the depoling will be driven by a weaker wave and the decoupling may not be complete.

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