Title

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Journal

Applied Physics Letters, 79(17)

ISSN

0003-6951

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Publication Date

2001-10-22

DOI

10.1063/1.1412595

Peer reviewed

Ultrasonic characterization of poling in lead zirconate titanate ceramics

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(Received 18 April 2001; accepted for publication 13 August 2001)

A method for measuring the spatial variation of the macroscopic poling state of a piezoelectric material using an ultrasonic transducer is described. The relatively simple method clearly indicates the distribution of poling within the material, demonstrated by the testing of a collection of partially poled lead zirconate titanate samples. © *2001 American Institute of Physics.*
[DOI: 10.1063/1.1412595]

Piezoelectric materials, especially lead zirconium titanate (PZT), have found widespread use as electromechanical transducers since their discovery in the late 1800s. For most applications, the piezoelectric materials are polycrystalline and must be *poled*, which reorients the unit domains within the structure more or less in the same general direction as an applied electric field or applied mechanical strain, the former being far more common. However, the extent that the PZT may be poled is limited by many factors, including the crystal structure, composition, and grain size. These factors influence the formation of domains, and the domain wall mobility. Further, in an actual application, the temperature, applied electric field, and time used in poling the piezoelectric material dramatically influence the quality of the poling of the final product. Unfortunately, the ability of the PZT to transduce energy between electrical and mechanical forms is directly dependent on these factors.

What is the quality of poling in a particular material, and what is the distribution of the poling within a sample for a given composition, applied electric field, and electrode configuration? A common method by which to answer this question is to measure the *average* poled state of a material by using electrodes deposited on the surface. This measurement is simple to make, especially when the electrodes are originally used to pole the material, and commercially available d_{33} meters perform such measurements. But measuring the *distribution* of poling across an electroded surface is difficult to perform in this way. In this letter we illustrate a method that uses ultrasonic transducers to send a train of pulses at several positions through a PZT sample to measure the distribution of the speed of sound within it, which is directly related to its poling at that point, and thus demonstrates that the method may be effective to determine both the poling quality and distribution.

Acoustic velocity measurements can provide a nondestructive means by which to determine the quality of poling in piezoelectric materials because of the relationship between a material's acoustic and elastic properties. An example of this is shown in the equation for longitudinal velocity in an isotropic material:

$$v_3 = \sqrt{\frac{E}{\rho}\left[\frac{1-\nu}{(1+\nu)(1-2\nu)}\right]} = \sqrt{\frac{c_{33}}{\rho}}, \tag{1}$$

where v_3 is the longitudinal velocity of the acoustic wave in the third direction (m/s), E is the elastic modulus of the material carrying the acoustic wave in the longitudinal direction (N/m^2), ρ is the material's density (kg/m^3), c_{33} is the stiffness of the material in the third direction (N/m^2), and ν is Poisson's ratio. Longitudinal sound velocity measurements taken in the transverse direction of poling in piezoelectric ceramics produce a higher value compared to the unpoled condition, and corresponds to the expected increase in the modulus reported by Jen *et al.*[1] Although not pursued in this study, measurements taken in orthogonal directions should show a corresponding decrease in value. Shear wave data can also provide similar information related to changes in shear modulus, as well as measurement of the Rayleigh wave velocity as reported by Jen *et al.*[2,3]

In this study, an immersion pulse–echo technique was used to perform measurements on circular PZT specimens which were poled over one half their surface area in the transverse direction. This was accomplished by sputtering approximately 2 μm of Au over the entire surface of one side of each specimen and over only one half of the opposing surface. The effect of the difference in thickness of the sample due to the 2 μm of gold being on only one half of the sample is negligible, amounting to a difference of less than 1 ns. An electric field of 24 kV/cm was then applied to pole the sample, which was then machined into a disk 1.6 cm in diameter and 0.04 cm in thickness. The electrode material was removed from one side of the specimen prior to physical or acoustic measurements. The thickness of each sample was then measured at eight locations using a micrometer.

A scanning acoustic microscope (SAM) was used for data acquisition, imaging, and analysis. The SAM was set up to produce a 256×256 pixel image of the specimen, providing a spatial resolution of 0.07 mm for each pixel. The water

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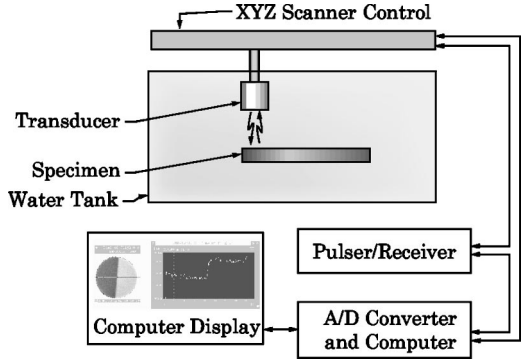

FIG. 1. Experimental setup to detect poling in piezoelectric samples.

path was adjusted initially to provide maximum intensity from the back surface reflection. The system components included a three-axis scanner, pulser, 250 MHz bandwidth receiver, analog-to-digital converter, and software to control the scan setup, pulser/receiver, and image display functions. A block diagram of the arrangement is shown in Fig. 1. The oscilloscope provided a time resolution of 1 ns. The image display capabilities included images of peak amplitude and time-of-flight (TOF) data, signal response amplitude data with respect to time at a specific scan position, and TOF data along cross sections of the sample.

Two transducers were used, 50 and 100 MHz broadbanded probes, both with a focal length of 12.7 mm in water using a fused quartz delay. The focal spot size was approximately 0.1 mm for both probes, and the nominal frequency of the back surface reflection was about 35 MHz. The two probes were used as a crude method by which determine whether the PZT samples were dispersive. Although methods to accurately measure dispersion are much more involved, the output frequency spectrum of each of the two probes is vastly different from the other. Thus, making the same measurement with the two probes would have a noticeable effect on the results if the PZT were indeed dispersive in this frequency range.

Longitudinal velocity measurements were made on the open-circuited sample in the d_{33} direction, and images of peak amplitude and TOF data were generated. After an image of the sample was acquired, two TOF data points were taken at each location, one being the time from the first to the second back reflection, and the other being that from the second to the third. For each measurement the transducer was focused to produce nearly equivalent amplitudes from each of the back reflections being measured. To accomplish this, the water path was adjusted so that the two back surface multiples being measured were approximately equal in amplitude and shape. In the experiment, they indicate the magnitudes of those values along a diameter of the sample roughly oriented in a direction perpendicular to the poled/unpoled interface.

Fuji Ceramics' C-213 hard PZT was used in the construction of the samples for this study. Several measurements were made along the surface of each sample. The material property data that are relevant to this analysis were provided by Fuji Ceramics.[4] Ultrasound measurements to determine the time of flight on each sample were made at eight specific

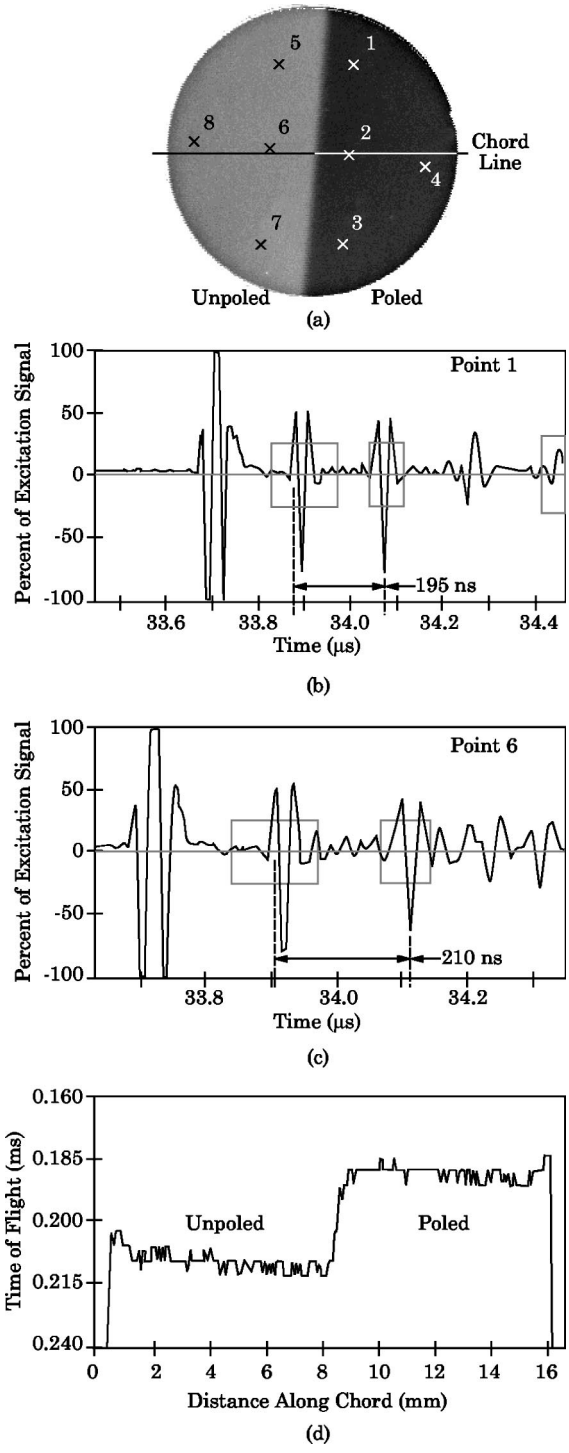

FIG. 2. Example results of the experiment on a specimen with (a) a scan of the entire specimen indicating locations and the chord line where additional data were taken, (b) amplitude and TOF ($=195$ ns) data at point 1, (c) amplitude and TOF ($=210$ ns) data at point 6, and (d) the TOF along a chord line across the sample.

locations at almost the same physical locations on each sample.

The distribution of the speed of sound across the sample is provided in Fig. 2 as a gray scale image [Fig. 2(a)], with the lighter gray on the left indicating a slower speed of sound in the unpoled portion of the sample. Note the transition from the unpoled section to the darker poled section on the right. The width of the transition zone is approximately the same as the thickness of the sample. The data at points 2, 3,

TABLE I. Longitudinal velocities in the Fuji PZT samples: Raw data.

Location No.	Average TOF (μs)	Thickness (mm)	Sound Velocity (m/s)
Sample 1			
1	0.5232	225	4650
2	0.5181	225	4606
3	0.5181	224	4626
4	0.5181	226	4584
5	0.5156	244	4226
6	0.5156	244.5	4218
7	0.5156	243	4244
8	0.5156	243	4244
Sample 2			
1	0.5207	224	4649
2	0.5258	224.5	4684
3	0.5207	225	4628
4	0.5181	225.5	4595
5	0.5156	245	4208
6	0.5156	244.5	4217
7	0.5156	244	4226
8	0.5131	244.5	4197

and 4 were almost identical to the data at point 1, and the data at points 5, 7, and 8 matched the data at point 6 for the two samples. As indicated from the response amplitude plots in Figs. 2(b) and 2(c), the time of flight at points 1 and 6 was 195 and 210 ns.

The unpoled and poled regions are clear in the chord-line TOF plot in Fig. 2(d), even though the TOF changes only 20 ns.

Table I lists the TOF data measured on two different samples along with the calculated sound velocities. The ultrasonic measurement permits the determination of two TOF values based on the first, second, and third reflections of the ultrasound pulse. In all the measurements made on the samples, the two TOF values were virtually identical. The averages of these TOF values are provided in Table I; they were used in conjunction with the measured thickness to compute the sound velocity at each location on the sample.

TABLE II. Mean longitudinal velocities in the Fuji PZT samples.

	Mean longitudinal velocity (m/s)	Standard deviation (m/s)
Sample 1		
Unpoled	4233	11
Poled	4616	24
Change	383	···
Sample 2		
Unpoled	4212	11
Poled	4639	32
Change	427	···

Note in Fig. 2 the location of each of the measurements on the samples' surfaces. Table II provides the mean, standard deviation, and percent of change of the longitudinal velocity of the unpoled and poled portions of the specimen.

A method for experimentally determining the distribution of poling within piezoelectric materials has been presented. Even if the material properties of the poled piezoceramic are unknown, the measurement technique can indicate the distribution of the poling within samples of the material. Improvement of electroding and poling processes can then be more easily performed, and the detection of poling and material defects can quickly be found. Samples made from Fuji Ceramics' C-213 were used as an example of the technique, and the distribution of poling was measured within the samples, illustrating the technique's usefulness.

Support of this work by AlliedSignal Aerospace, Kansas City Division, under Grant Nos. 052G604254 and 052G604904 is gratefully appreciated.

[1] C. K. Jen, C. J. Chung, G. Shapiro, J. Monchalin, P. Langlois, and J. F. Bussiere, J. Am. Ceram. Soc. **70**, C256 (1987).
[2] C. K. Jen, P. Cielo, X. Maldague, and K. Elassal, J. Am. Ceram. Soc. **68**, C146 (1985).
[3] C. K. Jen, G. Shapiro, P. Cielo, and J. F. Bussiere, Rev. Prog. Quant. Nondestr. Eval. **5A**, 625 (1985).
[4] *Piezoelectric Ceramics* (Fuji Ceramics Corporation, Tokyo, Japan, 1998).