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R. W. Smith

July 1970

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by

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July 1970

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ABSTRACT

Lithium niobate can be used to significantly extend the operating temperatures of ultrasonic transducers. Since some high-temperature transducer applications involve exposure to nuclear radiation, it is important to determine the effect of this radiation on the piezoelectric propertiesof the transducer crystal. In this work we measured the effect of gamma radiation on the piezoelectric properties of lithium niobate crystals. We exposed x-cut and z-cut lithium niobate transducer plates to gamma radiation from a ^{60}Co source. The electromechanical coupling factor, the dielectric constant and the frequency constant were measured before exposure and at periodic intervals up to 10^{10}R, total dose. We could detect no significant change in any of these parameters up to 10^{10}R. This indicates that lithium niobate is well suited for applications involving exposure to high levels of gamma radiation.

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EFFECTS OF GAMMA RADIATION ON THE PIEZOELECTRIC PROPERTIES OF LITHIUM NIOBATE

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INTRODUCTION

Lithium niobate, a relatively new ferroelectric crystal, is an important material because it can be used to significantly extend the operating temperature of ultrasonic transducers. Due to its high curie temperature (1210 °C) and the high piezoelectric coupling factors associated with some cuts, improved high temperature ultrasonic transducers are possible. Since some high-temperature transducer applications involve exposure to nuclear radiation (e.g., under-sodium viewing and acoustic emission monitoring), it is important to determine the effect of this radiation on the piezoelectric properties of the transducer crystal.

In this work we measured the effect of gamma radiation on the piezoelectric properties of lithium niobate crystals. The effect of gamma exposure was determined by irradiating some crystals with a gamma source and measuring the piezoelectric electromechanical coupling factor, the dielectric constant, and the frequency constant at progressively higher levels of ^{60}Co radiation. This report outlines the experimental procedure and presents our results of the irradiation effects.

SUMMARY AND CONCLUSIONS

We exposed x-cut and z-cut lithium niobate transducer plates to gamma radiation from a ^{60}Co source to determine the effect of this radiation on the piezoelectric properties of the material. The electromechanical coupling factor, the dielectric constant and the frequency constant were measured before exposure and at periodic intervals up to 10^{10}R, total dose.

We could detect no significant change in any of these parameters up to and including the maximum exposure of 10^{10}R. This result indicates that lithium niobate is well suited for applications involving exposure to high levels of gamma radiation.

DISCUSSION

The piezoelectric coupling factor is a direct measure of the strength of the piezoelectric activity of a crystal. The piezoelectric coupling factor can be defined as the square root of the ratio of the electrical energy available from a crystal to the total mechanical energy input. An alternative definition is the square root of the ratio of the mechanical energy available to the total input electrical energy. The coupling factor for a particular mode of vibration is thus a measure of the effectiveness of the crystal as an electromechanical energy converter for that particular mode.

Both x-cut (shear) and z-cut (longitudinal) lithium niobate are of interest for ultrasonic transducer applications, because both of these cuts have fairly pure vibrational modes.[1] The z-cut plate is a pure extensional mode. An x-cut plate has two shear modes; however, the coupling to one of these modes is very weak while the other is quite strong. The coupling factors for these two cuts can be readily measured to indicate the strength of the piezoelectric activity. Since these cuts are important for transducer applications and their coupling factors can be easily measured, both x and z-cut plates were irradiated during the experiment. The coupling factors were measured at progressively higher levels of radiation from a ^{60}Co source to detect any decline in the piezoelectric activity.

The coupling factor can be measured in several ways.[2-9] The technique recommended in the IRE Standards on Piezoelectric Crystals, 1957 and 1958 involves measuring the series resonant frequency f_s and the parallel resonant frequency f_p for one particular vibrational mode of the crystal. The effective coupling factor k for that mode is then calculated from

$$k = \left[\frac{f_p^2 - f_s^2}{f_s^2}\right]^{1/2}$$

In practice, the parallel resonance frequency may be obscured by unwanted resonances, making the parallel resonant frequency difficult to determine.

A method for determining the coupling factor which does not involve measuring the parallel resonant frequencies was investigated by Onoe et al.[2] and used by Warner et al.[1] to measure the crystal parameters of lithium niobate. This techniaue makes use of the fact that the overtone frequencies of a thickness mode piezoelectric resonator having a high coupling factor are not integral multiples of the fundamental, but instead are shifted toward the higher frequencies. For a pure mode these frequencies can be calculated from

$$\tan \frac{\omega T}{2V} = \frac{\omega T}{2V} \cdot \frac{1}{K^2}$$

where ω is the angular frequency, T is the thickness, and V is the phase velocity of the wave. This equation is exact for a z-cut lithium niobate plate and is a good approximation for an x-cut plate. Onoe has tabulated the coupling factors for a thickness mode resonator as a function of the ratios of the overtone frequencies (up to the third overtone) to the fundamental resonance. Measurement of several overtone freouencies yields several values for k and provides a check on the consistency of the results.

The dielectric constant and the frequency constant were also determined for each plate at each level of radiation. These parameters are sensitive to changes in the crystal and are thus good indicators of damage. The frequency constant is the product of the fundamental series resonant frequency f_s and the controlling dimension (thickness in this case) of the plate.

Determination of the dielectric constant at high and low frequency was made from capacitance measurements. The "free" or static dielectric constant $\varepsilon^T/\varepsilon_0$ was measured at a frequency below the fundamental series resonance (500 kHz for all the plates). Measurement of the "clamped" dielectric constant $\varepsilon^S/\varepsilon_0$ was made at high freauency (— 100 MHz), away from any overtone resonance. At this high frequency the crystal is self-clamped.

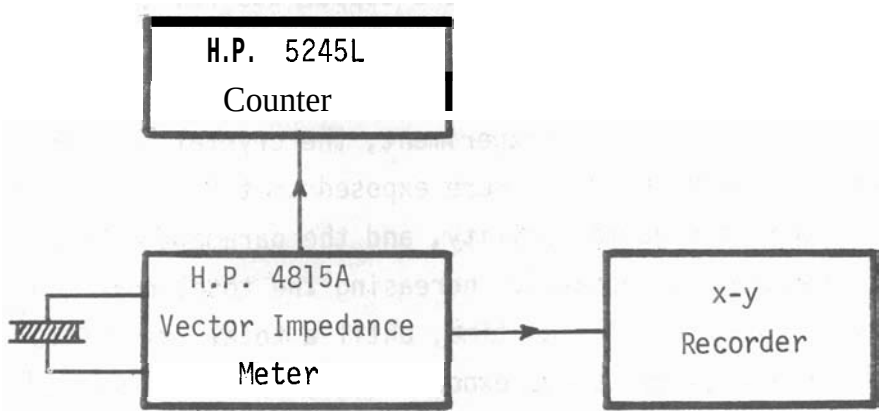
EXPERIMENTAL

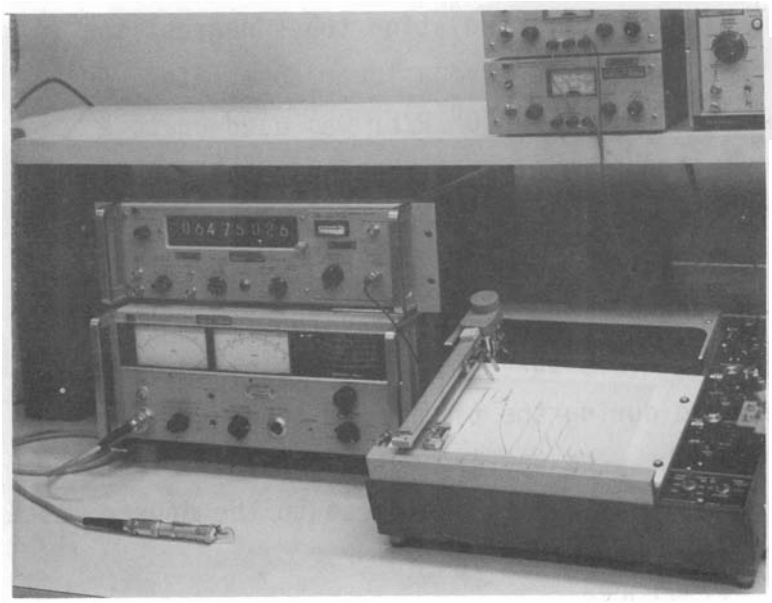
Measurement of the crystal parameters of the lithium niobate transducer plates used in this experiment were made with the instrumentation shown in Figure 1. Accurate frequency measurements were made with a 200 MHz electronic counter, Hewlett Packard Model 5245L. The impedance measurements were made with a Hewlett Packard Model 4815A RF Vector Impedance Meter. This instrument has its own oscillator and reads out both the magnitude of the impedance and the phase angle. Analog recorder outputs are provided for the impedance magnitude and phase angle as well as the frequency. The impedance magnitude was recorded as a function of frequency using an X-Y recorder. After the resonant frequencies were located approximately, the scale on the recorder was expanded and a more detailed graph of the impedance characteristics in the area of the resonance was taken.

A total of four plates were used, two x-cuts and two z-cuts. The geometry of the plates is summarized in Table 1. Two thicknesses of each cut were used, one approximately 1 mm thick and one about 0.2 mm thick. The plates were cut from a single domain optical quality lithium niobate crystal. The faces were lapped and polished flat and parallel within 0.5 μ (micron).

Table 1. Initial Measurements on the Unirradiated Crystals

Crystal	Coupling Factor		Dielectric Constant $\frac{\varepsilon^T}{\varepsilon_o}$		Frequency Constant Hz-meters	
	meas.	publ.	meas.	publ.	meas.	publ.
x-cut Area - 1.15 cm^2 Thickness - 0.518 mm	0.68	0.68	85	84	1820	1838
x-cut Area - 1.39 cm^2 Thickness - 0.254 mm	0.69	0.68	81	84	1840	1838
z-cut Area - 2.41 cm^2 Thickness - 0.254 mm	0.21	0.17	28	30	3650	3615
z-cut Area - 2.65 cm^2 Thickness - 0.991 mm	0.23	0.17	29	30	3600	3615





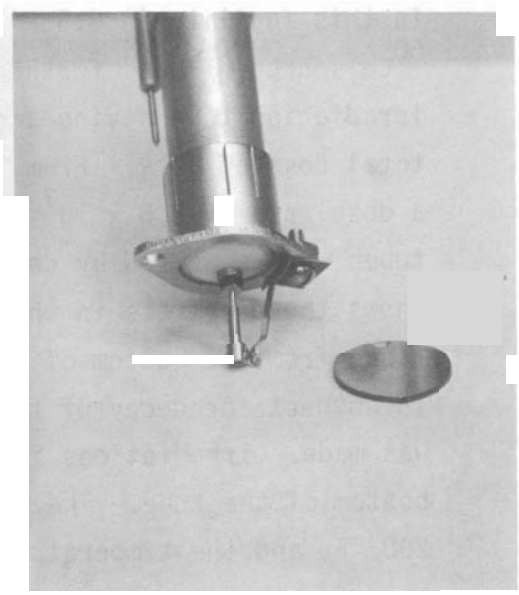


Figure 1. Measurement System

Initially, copper electrodes about 0.5 μ thick were deposited on the crystals. These electrodes were oxidized during the experiment in the gamma facility. Consequently, the copper was removed with a dilute nitric acid solution and chrome gold electrodes were redeposited. The thicknesses of chrome and gold were about 200 and 2000 Å, respectively.

In the course of the experiment, the crystal parameters were first measured, then the crystals were exposed to the desired level (10^5R initially) in the gamma facility, and the parameters were measured again. This procedure was repeated, increasing the total dose to an order of magnitude higher level each time, until a total dose of 10^{10}R was attained. One of the x-cut plates was exposed ahead of the others to detect any change and to make it possible to adjust the radiation level of the remaining plates, if necessary.

The transducer plates were irradiated in the Hanford Laboratory Gamma Radiation Facility (Figure 2). The highest radiation dose rate available in this facility is 3.5 x 10^7 R/hr in the irradiation tubes nearest the ^{60}Co source. Tubes further from the source have a lower dose rate. An irradiation tube having a dose rate of 1.5 x 10^6 R/hr was used up to a total dose of 10^7R. From 10^7 to 10^{10}R total dose the inner tube, having a dose rate of 3.5 x 10^7 R/hr, was used. Dose rates in the irradiation tubes were measured by ceric sulfate dosimetry techniques. Figure 3 shows the dose rate in the tube nearest the source as a function of distance from the bottom of the tube. This curve must be multiplied by 0.92 to correct for decay of the source during the time since the measurement was made. Irradiations in this tube were carried out about 7 in. from the bottom of the tube. The temperature in the tube closest to the source was 208 °F, and the temperature in the outer tube was 98 °F.

RESULTS

Our measurements on the unirradiated crystals show good agreement, in most cases, with published values. These measurements are summarized in Table 1 which also shows the published values[1] for the parameters. We measured a somewhat higher value for the coupling factor for z-cut plates than Warner[1]. This is the only significant difference.

Figure 2. Hanford Laboratory Gamma Radiation Facility

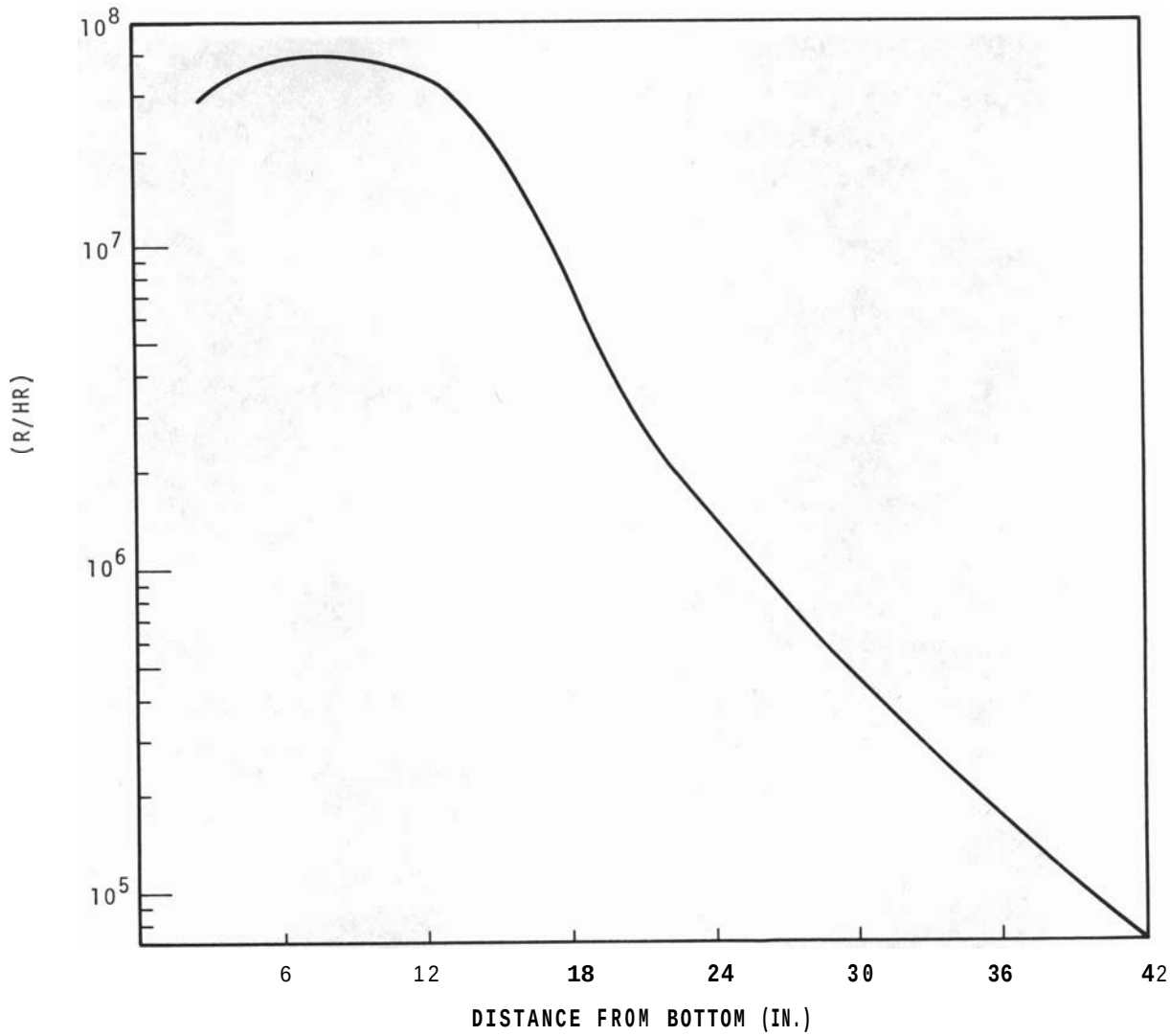


Figure 3. Gamma Irradiation Facility

We could detect no significant change in any of the parameters for radiation exposure levels up to 10^{10}R. These measurements are summarized in Tables 2 through 5. The deviations in the tables are all within the measurement errors. These data indicate that the crystal is essentially unaffected, as evidenced by the essentially constant values for the parameters up to 10^{10}R.

Table 2. Radiation Test of Lithium Niobate

Crystal: z-cut
Area 2.65 cm^2 Thickness 0.991 mm

Total radiation dose, rads	Dielectric constant $\frac{\varepsilon^S_{33}}{\varepsilon_o}$	$\frac{\varepsilon^T_{33}}{\varepsilon_o}$	Coupling Factor Calculated from the resonance ratios $\frac{f_2}{f_1}$	$\frac{f_3}{f_1}$	$\frac{f_4}{f_1}$	average	Frequency constant, Hz-meters
0	28	29	0.232	0.225	0.223	0.227	3599
1.5×10^5	28	29	0.231	0.226	0.223	0.227	3599
10^6	28	29	0.233	0.236	0.234	0.234	3597
10^7	28	28	0.234	0.226	0.224	0.228	3597
10^8	28	29	0.238	0.231	0.229	0.233	3594
10^9	28	29	0.239	0.231	0.229	0.233	3593
10^{10}	28	29	0.239	0.232	0.231	0.234	3593

Table 3. Radiation Test of Lithium Niobate

Crystal: z-cut

Area 241 cm^2 Thickness 0.254 mm

Total radiation dose, rads	Dielectric constant $\frac{\varepsilon^S_{33}}{E_o}$	Dielectric constant $\frac{\varepsilon^T_{33}}{E_o}$	Coupling Factor calculated from the resonance ratios $\frac{f_2}{f_1}$	$\frac{f_3}{f_1}$	$\frac{f_4}{f_1}$	average	Frequency constant, Hz-meters
0	27	28	0.213	- -	- -	0.213	3652
1.5×10^5	27	28	0.213	0.212	- -	0.212	3652
10^6	27	28	0.217	0.211	- -	0.214	3640
10^7	26	28	0.221	0.214	- -	0.217	3638
10^8	27	28	0.217	0.214	- -	0.216	3638
10^9	27	28	0.221	0.214	- -	0.217	3638
10^{10}	27	28	0.221	0.214	- -	0.217	3638

Table 4. Radiation Test of Lithium Niobate

Crystal: x-cut
Area 1.15 cm^2 Thickness 0.518 mm

Total radiation dose, rads	Dielectric constant $\frac{\varepsilon_{11}^S}{\varepsilon_o}$	Dielectric constant $\frac{\varepsilon_{11}^T}{\varepsilon_o}$	Coupling factor Calculated from the resonance ratios $\frac{f_2}{f_1}$	$\frac{f_3}{f_1}$	$\frac{f_4}{f_1}$	average	Frequency constant, Hz-meters
0	49	85	0.685	0.684	0.684	0.684	1821
1.5×10^5	45	85	0.685	0.684	0.684	0.684	1821
10^6	46	84	0.685	0.684	0.684	0.684	1821
10^7	46	84	0.685	0.685	0.685	0.685	1820
10^8	45	83	0.687	0.685	0.685	0.686	1820
10^9	45	84	0.687	0.685	0.685	0.686	1820
1.1×10^{10}	45	84	0.686	0.685	- - -	0.686	1820

Table 5. Radiation Test of Lithium Niobate

Crystal: x-cut

Area 1.39 cm^2 Thickness 0.254 mm

Total radiation dose, rads	Dielectric constant $\frac{\varepsilon_{11}^S}{\varepsilon_0}$	Dielectric constant $\frac{\varepsilon_{11}^T}{\varepsilon_0}$	Coupling Factor Calculated from the resonance ratios $\frac{f_2}{f_1}$	$\frac{f_3}{f_1}$	$\frac{f_4}{f_1}$	average	Frequency constant, Hz-meters
0	42	81	0.690	0.683	- -	0.690	1838
1.5×10^5	42	80	0.689	0.685	0.683	0.686	1838
10^6	42	80	0.689	0.685	0.683	0.686	1835
10^7	43	80	0.689	0.685	0.684	0.686	1835
10^8	42	80	0.689	0.686	0.683	0.686	1835
10^9	42	80	0.689	0.686	0.683	0.686	1835
10^{10}	42	80	0.689	0.686	0.683	0.686	1835

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