A DEVICE FOR MEASURING THE STATIC PIEZOELECTRIC EFFECT IN ROCHELLE SALT CRYSTALS

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A DEVICE FOR MEASURING THE STATIC PIEZOELECTRIC EFFECT IN ROCHELLE SALT CRYSTALS

INTRODUCTION

Piezoelectricity may be defined as the electric polarization that is produced by a mechanical strain in certain anisotropic substances. This polarization, known as the direct piezoelectric effect, is proportional to the strain and changes sign with it. In the converse effect, strain is produced by a polarizing field.

Of the thirty-two possible crystal classes, twenty classes have the asymmetry to be piezoelectric. Considering the three tension or compression and the three shear components of strain with the three components of electric polarization along the principle crystallographic axis, it becomes evident that there are eighteen possible independent piezoelectric constants. In general, the higher the degree of crystalline asymmetry, the greater will be the number of piezoelectric constants.

The piezoelectric equations for the direct effect in terms of stress are:

$$-P_x = d_{11}X_x + d_{12}Y_y + d_{13}Z_z + d_{14}Y_z + d_{15}Z_x + d_{16}X_y$$

$$-P_y = d_{21}X_x + d_{22}Y_y + d_{23}Z_z + d_{24}Y_z + d_{25}Z_x + d_{26}X_y$$

$$-P_z = d_{31}X_x + d_{32}Y_y + d_{33}Z_z + d_{34}Y_z + d_{35}Z_x + d_{36}X_y$$

where, P_X is the polarization in the X direction, X_X is a normal stress in the X direction, Y_Z is a shear stress in the Y-Z plane, and the d's are piezoelectric constants.

The equations for the converse effect in terms of strain are:

x_x = d₁₁E_x+ d₂₁E_y + d₃₁E_z y_y = d₁₂E_x+ d₂₂E_y + d₃₂E_z z_z = d₁₃E_x+ d₂₃E_y + d₃₃E_z y_z = d₁₄E_x+ d₂₄E_y + d₃₄E_z z_x = d₁₅E_x+ d₂₅E_y + d₃₅E_z x_y = d₁₆E_x+ d₂₆E_y + d₃₆E_z

where x_X is the normal strain in the X direction, and E_X is the electric field in the X direction.

In the triclinic system, which is of the highest asymmetry, all eighteen piezoelectric constants may differ from zero. In the case of Rochelle salt, which is in the rhombic system, only d14, d25, and d36 differ from zero.

Thus, the piezoelectric equations for Rochelle salt reduce to,

for the direct effect in terms of stress,

$$-P_X = d_{14}Y_Z$$
,

$$-Py = d25Zx$$

$$-P_z = d_{36}X_y$$
,

and for the converse effect in terms of strain,

$$y_z = d_{14}E_x$$
,

$$z_x = d_{25}E_y$$
,

$$x_y = d_{36}E_z$$
.

If one now considers only X-cut Rochelle salt, the piezoelectric equations reduce to only those containing the constant, d₁₄. Thus, in the direct effect, a shear stress in the Y-Z plane will produce polarization in the X-direction only. Also, for the converse effect, an electric field applied in the X-direction will produce only a shear stress in the Y-Z plane.

If the crystal is 45° X-cut Rochelle salt (Figure 1), the shear strain, y_Z , becomes a normal strain, y_Z , and the piezoelectric constant is now $\frac{d_14}{2}$. Thus the basic equation is

$$y_z \cdot = \frac{d_{14}}{2} E_x,$$

where
$$y_z^{\dagger} = \frac{y_z}{2}$$

Rochelle salt possesses piezoelectric and dielectric properties so unique that they are known as ferroelectric properties. This is because of the analogy of these properties to the magnetic properties of ferromagnetic substances. As the Curie point in iron is the critical temperature at which the permeability falls off to unity, so in Rochelle salt there are two critical

temperatures, the upper and lower "Curie points", at which a similar change takes place in the dielectric and piezoelectric constants. These temperatures are 24°C. and -18°C. This change may be observed with fields in the X-direction and shear stresses in the Y-Z plane.

The apparatus described in this paper has been designed for use as a Modern Physics laboratory experiment. A 45° X-cut Rochelle salt crystal, shown in Figure 1, was selected as the crystal to be used because of the relative ease with which the converse piezoelectric effect may be observed. The piezoelectric effect in X-cut Rochelle salt is very much affected by such variables as temperature, stress, and the particular electrode installation. Therefore, one of the most important aspects in the design of this apparatus was to make possible the observation of both the direct and converse effects in one particular crystal installation. Cady asserts that no comprehensive study of both effects has been made on the same crystal specimen (1, pp.545-546).

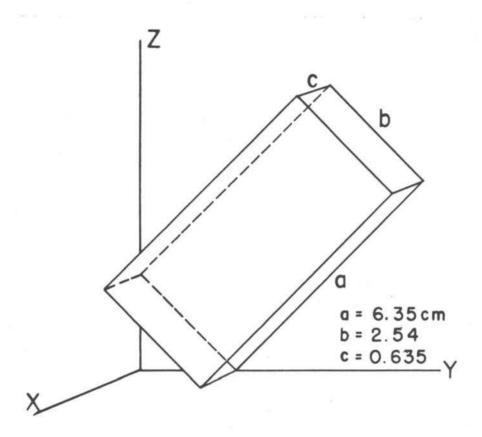


Fig. I. The 45° X-cut Crystal

GENERAL ARRANGEMENT

The temperature is controlled by use of a water jacket. Compression stresses are applied with a spring balance. All strains are measured with an optical system utilizing interference fringes. The polarization which is produced by straining a crystal, as in the direct piezoelectric effect, is measured by means of an electrometer tube circuit. The electric potentials applied to the crystal, to observe the converse effect, are obtained from a regulated direct current power supply.

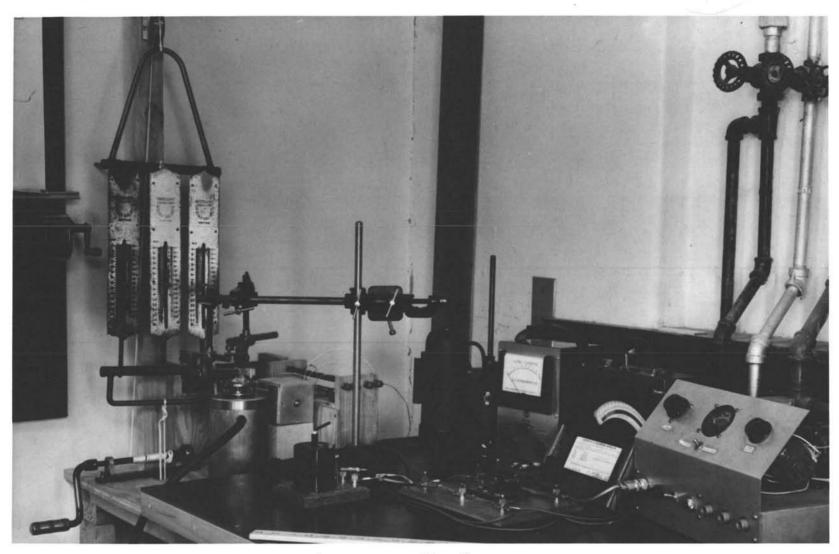


Fig. 2

MECHANICAL SYSTEM

as in Figure 3, which is attached to the rod that protrudes from the bottom of the water jacket. Any vibration of the spring balance is not transmitted to the rest of the apparatus because of the pulleys. The stress is then transmitted from the rod to a spider, through the spider to four rods, and finally to a disc that rests upon the top of the crystal. (Figure 4) The bottom of the crystal is supported by a steel spacer which rests upon the lower disc. Each disc is centered with three ball bearings, one of which is spring loaded.

An invar rod, which has a very low thermal coefficient of expansion, rises from the upper disc to support an optical flat. Two polystyrene tubes, paralleling the invar rod, carry the electrode leads to points outside the water jacket. The optical system is used to measure deformation between the optical flat and the upper part of the water jacket.

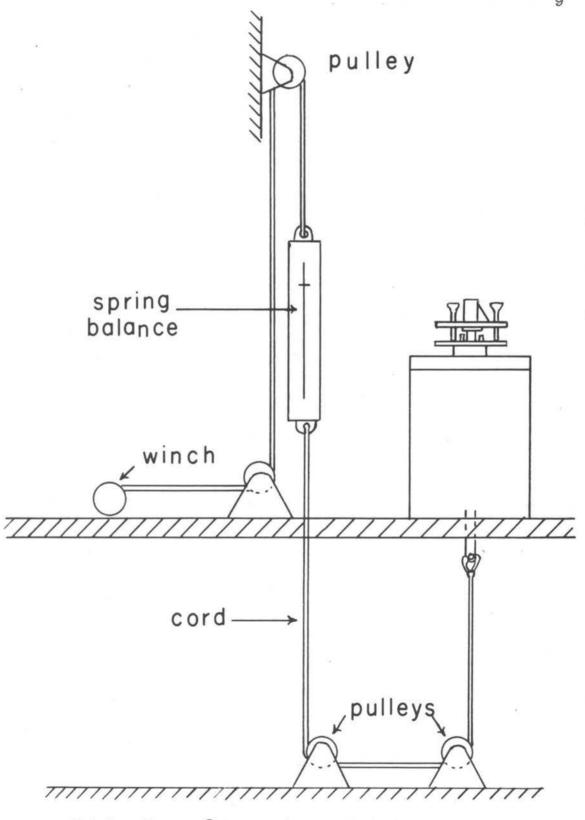


FIG. 3 Stressing Device

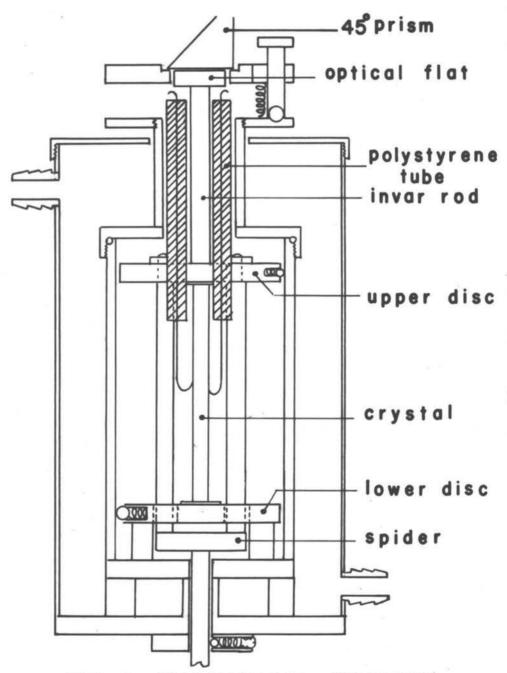


FIG. 4 MECHANICAL SYSTEM

THE OPTICAL SYSTEM

A schematic diagram of the optical system is given in Figure 5. Interference arises from the reflection of monochromatic light from the upper surface of the optical flat and the lower surface of the 45° prism. The prism rests upon a spring loaded disc. The three adjustment screws of the disc are mounted with one in a hole, one in a slot, and the other on a plane. The fringes resulting from the constructive and destructive interference are observed with a microscope. Thus a shift of one fringe indicates a deformation in length of one-half the wavelength of light that is used. The light source used in these observations was a mercury vapor lamp which was fitted with a green filter. The wave-length of this light is 5460A.

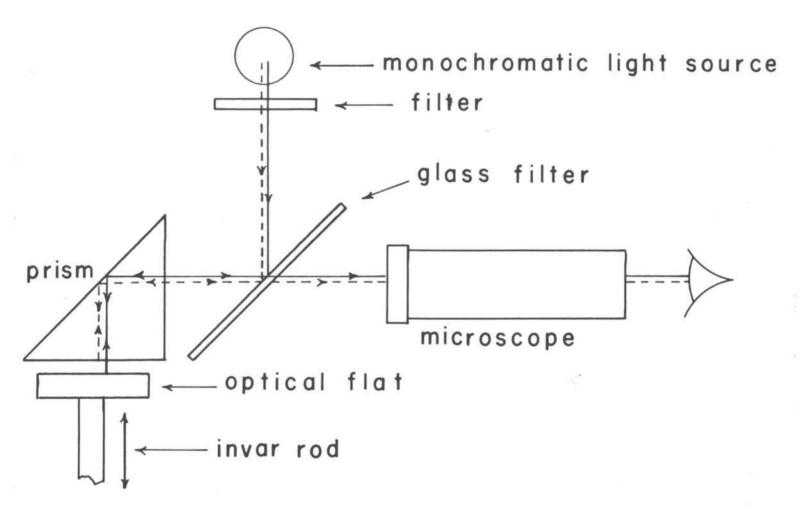


FIG. 5 Optical System

THE ELECTRONIC CIRCUIT

The circuit diagrams are given in Figures 6 and 7. One electrode lead is brought out to the grid of the electrometer tube and also to one side of a 0.25 microfarad capacitor. The other electrode lead is brought out to the other side of the capacitor. A change in the potential difference across the capacitor indicates the amount of charge produced on a crystal face by a corresponding change of stress on the crystal. Also, the grid potential of the electrometer tube controls the current through the plate resistor. Thus, the potential across the plate resistor may be calibrated to give the grid potential.

To apply an electric potential to the crystal, a direct current power supply and a high resistance potentiometer were used, the ground and tap leads being connected to the capacitor.

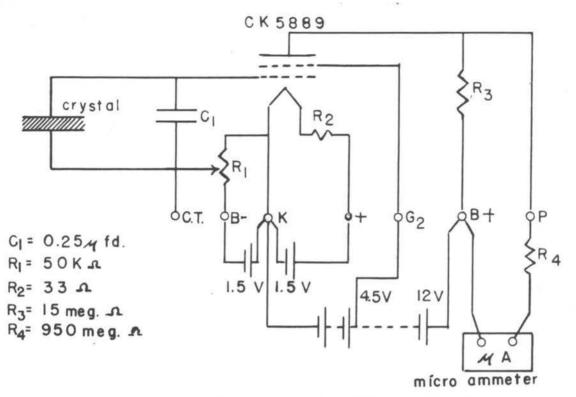


Fig. 6. Direct Piezoelectric Effect Circuit

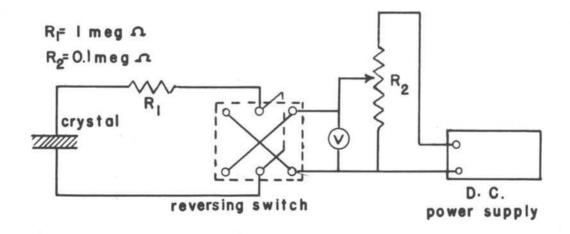


Fig. 7 Converse Piezoelectric Effect Circuit

THE INSTALLATION OF A CRYSTAL

The tin foil electrodes should make as close physical contact to the clean surface of the crystal as is practical. These electrodes should stress the crystal as little as possible. A collodion cement is used to hold the electrode leads in place. Silver paint is used to make good electrical contact.

First, the crystal and the quartz insulator are fixed to the upper disc with Ward's bio-plastic cement. To do this, one may hold the upper disc upside down by clamping the invar rod in a vise. Then the electrode leads are threaded through the polystyrene tubes and the crystal and quartz insulator are cemented in place. Another quartz insulator is then cemented to the bottom of the crystal. Ceresin wax was painted on all surfaces to help prevent surface leakage of the electric charge. Although the bio-plastic cement does not begin to set for a few hours, the Ceresin wax will hold the crystal in place during the rest of the installation.

After the lower disc and spacer are cemented in place, the lower face of the quartz insulator is cemented and the crystal is lowered into place. This may be accomplished by holding the ball bearings in place with the fingers of one hand while holding the invar rod with the other hand. The upper disc may now be bolted to the

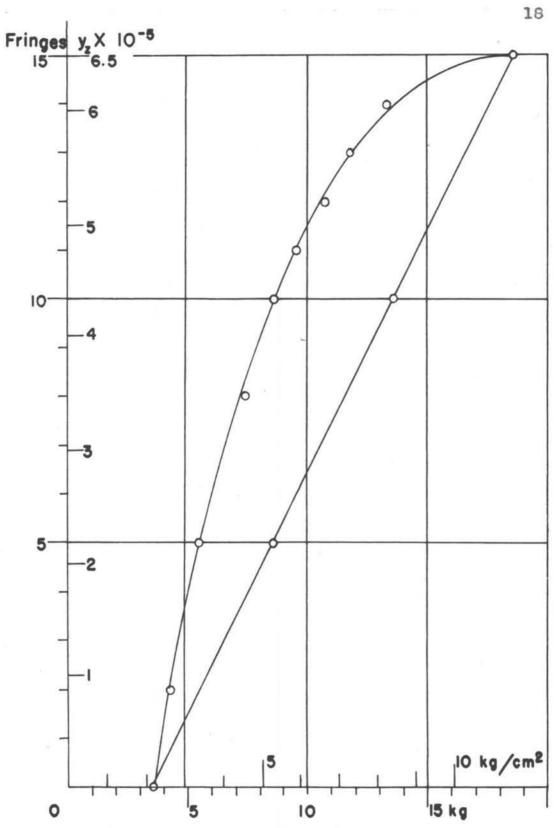
four rods that lead to the spider.

Setting of the bio-plastic cement is accelerated by warming the apparatus for a few hours. It must be remembered, however, that Rochelle salt "melts" into a solution of Na and K tartrates at 55.6°C. This process is irreversible.

MEASUREMENTS

In making any piezoelectric measurements on X-cut Rochelle salt crystals, it must be noted that temperature is a very critical variable. At least two hours are needed for the size crystal used in these experiments, Figure 1, to come to temperature equilibrium.

Elasticity measurements are taken with the electrodes at the same potential, so that the electric field is zero. A stress-strain hysteresis curve was observed, and is shown in Figure 8. Only increasing compression stresses produced a linear relationship. The crystal must be made to produce this hysteresis curve several times before reproducible results are obtained. It is not known if this hysteresis curve is peculiar to the particular crystal installation tested, or is true of all Rochelle salt crystals. Two crystals have been tested and both show this hysteresis effect. All reported values of other workers for the elastic constants were obtained from resonant frequencies of lengthwise vibrations, except those of Hinz (2, pp.617-632), who used a static method involving an optical lever. Hinz does not report an elastic hysteresis effect.



Elasticity

Strain of the apparatus, which would be part of the observed strain of a crystal, was found to be 1.0 x 10^{-6} cm/kg. This was determined by replacing the Rochelle salt crystal with a steel blank. This is negligible compared to the crystal strain.

must first determine the time constant of the crystal and electronic circuit in order to know how much error is introduced by leakage. This was done by determining the resistance between the electrodes by the use of a micro ammeter. This resistance, when multiplied by the capacitor size of 0.25 microfarads, is equal to the time constant. This same micro ammeter was used to indicate the potential drop across the plate resistor in the electrometer tube circuit, as indicated in Figure 6. A known negative potential difference, of from zero to two volts, was applied across the 0.25 micro farad capacitor to calibrate the micro ammeter readings with respect to the capacitor voltage. A typical calibration curve is given in Figure 9.

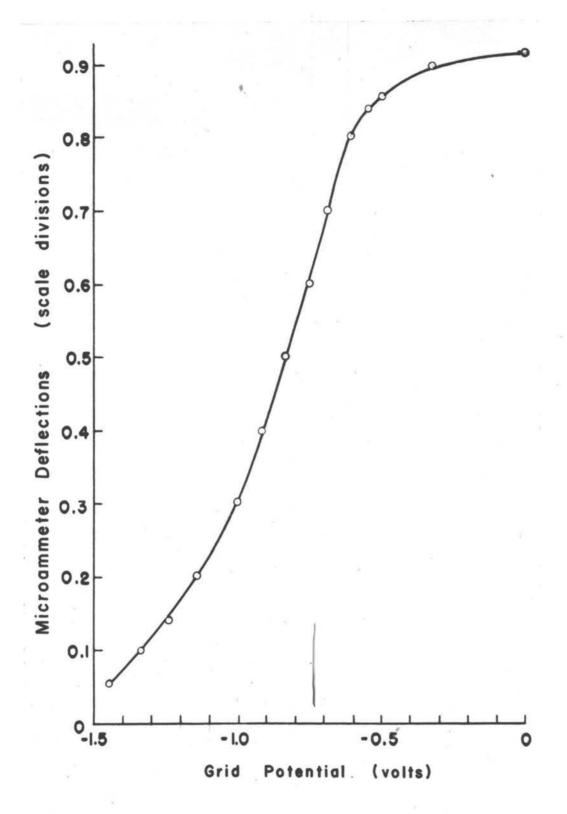


Fig. 9. Typical Calibration Curve of Electrometer Circuit

Before the piezoelectric measurements were taken, the crystal was given a stress and strain history. was done in the same manner in which one would observe a stress-strain hysteresis curve between forces of 3kg and 20kg. The piezoelectric measurements were then taken by increasing the compression force from 6kg to higher values up to llkg. If necessitated by a very short time constant of about five minutes or less, the readings may be taken in an absolute manner. An observer is able to stress the crystal to a predetermined value and read the micro ammeter within twelve seconds. However, when taking absolute measurements, it must be remembered to maintain the elastic history of the crystal. With a longer time constant, continuous readings may be taken. Readings taken by both methods gave essentially the same results. The micro ammeter readings were then transformed by use of the calibration curve into capacitor voltages and then into crystal polarization values.

After stressing the crystal to 6kg/cm², and observing an immediate polarization, an increase in polarization was observed for the next twenty minutes, at which time the electrometer tube current became saturated. Because of a relatively short time constant, it was not possible to measure the effect. This lag in the polarization was reported by Shulvas-Sorokina (6, pp.700-706) for stresses under lkg/cm².

The piezoelectric constant, d₁₄, is represented by the slope of the polarization-stress curves, shown in Figure 10. From these polarization curves, it is evident that d₁₄ is not a constant, but is a variable that depends both upon stress and temperature. Almost all the published data on the polarization-stress relationship may be found in papers by Iseley (3, pp.569-574) and Shulvas-Sorokina (6, pp.700-706). Iseley's curves include only stresses under 2kg/cm². Although Shulvas-Sorokina's polarization-stress curves cover a wide range of stresses, she has failed to state the unit of polarization, which she reports to be different for each curve.

Shulvas-Sorokina's highest value of d₁₄ is about ten times as great as Valasek's (6, pp.235-236). Although Valasek fails to specify the stress, this discrepancy may be accounted for by stresses resulting from the mounting, the depolarizing effect of surface layers between the electrodes and the crystal, and differences in the different crystal specimens. The magnitude of the direct piezoelectric effect presented in this paper was found to fall between the values reported by Valasek and Shulvas-Sorokina.

From the slope of the direct effect curves, constant stress values of d_{14} may be calculated and plotted as functions of temperature. These curves are

shown in Figure 11. The characteristic shape of these curves is indicated in the results of Shulvas-Sorokina and Valasek.

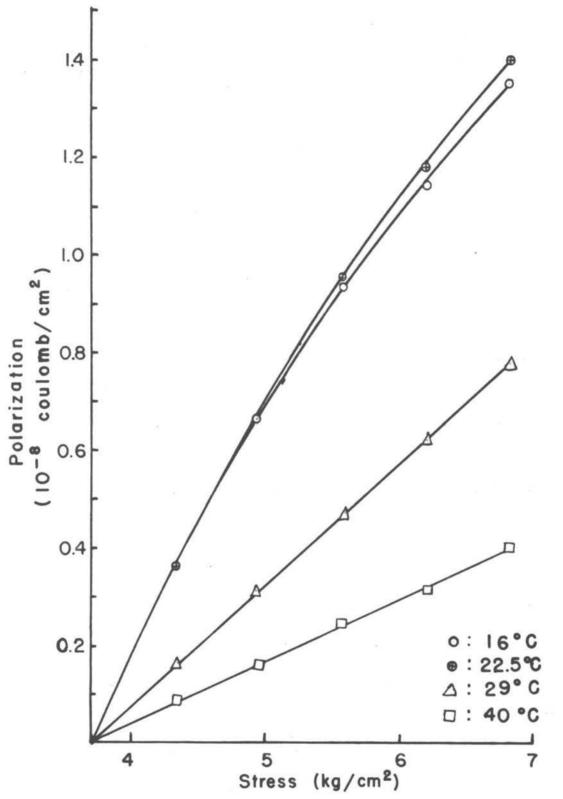


Fig. 10. Direct Effect at Various Temperatures

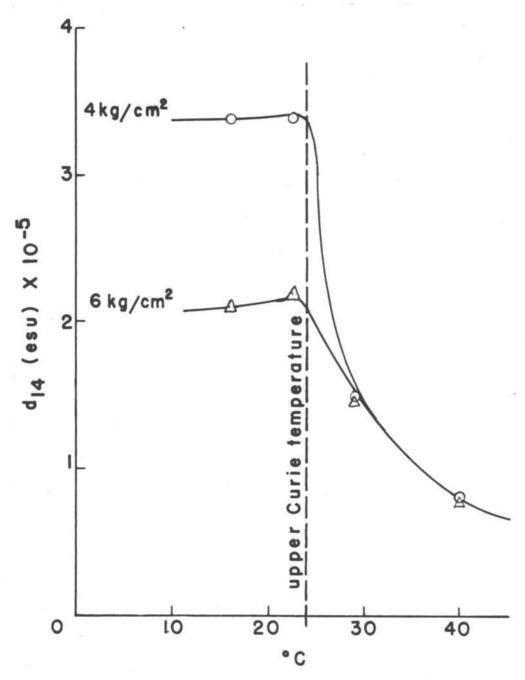


Fig. II. d_{14} from the direct effect as a function of temperature for two different stresses

The converse effect was observed by applying a polarizing field to the crystal, and measuring the resulting deformation by use of the optical system. The applied voltage was obtained from a direct current power supply and a high resistance potentiometer, and was measured with a voltmeter. A reversing switch was used to change the direction of the applied field. To protect the crystal from high charging currents, a megohm resistor was wired in series with the electrode leads. The converse curves obtained, shown in Figure 12, agree with those of Hinz (2, pp.617-632) to within 5%.

The converse effect is also a function of stress, as well as temperature. Thus, additional converse effect curves, shown in Figures 13 and 14, were observed by adding various static compression stresses to the crystal. This was done with the use of the same device that stressed the crystal in obtaining the direct effect observations. These curves clearly show a decrease in the converse piezoelectric effect with increasing stress. The piezoelectric constant d₁₄ was computed from these curves and is shown in Figure 15 as a function of stress.

For all converse effect observations, the electric field range was $\frac{1}{2}$ 236 v/cm. The minimum static stress on the crystal, y_z , was 0.25kg/cm².

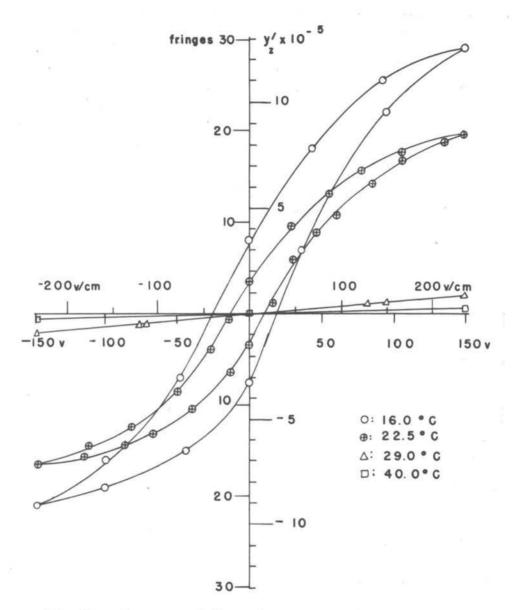


Fig. 12 Converse Effect for Various Temperatures With a Static Stress of O. 25 Kg/cm²

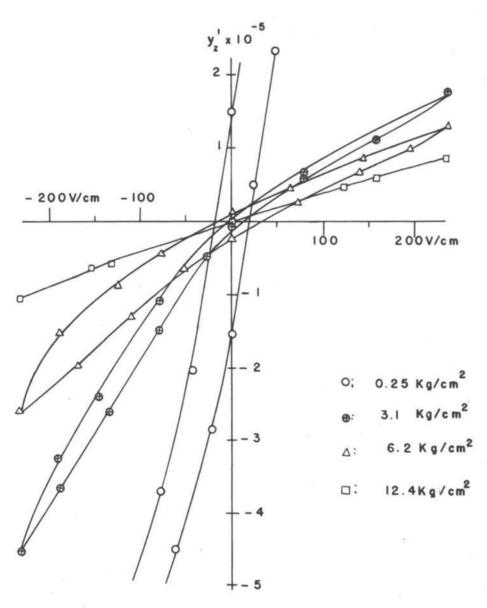


Fig. 13 Converse Effect at 22.5 °C for Various Static Stresses

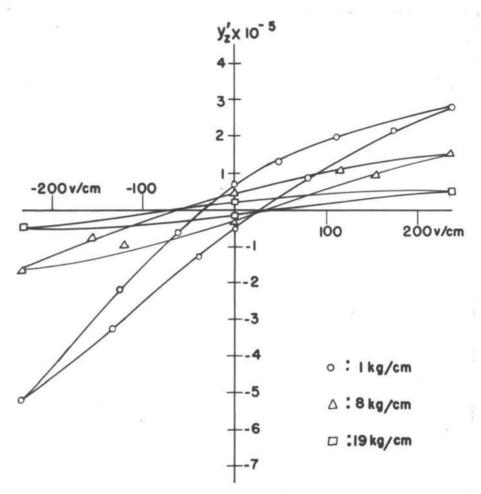


Fig. 14. Converse Effect at 21 °C for Various Static Stresses

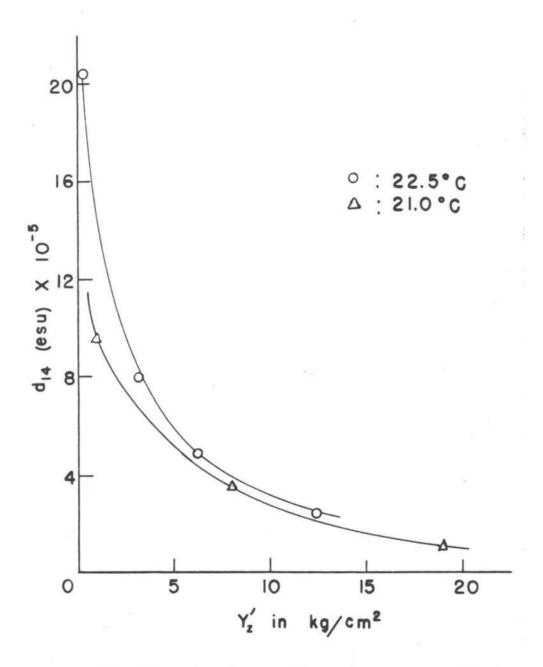


Fig. 15. d₁₄ from the converse effect as a function of stress for two different temperatures

CONCLUSIONS

The Modern Physics laboratory experiment, for which this apparatus was designed and constructed, is of two hours duration. During this time any one of a number of piezoelectric experiments may be performed. Although the apparatus is designed for use at various temperatures, only one such temperature may be used in any one laboratory period. It is suggested that the following experiments may be performed.

Both the direct and converse effects could be observed in two hours. Here, in the converse effect, a static load would be applied which would be of such a value as to fall within the range of the stresses applied in observing the direct effect. Thus, the computed values of the direct and converse d₁₄'s could be compared under equal conditions.

Also, the converse effect may be observed under various static loads. The constant, d_{14} , for the converse effect could then be computed and plotted as a function of stress.

Again, the converse effect may be observed with various maximum electric field strengths, and the constant, d₁₄, computed and plotted as a function of the applied electric field.

In evaluating the data obtained from this apparatus, it should be remembered, as stated in the introduction, that both the direct and converse observations are made on one crystal specimen. Thus, in comparing the two effects, any error due to individual differences in different crystal specimens is eliminated.

The data presented in this paper is in reasonable agreement with the published findings of other investigators. A search of the literature reveals no findings with which to verify the results obtained in the converse effect wherein the crystal is subjected to a constant stress. Still, these results show the same stress anomaly that exists in the direct effect.

A comparison of the converse and direct effects found in this paper show the converse effect to be several times as great as the direct effect. This discrepancy is found throughout the literature in the field. At present it is not known if this is due to an irreversibility of the piezoelectric effect or is caused by consistent errors in the experimentation.

BIBLIOGRAPHY

- Cady, Walter Guyton. Piezoelectricity. New York, McGraw-Hill, 1946. 806p.
- Hinz, H. Elastische Deformationen am Seignettesalz. Zeitschrift für Physik 111:617-632. 1939.
- 3. Iseley, F. C. The relation between the mechanical and piezoelectrical properties of a Rochelle salt crystal. Physical review 24:569-574. 1924.
- 4. Mason, Warren P. Piezoelectric crystals and their application to ultrasonics. New York, D. Van Nostrand, 1950. 508p.
- 5. Shulvas-Sorokina, R. D. Piezoelektrische Eigenschaften von Seignettesalzkristallen. Zeitschrift fur Physik 73:700-706. 1932.
- 6. Valasek, J. Note on the piezoelectric effect in Rochelle salt crystals. Science 65:235-236.