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TRANSITIONAL CHARGE RELEASE

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Technical Report 3672

TRANSITIONAL CHARGE RELEASE

by

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May 1968

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ABSTRACT

The pressure-enforced ferroelectric-to-antiferroelectric phase transition in the system $PbZrO_3$-$PbTiO_3$-$PbSnO_3$-$PbNb_2O_6$ was investigated as a charge-release mechanism for fuzing. Capacitance, dielectric loss, thermal expansion, and hydrostatic transition pressure were measured over the Mil Spec temperature range.

This effort was conducted under the Independent Research Program, AMCMS Code 5016.11.844, of the Director, Feltman Research Laboratories.

INTRODUCTION

Many fuzing systems require a setback-actuated power source to produce a uniform output over a wide range of accelerations. In practice, a piezoelectric element is often used to generate the voltage, and mechanical shorting beams or Zener diodes are used to regulate the voltage to a constant value.

The use of a material which undergoes a pressure-enforced ferroelectric-to-antiferroelectric phase transition as a power source should result in a much less complex power source design. These materials are completely depolarized when they become antiferroelectric. Since the entire remanent polarization is released, the output is dependent only on the original polarization of the element rather than exhibiting a pressure dependence as in a conventional element. The use of an element of this sort in setback fuzes could eliminate rectifying and voltage regulating diodes and mechanical shorting beams as well as reduce the weight of the proof mass. Also, such an element employed in a hydrostatic vessel could be used as an improved impact power source of a sensitivity which, from the standpoint of force or shock requirements, would be greater than that of the standard piezoelectric element; besides, it would allow a multi-directional (graze) capability.

A more extensive explanation of the difference between these materials and conventional ferroelectrics may be found in Picatinny Arsenal Technical Report 3481, which includes the results for the first year of this project.

A range of compositions which appear to meet the requirements for a power source of this type has been selected. Minor compositional changes can effectively control temperature dependency in this material. Thus, once the effect of temperature on the various compositional modifications of this material has been determined, it will be possible to use the material with confidence over the entire temperature range relevant to military application.

PROCEDURE

Capacitance and dissipation factor were measured as a function of temperature with a General Radio Type 1610 capacitance measuring assembly. The specimens were immersed in silicon oil, and data were plotted every 5° from 0° to 200°C on both heating and cooling.

Thermal expansion measurements were made with a fused silica dilatometer designed for use with specimens approximately 1 mm in diameter by 1 cm long. A "Daytronic" differential transformer displacement measuring system was used in conjunction with an X-Y recorder and a thermocouple to produce an automatic plot of temperature vs expansion. The furnace was cooled to approximately -70°C before each run, and thermal expansion data were recorded from -50° to +200°C on heating, and from 200°C to room temperature on cooling. The furnace, was programmed to cycle in approximately six hours, by means of a motor-driven variac, and a limit controller was employed to initiate the cooling portion of the cycle.

Hysteresis loop measurements were made at a frequency of 1/2 hz with a modified Sawyer-Tower type circuit. The previous report included hysteresis loop data over the temperature range relevant to military application. This report includes the measurement of loop data as a function of axial loading, and hydrostatic pressure. Silicon rubber "RTV" potting compound was used as a "fluid" in the hydrostatic vessel shown in Figure 1. Copper wires were attached with low-temperature solder to the silvered electrodes of the specimen. The ends of the 10-mil copper wire made mechanical contact with the steel pistons which were used to apply pressure. This vessel worked well up to 50,000 psi and was also used to measure the effect of temperature on the hydrostatic transition pressure. A second hydrostatic vessel was constructed as shown in Figure 2. This system also appeared to closely approximate hydrostatic conditions. A thermocouple was welded to the brass electrode below the specimen, so that temperature could be measured. Specimens could be replaced by removing the top piston. Since the second system was more convenient to use, it was used for most of the high voltage hydrostatic measurements over the temperature range.

Hysteresis loops as a function of axial loading were measured with the fixture shown in Figure 3. With this fixture, it was possible to reach a pressure of 50,000 psi without breaking the specimens. A short length of PVC tubing was forced over the lower electrode to contain a few drops of silicon oil. The measurements were made at a field strength of approximately 25,000 volts per centimeter.

RESULTS

The materials had room temperature dielectric constants on the order of 320 and curie temperatures which ranged from 160°C for the 30% $PbSnO_3$ compositions to 200°C for the 0% $PbSnO_3$ compositions. The ferroelectric-to-antiferroelectric phase transition was accompanied by a sharp decrease in the dielectric constant, which was easily observed on the temperature-vs-capacitance plots. Figure 4 shows the phase stability as determined by the capacitance measurements.

The thermal expansion measurements showed a number of ferroelectric-to-antiferroelectric transitions as well as the previously noted ferroelectric-antiferroelectric-paraelectric transitions.

Figure 5 shows the general shape of the curves produced by the thermal expansion measuring system. The temperature stability as determined by these diagrams has been plotted as a function of composition in Figure 6. Compositions containing more than 20% $PbSnO_3$ and very close to the antiferroelectric phase boundary became antiferroelectric upon heating. None of the compositions tested became antiferroelectric when cooled below room temperature.

Static axial loading affected the shape of the hysteresis loops as shown in Figure 7. The remanent polarization P_R became less as pressure was increased. Pressure had no effect on the critical field strength E_f. Compositions close to the phase boundary, that is, containing less $PbTiO_3$, suffered a greater reduction of remanent polarization due to the application of pressure than compositions 2 or 3 atom percent away from the boundary, as shown in Figure 8.

Two compositions, those containing 10% and 20% $PbSnO_3$, showed a nonlinear depolarization due to axial loading. Both of these compositions are very close to the antiferroelectric phase boundary.

A hydrostatic vessel was constructed using RTV - silicon rubber as a "fluid." Before pressure was applied, a normal hysteresis loop was obtained, as shown in Figure 9. When pressure was applied, the specimen depoled spontaneously at a critical pressure. Loops made at pressures above this critical level were antiferroelectric, as shown in Figure 10.

When the pressure was reduced to below the critical point, it was possible to switch the material back to the ferroelectric state with a field of approximately 18,000 volts/cm. Once switched, the material remained ferroelectric, as shown in Figure 11.

The pressure vessel used with the RTV silicon rubber failed structurally when used with a second composition. A second vessel was constructed using PVC as a pressure medium. This vessel was much easier to use, as the specimens could easily be replaced. A thermocouple was also provided.

The transition pressures, as determined with this system, are shown as a function of temperature and composition in Figure 12. The compositions containing 10% and 20% $PbSnO_3$ showed the best temperature stability. The effect of composition and temperature on charge output is shown in Figure 13. The two compositions containing 4% $PbTiO_3$ with 10% and 20% $PbSnO_3$ have charge outputs which vary greatly with temperature. These two compositions also showed good pressure stability over the temperature range in Figure 12.

As a compromise between pressure stability and charge output stability, a composition containing 20% $PbSnO_3$ and 5% $PbTiO_3$ has been selected. This composition has a relatively high transition pressure (15-30,000 psi) and a somewhat lower charge output than many of the other compositions. However, these values are relatively stable over the temperature range.

CONCLUSIONS

Either RTV silicon rubber or soft PVC plastic will work satisfactorily as a hydrostatic medium to induce the pressure-enforced ferroelectric-to-antiferroelectric phase transition.

The hysteresis loops made under axial loading and hydrostatic loading show that, in the compositions studied, and with this specimen shape, the ferroelectric-to-antiferroelectric phase transition may be produced by hydrostatic pressure, but not by axial loading.

Further compositional work should be done in order to lower the transition pressure so that a low-cost pressure vessel may be used.

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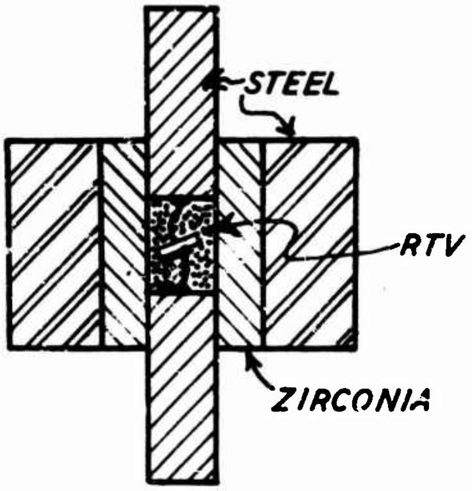

Fig 1 50,000 psi hydrostatic vessel for hysteresis loop with RTV pressure medium

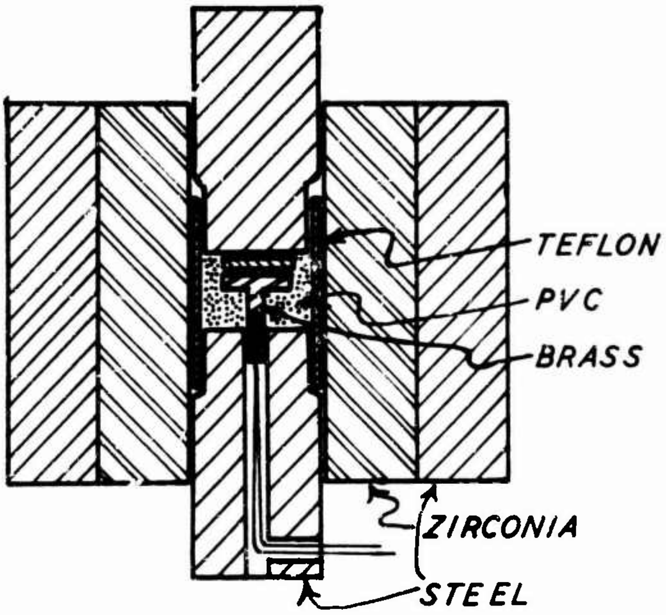

Fig 2 Hydrostatic vessel with soft polyvinyl chloride pressure medium

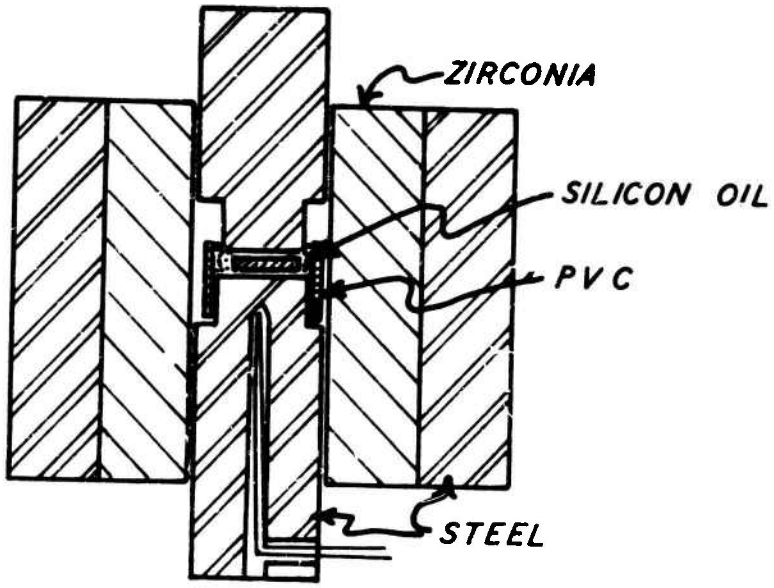

Fig 3 High voltage axial loading sample holder

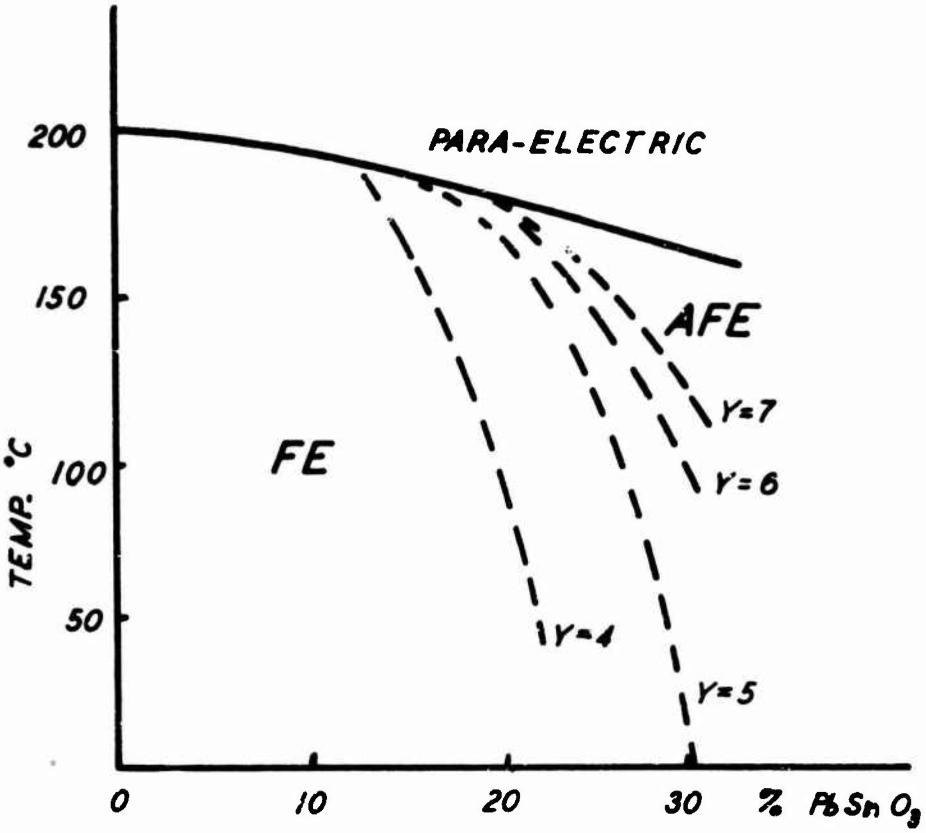

Fig 4 Effect of composition on phase stability

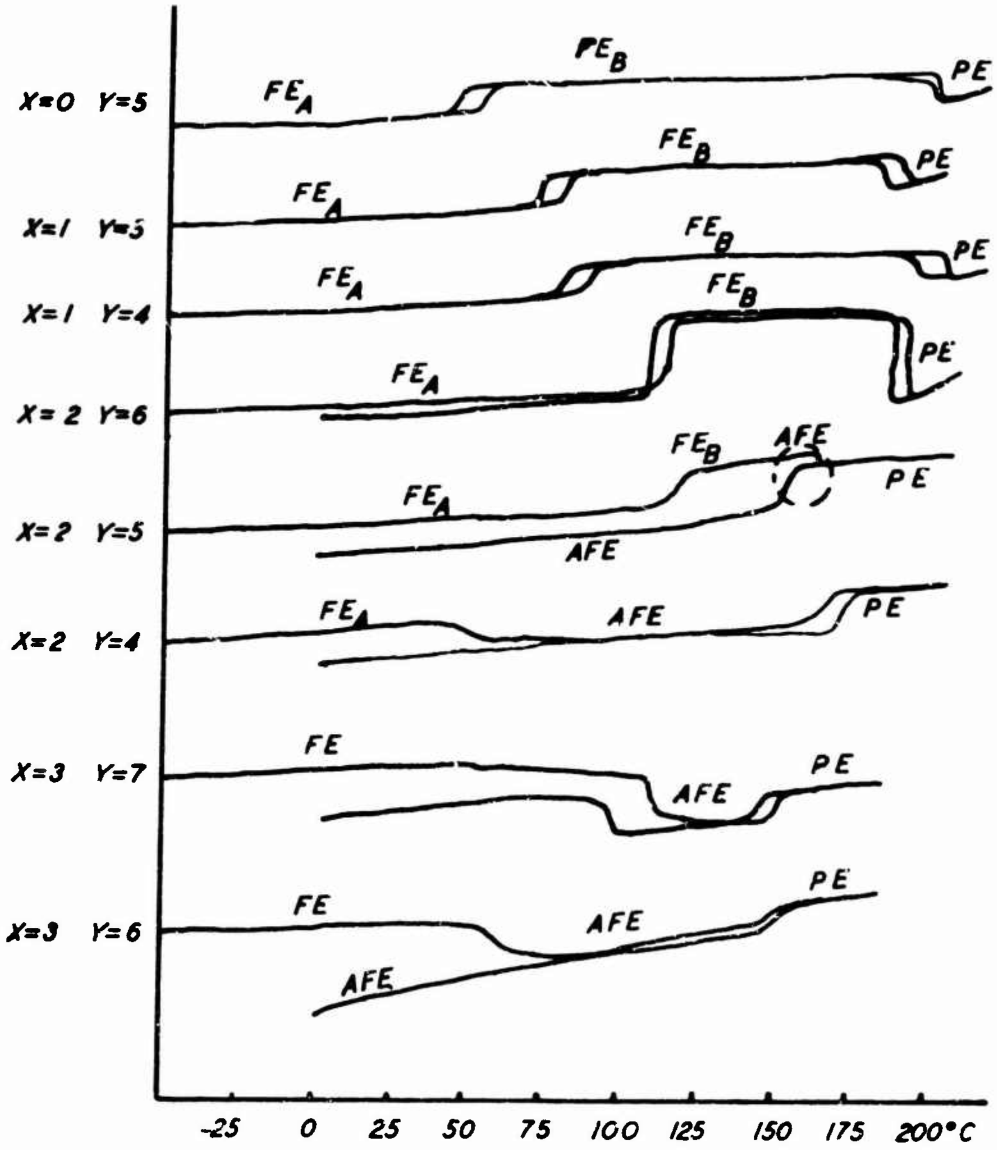

Fig 5 Thermal expansion data, FE_A-rhombohedral, FE_B-rhombohedral, AFE-tetragonal, PE-para-electric-multiple cell cubic

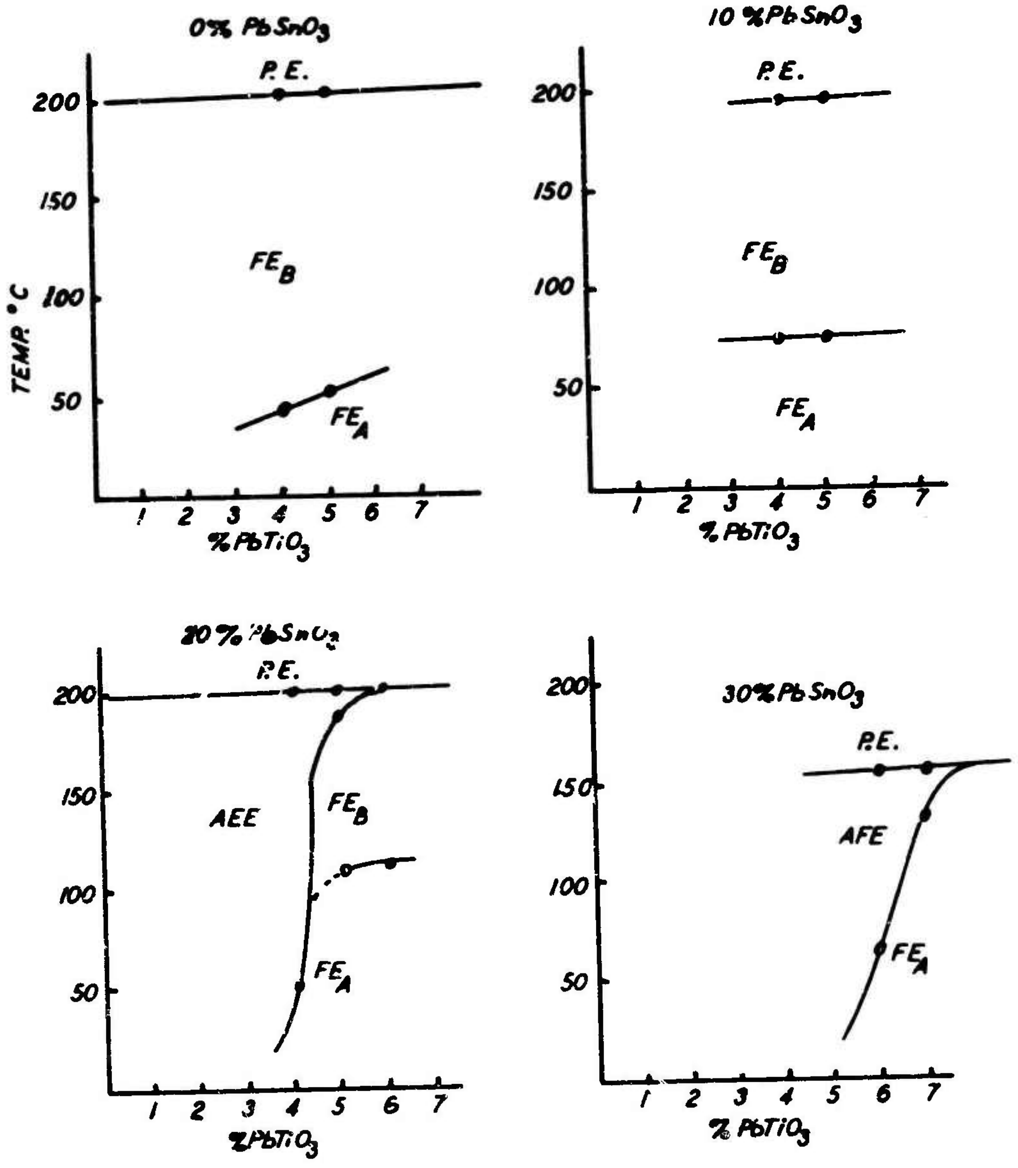

Fig 6 Effect of composition on temperature stability

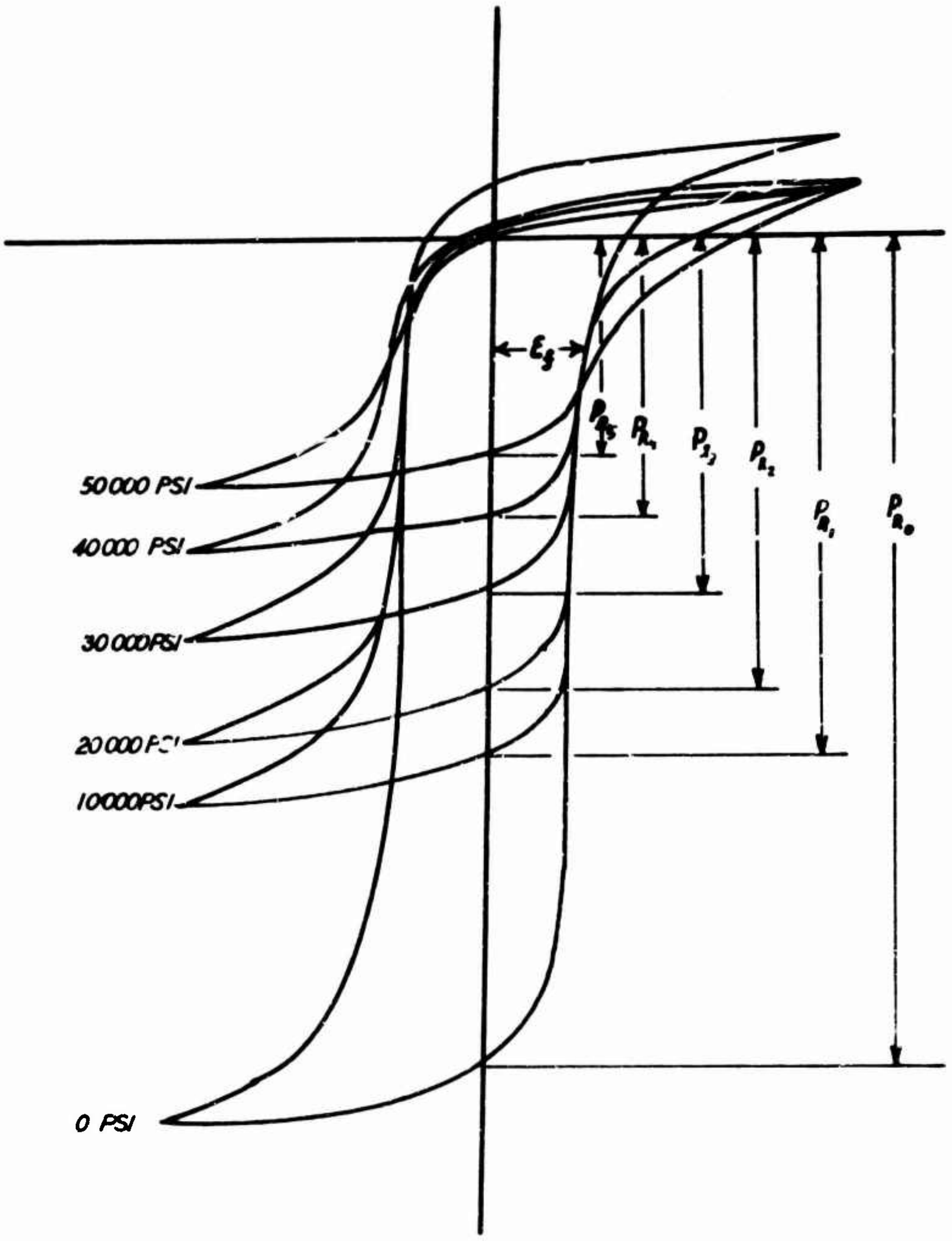

Fig 7 Hysteresis loops vs axial pressure

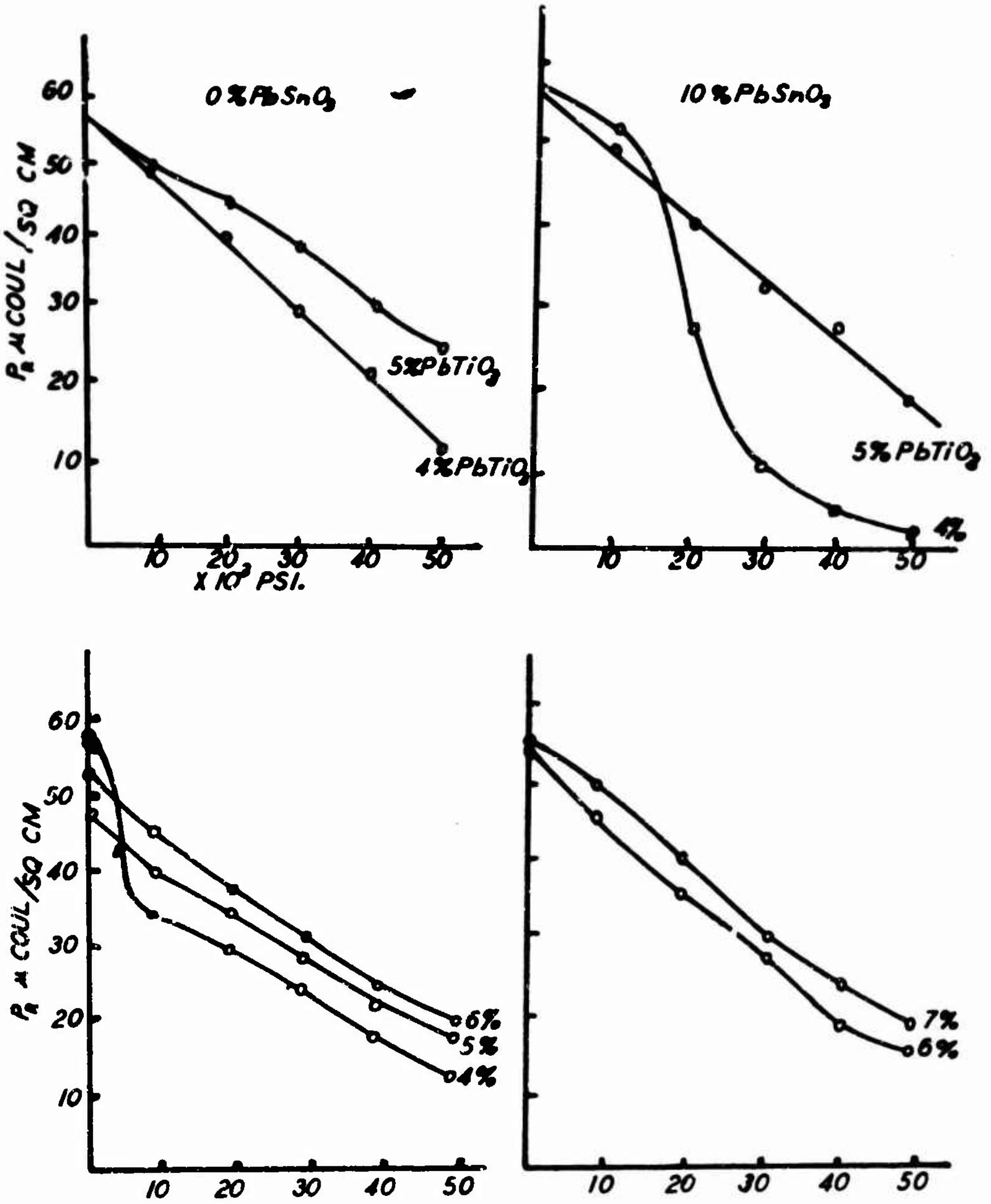

Fig 8 Remanent polarization versus axial pressure. Data from slow hysteresis loops

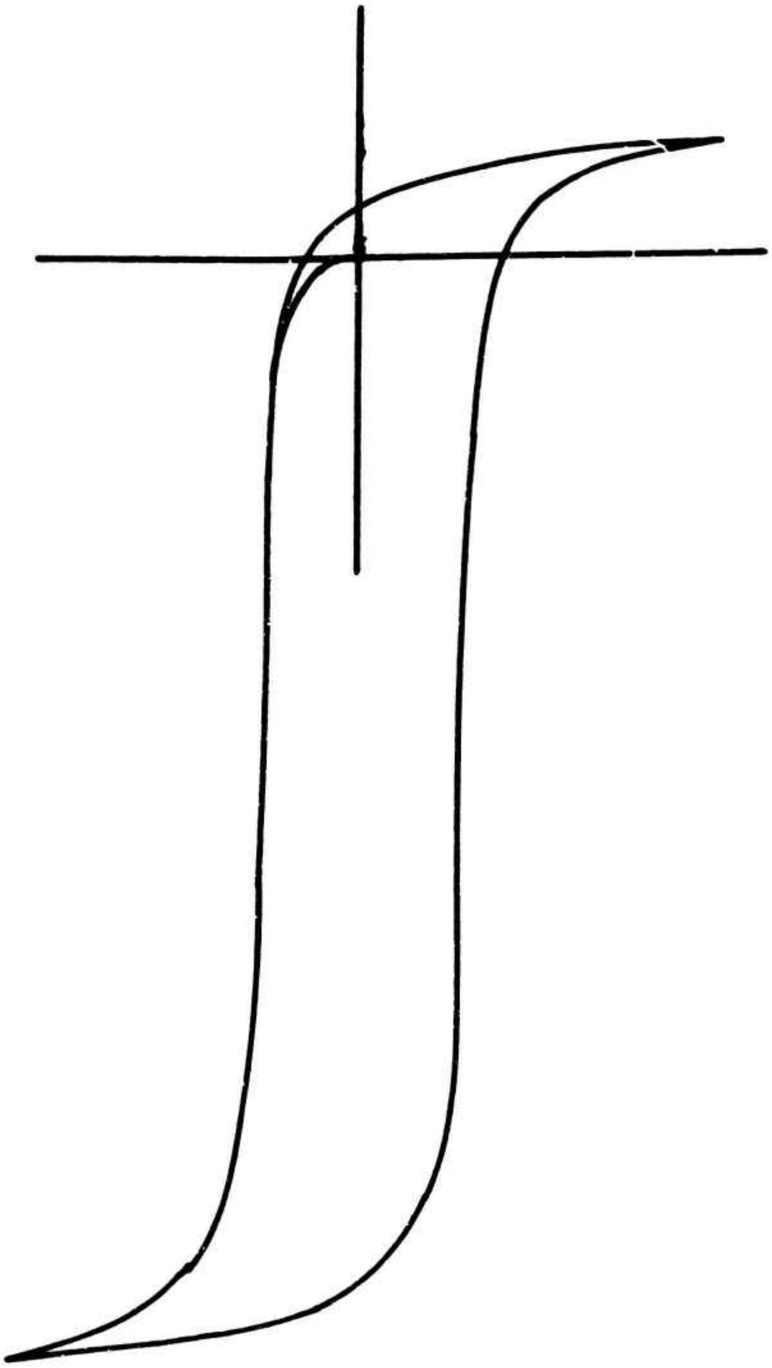

Fig 9 Loop shape of $X = 3$, $Y = 6$, at 0 psi

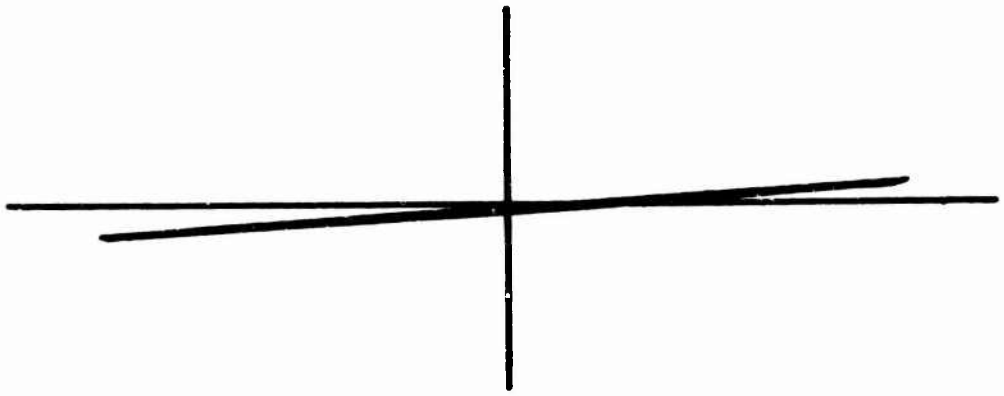

Fig 10 Antiferroelectric loop exhibited by composition X = 3, Y = 6 at pressures above 20,000 psi hydrostatic

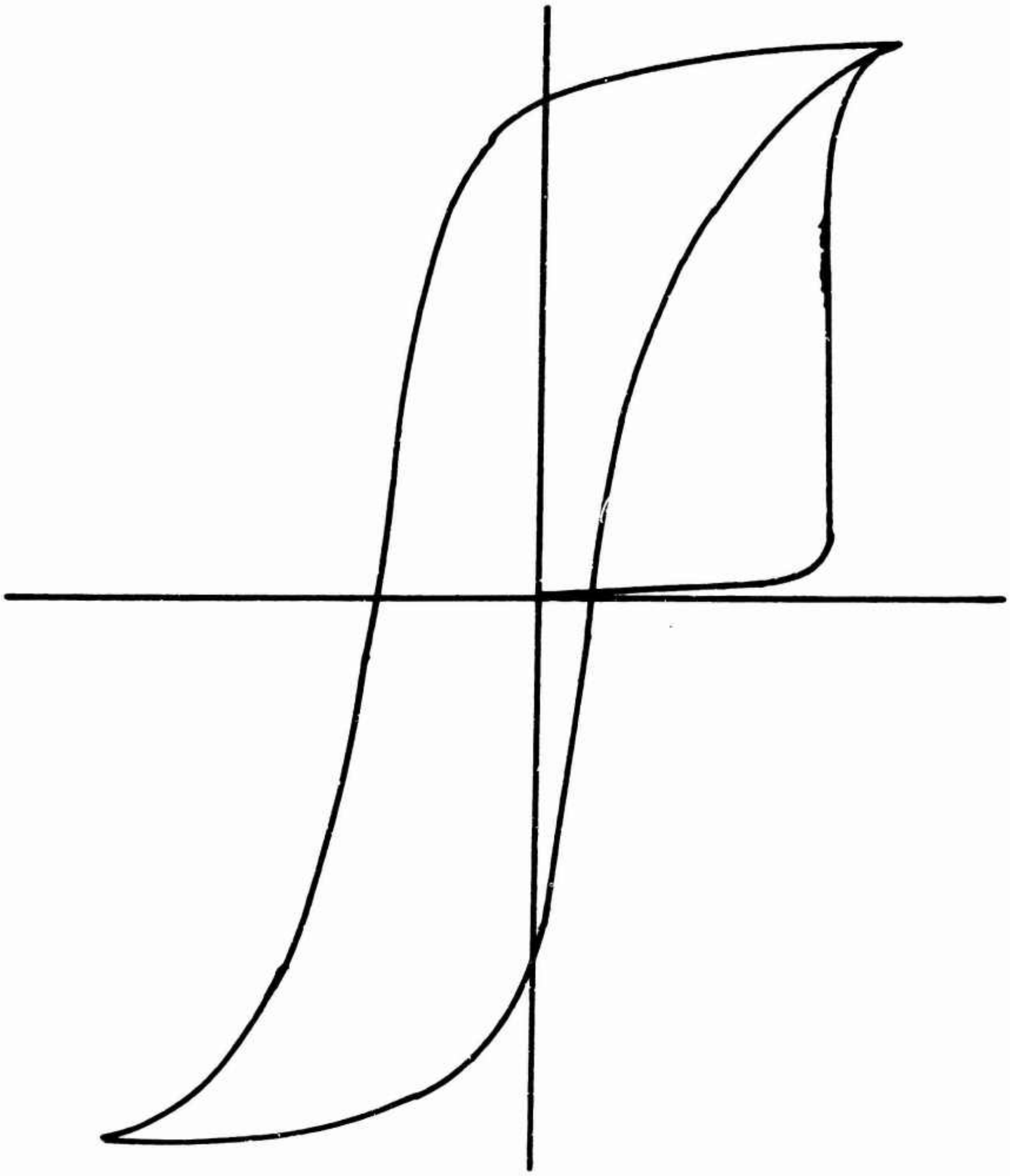

Fig 11   Loop shape at 18,000 psi after cycling to 40,000 psi

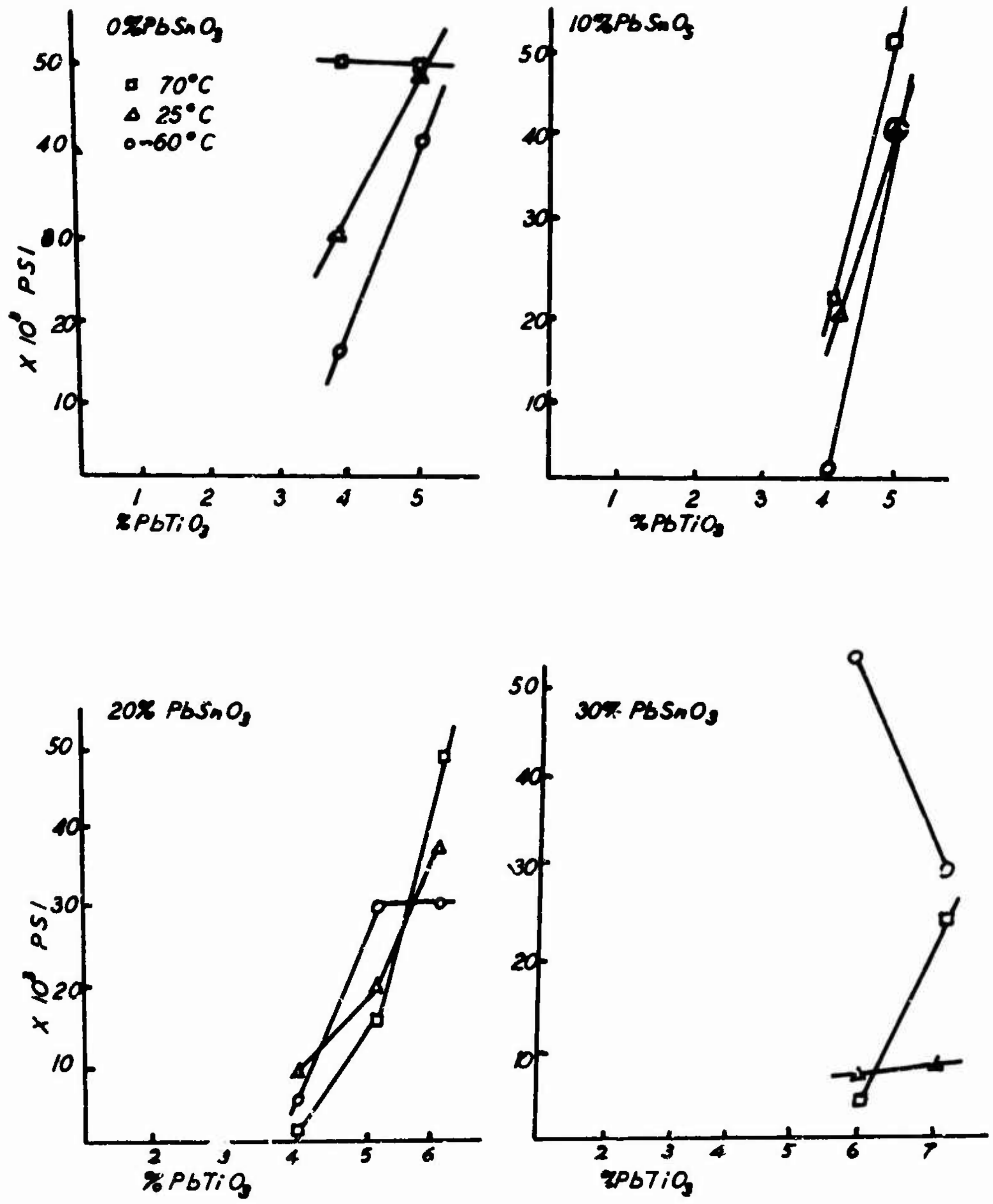

Fig 12 Effect of composition and temperature on transition pressure with hydrostatic loading

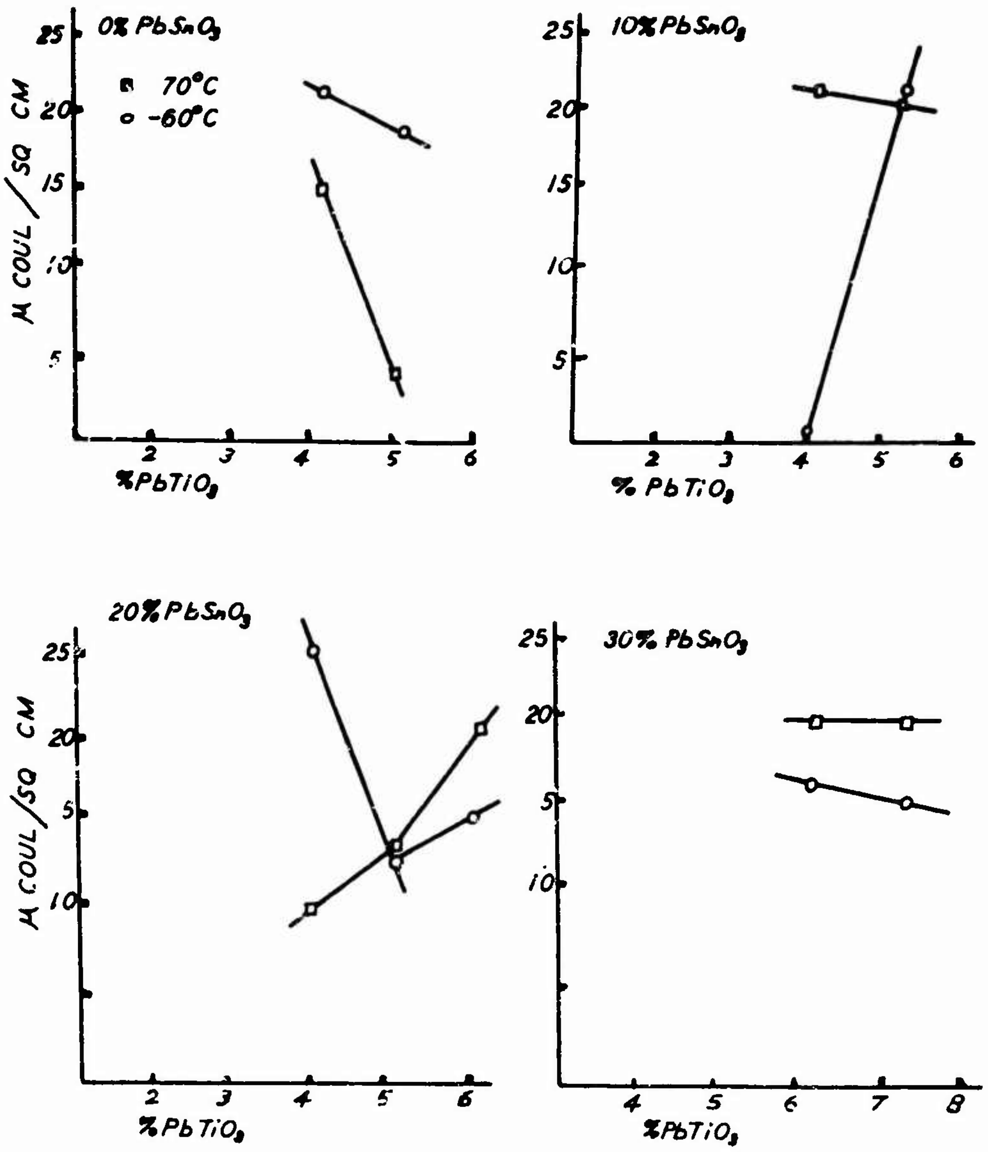

Fig 13 Effect of composition and temperature on output

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Transitional charge release						
Ferroelectric-to-antiferroelectric phase transition						
Fuzing systems						
Piezoelectricity						
Polarization						
Ferroelectrics						
PbSnO$_3$ compositions						
RTV silicon rubber						
Hydrostatic transition pressure						