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# A BUTTERFLY LOOP AUTOMATIC RECORDER FOR FERROELECTRIC AND FERROMAGNETIC MATERIALS

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# ABSTRACT

A Butterfly Loop Automatic Recorder is described which plots the variation of dielectric constant of a specimen of ferroelectric material as a function of applied d-c electric field. Such automatic recording equipment is necessary to accumulate and plot a large volume of data in a reasonable time, as in a survey of ferroelectric materials.

The equipment may be modified to obtain the hysteresogram, or P-E Loop, of a ferroelectric material. With further modification it may be used to plot the variation of permeability of a magnetic toroidal core as a function of dc magnetic field, applied either parallel or perpendicular to the ac field direction.

Specimen preparation, equipment operation and sample results on typical ferroelectric and ferromagnetic materials are described.

# A BUTTERFLY LOOP AUTOMATIC RECORDER FOR FERROELECTRIC AND FERROMAGNETIC MATERIALS

## 1. INTRODUCTION

A Butterfly loop is a plot of dielectric constant, $\epsilon$, vs dc electric field, E, when the latter is cycled symmetrically about zero field. Butterfly loops for dielectric materials take a characteristic shape when the material has nonlinear dielectric properties. Ferroelectric materials, such as barium titanate and the titanate ceramic compounds, when operated in the vicinity of the Curie point[1] or at lower temperatures, exhibit considerable nonlinearity, and are therefore useful in dielectric amplifiers, electric tuning of rf circuits, and other voltage sensitive applications.

For a survey of ferroelectric materials[2] at various temperatures, automatic equipment is required to obtain in a reasonable time the large volume of data needed. The Butterfly Loop Automatic Recording Equipment, or BLARE, was designed to fill this need. A somewhat similar apparatus for investigating barium titanate crystals is described by Drougard and Young[3].

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[1] The Curie point for a ferroelectric material is the temperature at which the maximum dielectric constant is obtained with zero dc electric field.

[2] See L. W. Orr, "$\epsilon$-T-E Surfaces of Ferroelectric Ceramics," Electronic Defense Group, Technical Report No. 53, University of Michigan, October, 1955.

[3] M. E. Grougard and D. R. Young, "Domain Clamping Effect in Barium Titanate Single Crystals," Phys. Rev. 94, 1561 (June 15, 1954).

BLARE may be modified to plot, at a very slow rate, a hysteresogram, or P-E loop, for a ferroelectric material. Such a plot of polarization vs electric field is useful in studying the behavior of certain ferroelectric ceramics and single crystals.

Magnetic materials also exhibit Butterfly loops. In this case the plot is one of permeability vs magnetic field when the specimen is cycled symmetrically about zero field. A further modification of BLARE makes it possible to obtain such plots from toroidal samples of magnetic materials.

## 2. DESCRIPTION OF EQUIPMENT

Figure 1 shows a perspective view of BLARE. Units are rack mounted, and a shelf (7) is furnished to hold the thermos flask (3) containing the specimen, and any auxiliary equipment needed. All units are powered by the Sorensen regulator (8) which insures stability of operation.

A block diagram of the system is shown in Fig. 2. The specimen (3) is excited by a 1000 cycle generator (1). The ac current flowing in resistor $R_2$ is equal to $A \frac{dp}{dt}$, the specimen area multiplied by its time-rate of change of polarization. This ac current is thus proportional to the dielectric constant, $\epsilon$, at any particular value of applied electric field, E.

The voltage across $R_2$ is the input to the amplifier and detector (4) whose dc output is recorded by the pen of the Moseley Autograf X-Y recorder (5). The pen displacement is therefore proportional to $\epsilon$, the dielectric constant. Standard capacitors built into the equipment permit a direct capacitance calibration. The relative dielectric constant, $\epsilon$, is then obtained from the relation

$$\epsilon = 4.45 \frac{dC}{A} \qquad (1)$$

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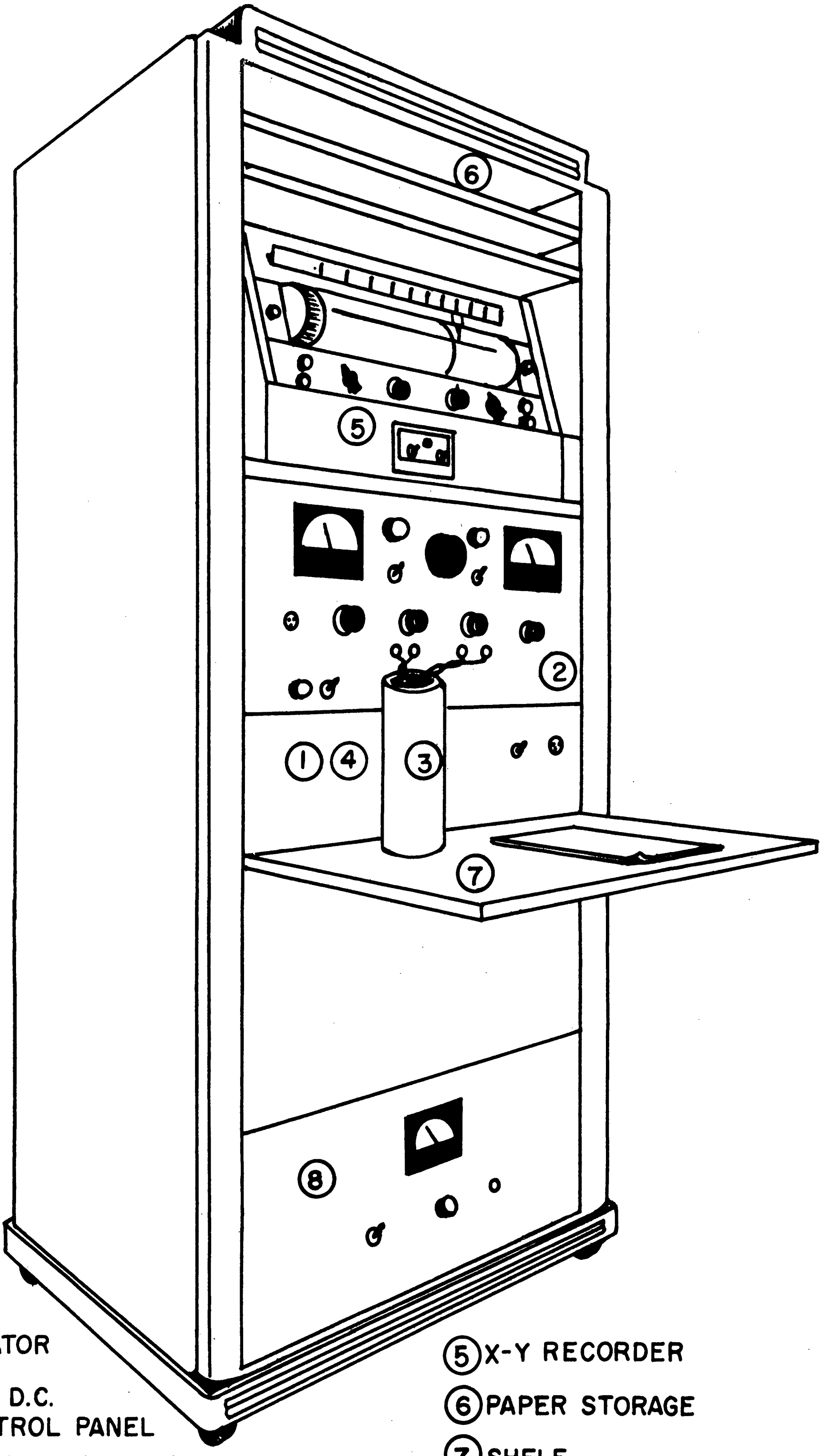


① SIGNAL GENERATOR

② MOTOR DRIVEN D.C.
SUPPLY & CONTROL PANEL

③ SPECIMEN IN THERMOS FLASK

④ AMPLIFIER & DETECTOR

⑤ X-Y RECORDER

⑥ PAPER STORAGE

⑦ SHELF

⑧ 500 WATT SORENSON
REGULATOR

FIG. 1
BLARE
(BUTTERFLY LOOP AUTOMATIC RECORDER)

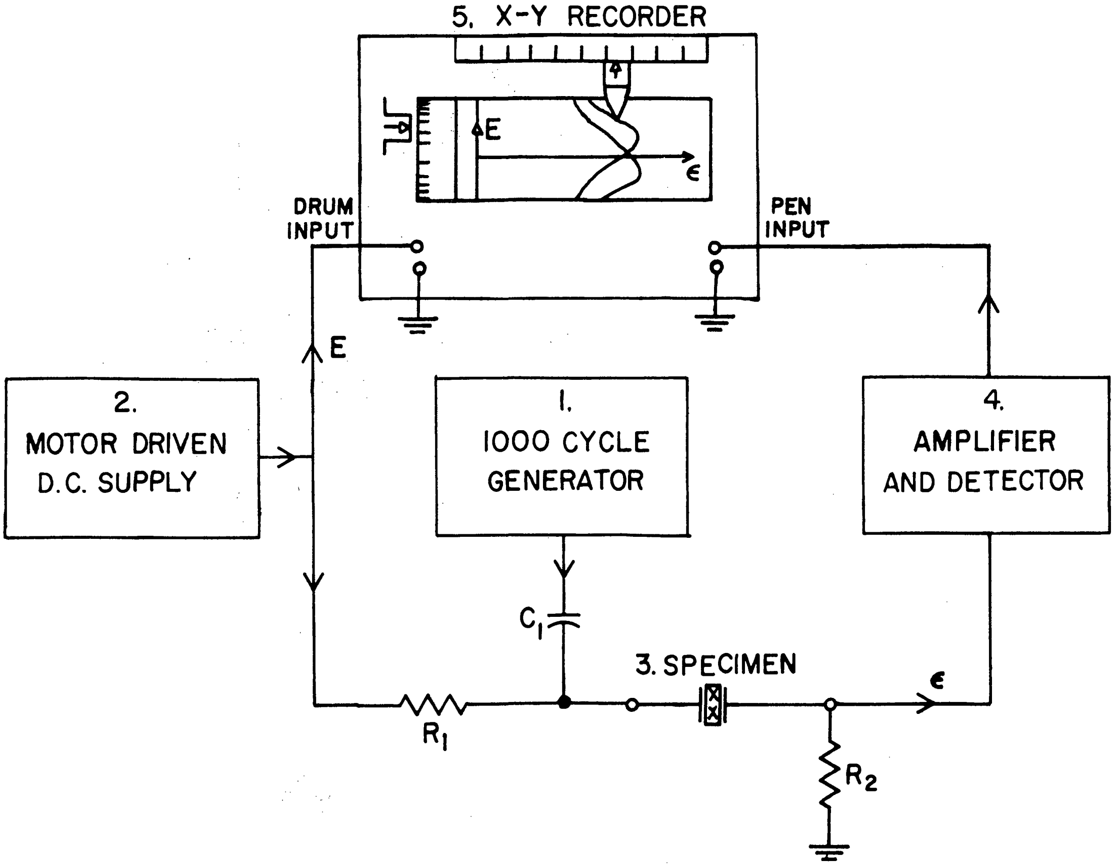


FIG. 2

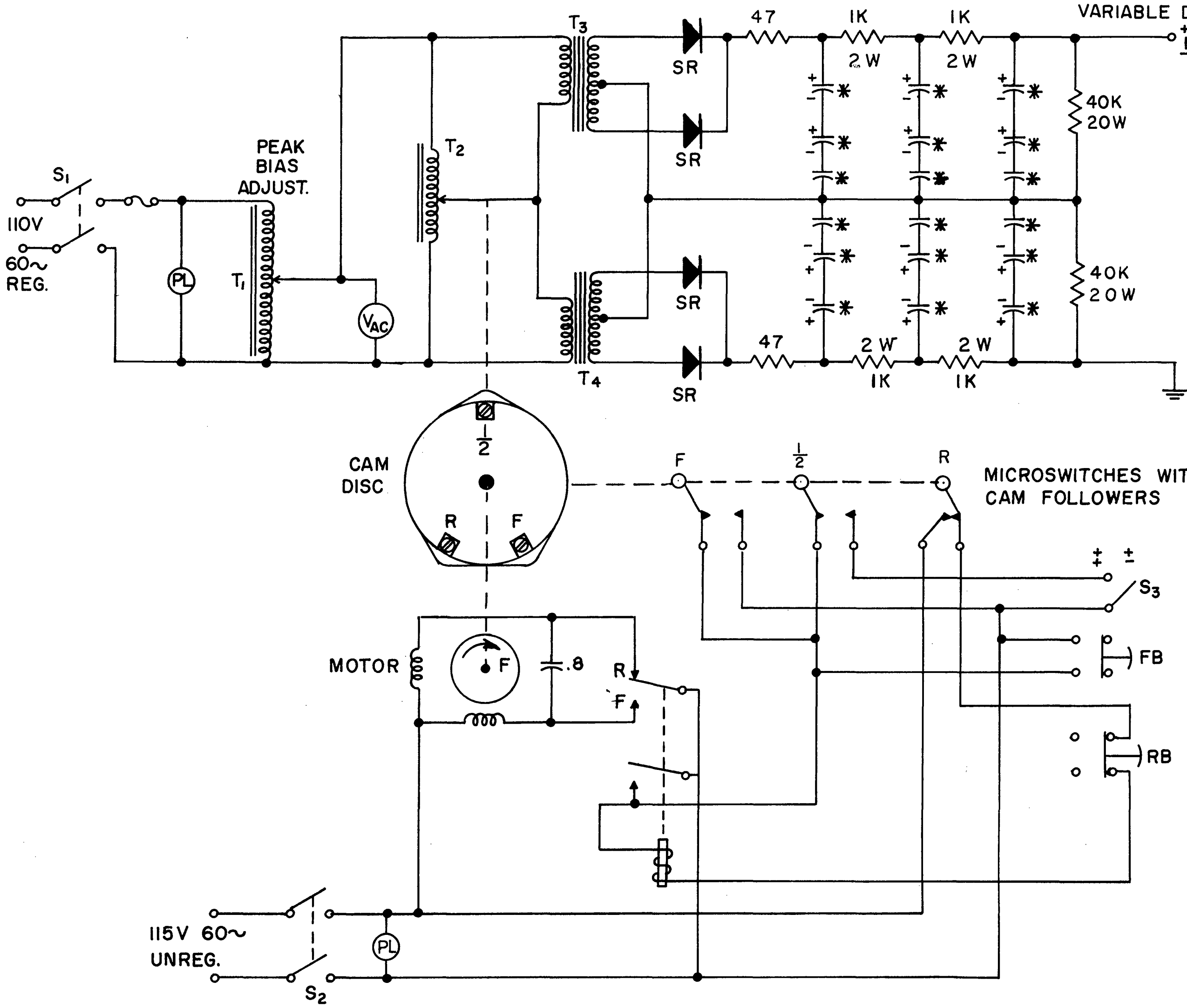
BLOCK DIAGRAM OF BLARE

where C is the specimen capacitance (in micromicrofarads), d is the thickness of the dielectric (in inches), and A is the area of one electrode (in square inches).

While the dielectric constant is being indicated in this manner by the pen position, a slowly varying electric field is applied to the specimen by the motor-driven supply (2). The variable voltage, E, from this supply is fed to the specimen by resistor $R_1$. A fixed fraction of this voltage is also applied to the drum input of the X-Y recorder. Upon lowering the pen of the recorder, a Butterfly loop of the specimen is obtained after one cycle of the voltage supply.

## 3. VARIABLE DC VOLTAGE SUPPLY

A smoothly varying voltage is required to drive the specimen in an automatic reversing cycle. The circuit of this supply is shown in Fig. 3. To obtain smooth variation over the full cycle from positive to negative voltage, a pair of supplies in push-pull are used. These are fed from the upper and lower sections of variac $T_2$. This variac is coupled by a V-belt and cone pulleys to a two phase reversible motor equipped with reduction gearing. The reversing cams marked R, 1/2, and F are staggered axially, and each one engages only the follower on the microswitch which is marked in the same manner. The cam disk is directly connected to the variac shaft. With the switch S3 open, the cam disk will oscillate between position R and F, engaging in turn the R, 1/2 and F microswitches, which are mounted in a line. When S3 is closed, the system will oscillate between cam positions 1/2 and R. The forward and reverse buttons marked FB and RB afford manual control of the direction of rotation when it is desired to override the cam control. All three cams can be moved on the rim of the cam disk to give any desired reversing positions. The variable dc output of this network, can be swept from +1000 v to -1000 v or any intermediate



SR - SELENIUM RECTIFIER 2KV PEAK INVERSE (SARKES - TARZIAN)

✱ - 450 V 40$\mu$f ELECTROLYTIC

FIG. 3

MOTOR DRIVEN D.C. HIGH VOLTAGE SUPPLY UNIT FOR BLARE voltage. When S3 is closed, the voltage is swept from 0 to plus 1000 v or to any intermediate voltage. The peak voltage excursion is determined by the setting of variac $T_1$.

By means of the cone pulley, two rates are available. At the slow rate, variac $T_2$ is driven from one extreme to the other in 40 seconds, and at the fast rate, in 10 seconds.

## 4. OSCILLATOR AND AMPLIFIER-DETECTOR

The amplifier detector circuit is shown in Fig. 4 A 1 kc signal from a General Radio Unit Oscillator is applied to the sample. The signal voltage to the sample may be varied in increments between 10 mv and 10 v rms by switch $S_1$. The selector switch $S_2$ permits selection of any one of three standard mica capacitors, or the test specimen, X. A direct capacitance calibration is thus furnished by the standard capacitors which are not voltage sensitive. The drum and the pen deflection sensitivities can be controlled continuously by the two variable resistors marked drum gain and pen gain. The closing of switch $S_3$ gives a zero reference output to permit the setting of the pen zero on the recorder.

The three stage audio amplifier is designed with a restricted bandwidth to reject high frequency noise and hum pickup. The output is rectified by the 1N34 diode giving a dc voltage proportional to the 1000 cycle capacitor current in the test specimen or the the current in the calibrating capacitor selected by $S_2$. Gain settings are adjusted to operate the diode in the most linear region of its characteristic, and a high degree of linearity is thereby obtained. At the maximum voltage gain, the ratio of dc output to rms input is approximately 3000 at 1 kc.

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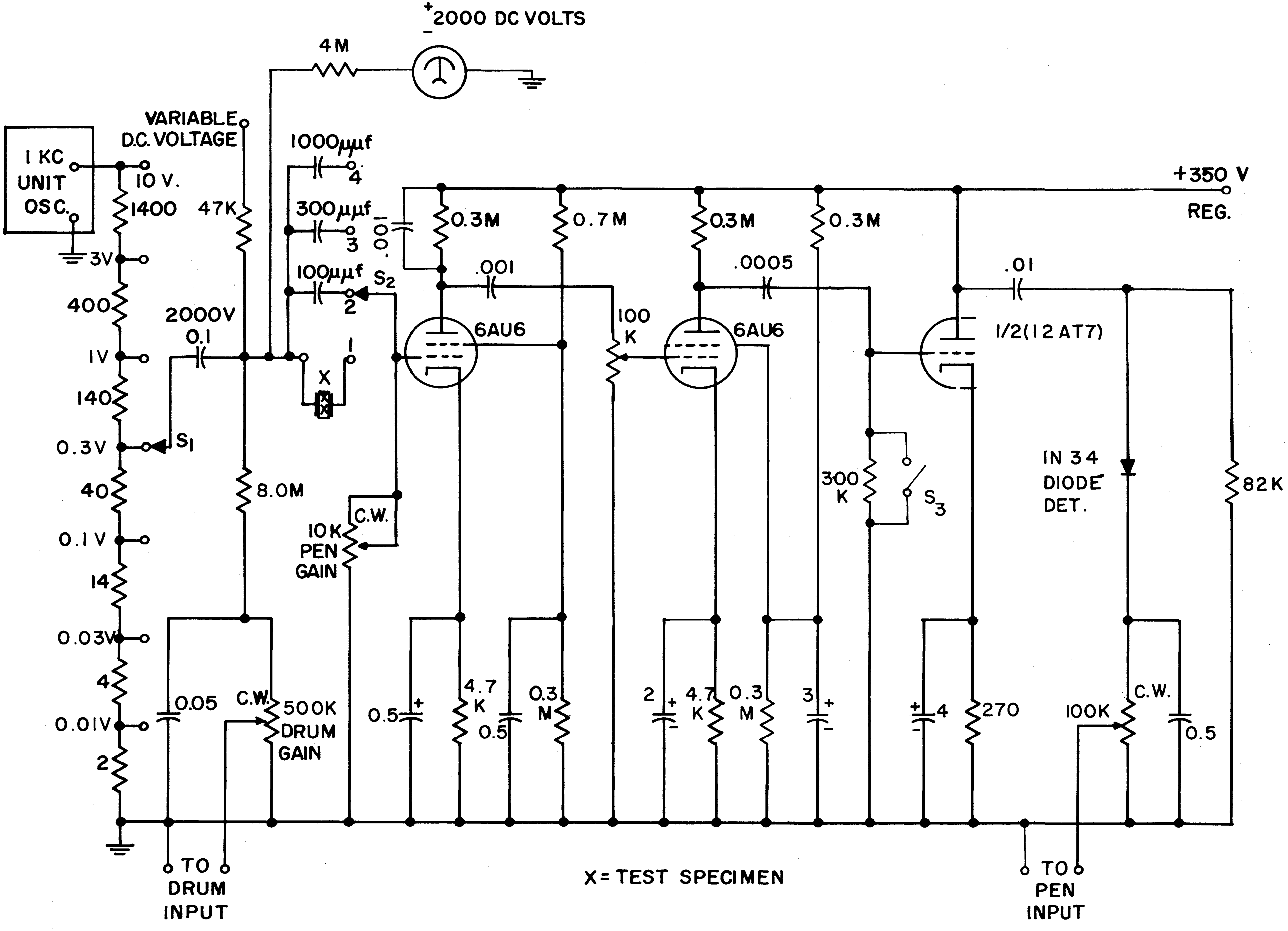

FIG. 4

AMPLIFIER-DETECTOR FOR BLARE

The variable dc driving voltage is fed to the test specimen through a 47K ohm isolating resistor. This prevents attenuation of the 1000 cycle signal at the injection point. A 2000 volt capacitor is used to inject the 1000 cycle signal since this capacitor is also subjected to the high voltage excursions impressed upon the specimen.

Charging and discharge currents due to the variation of the high voltage drive must flow through the 10K ohm Pen Gain potentiometer, but these currents are so small as to have no appreciable effect on the grid bias of the first amplifier stage.

To obtain the drum input, which must be proportional to the dc driving voltage, a voltage divider consisting of an 8 megohm resistor in series with the 500K ohm Drum Gain potentiometer is used. A capacitor across the Drum Gain potentiometer prevents 1000 cycle and hum pickup from entering the drum input of the X-Y recorder.

A dc voltmeter with zero center scale is used to monitor the voltage applied to the test specimen.

The heater power and 350 volt regulated dc power required by the unit are furnished by a conventional power supply and an Applegate regulator unit.

## 5. SPECIMEN PREPARATION AND PROCEDURE

To avoid the necessity of making corrections for fringing capacitance and for other reasons[1], specimens are prepared with electrodes plated to the edge. This is done by applying a metallic coating to both sides of a thin wafer of the material, and dicing the specimen with a narrow diamond wheel. For the

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[1] It was found that better high frequency performance resulted from this construction.

dicing operation the specimen is temporarily cemented to a glass microscope slide. The slide is cemented to a steel flat and located on the magnetic chuck of a precision grinder as shown in Fig. 5. After cutting, the plated squares of ceramic are removed, cleaned and dried.[1] A typical specimen is 0.1 to 0.02 inch square and 0.02 inch thick.

The electrode material generally used is silver. It may be applied by silver painting and firing or by vacuum evaporation. A thin even coating is desirable.

For a quick test, the specimen may be placed in a clip holder (Fig. 5A) and operated in transformer oil. For a more permanent unit, electrode wires are attached by a careful solder operation (Fig. 5B). The specimen is then encapsulated in a resin bead to exclude moisture and avoid surface breakdown along the edge of the ceramic when the high voltage is applied. Care must be taken to remove all traces of mositure from the specimen before encapsulating. Even small traces of moisture cause leakage losses and premature electric breakdown.

If the specimen is heated in preparation, it must be properly aged before measurements are taken. After a temperature excursion, many ferroelectric materiald experience an aging process indicated by a slow decrease in capacitance with time. Typical aging of a ferroelectric material after being heated to the boiling point and chilled to room temperature is indicated in Fig. 6. It is seen that after 24 hours the capacitance value is still slowly decreasing. High $\epsilon$ dielectrics appear to be more sensitive in this regard than low $\epsilon$ materials.

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[1] For a full discussion of ferroelectric specimen preparation and testing, see H. Diamond, "Miniature Nonlinear Capacitors," Electronic Defense Group Technical Report No. 54, University of Michigan, December, 1955.

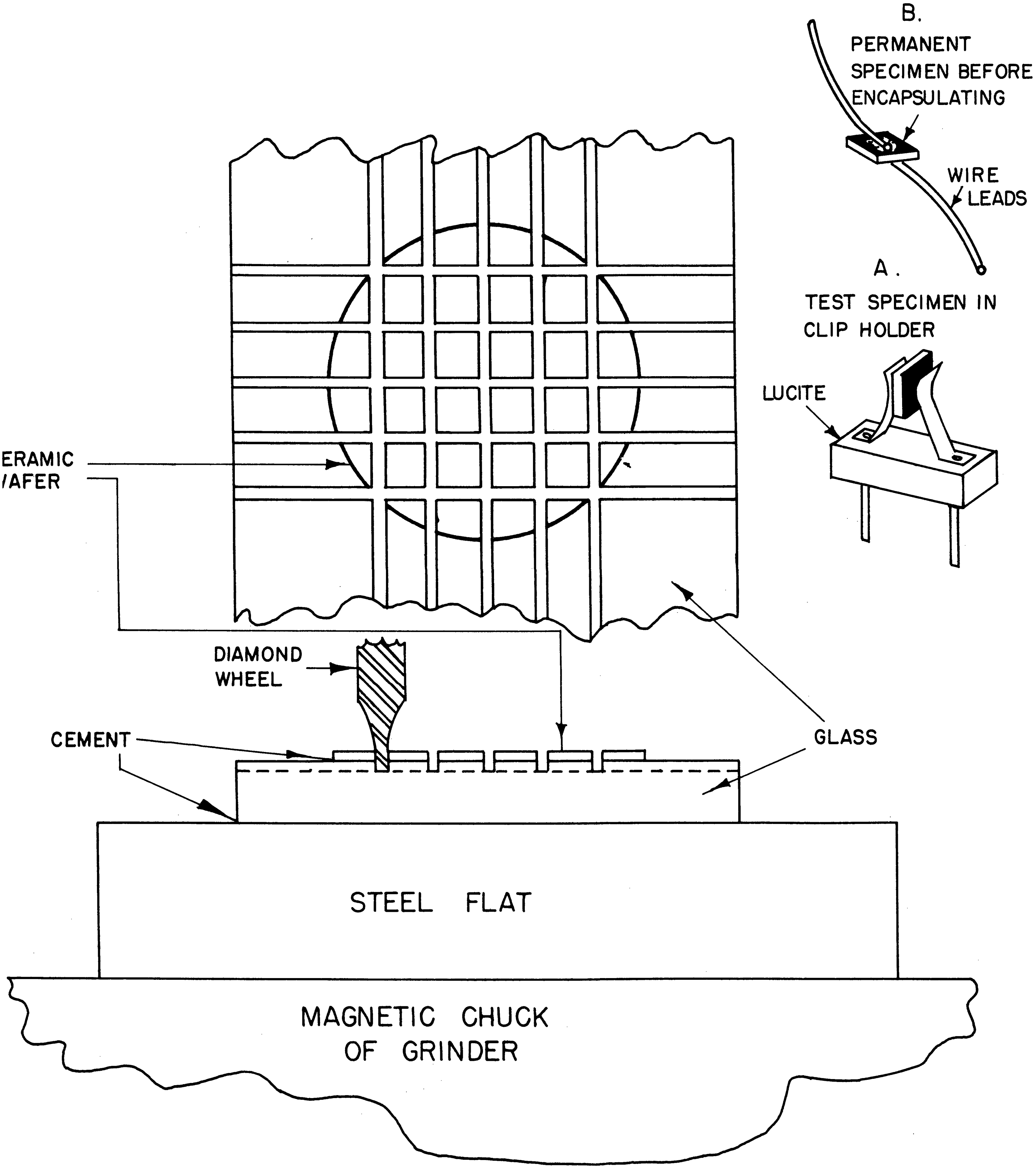
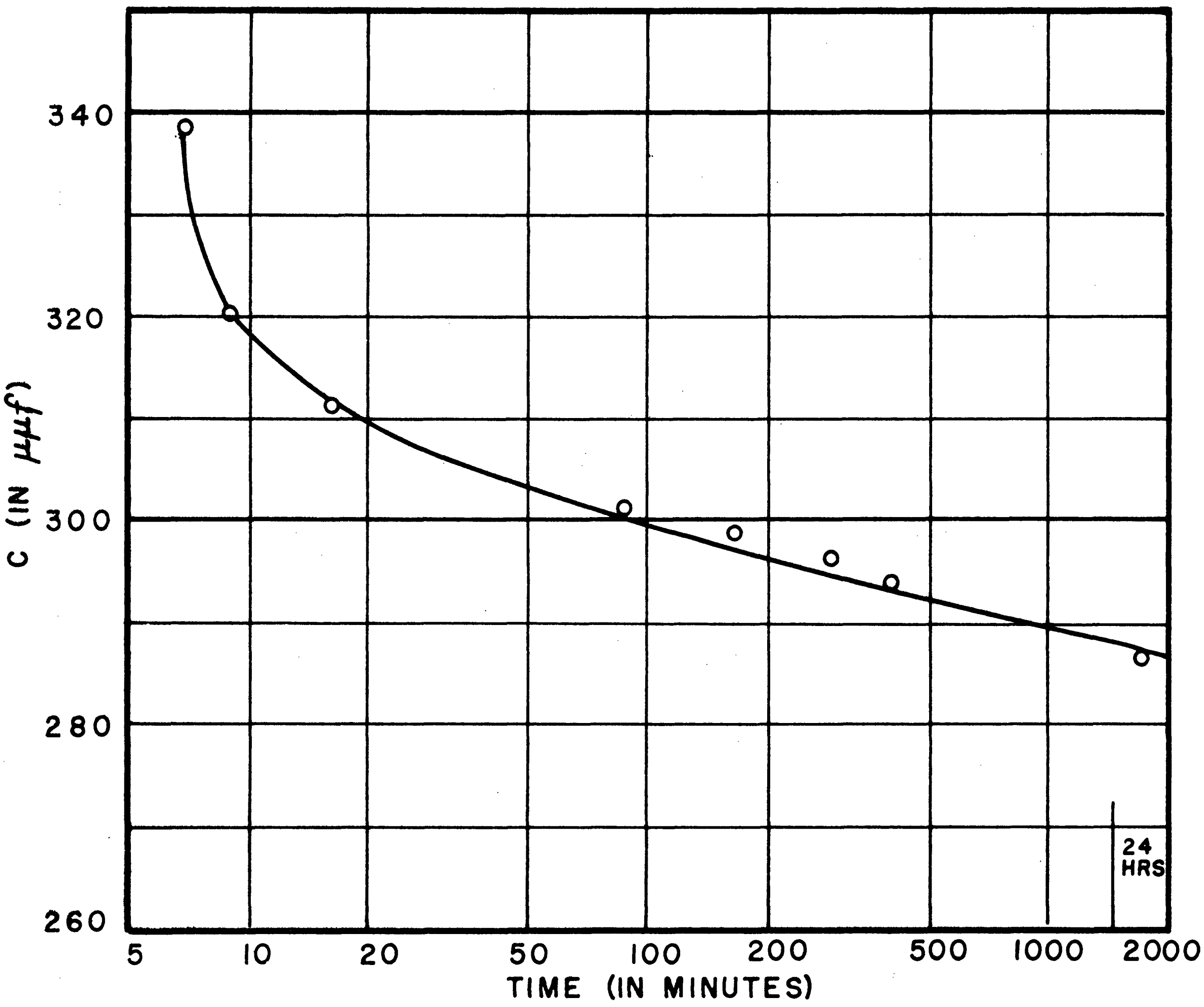


FIG 5

SPECIMEN PREPARATION



C VS TIME. AGING AFTER HEATING TO 100°C AND QUENCHING AT 25°C, AEROVOX "HI-Q" BODY NO 41.

FIGURE 6

This point must be kept in mind when data are to be taken at different temperatures. The standard procedure is to keep the specimen at the lowest temperature for a considerable period of time before making the first recording of the temperature run. When the temperature is raised, the material responds relatively quickly to its ultimate value, and a second recording may be made without waiting.

When a set of Butterfly loops at different temperatures is to be taken, the specimen is immersed for a long period in a cold oil bath as shown in Fig. 7. After the data are taken at the lowest temperature, power is applied to a heater in the oil bath for a short interval. A variac and heater terminals are furnished on the front panel of BLARE for this purpose. After adequate heat has been added, the bath is stirred with a stirring motor until the thermometer reading becomes steady. The Butterfly loop for that temperature is then recorded.

## 6. MODIFICATION OF BLARE FOR P-E LOOP PLOTTING

By removing the oscillator drive and amplifier detector, BLARE may be rearranged for P-E loop plotting as indicated in the block diagram in Fig. 8. When the test specimen is cycled by the variable dc voltage, the charging current is $A \frac{dp}{dt}$. If this current is integrated by an integrating capacitor $C_2$, the capacitor voltage will be proportional to the polarization, p. In making this plot, the same motor-driven dc supply is used. A high input impedance Kiethley Electrometer measures the potential across the integrating capacitor. The electrometer delivers an amplified signal which drives the pen of the recorder. In this way, a slow P-E hysteresis loop can be recorded.

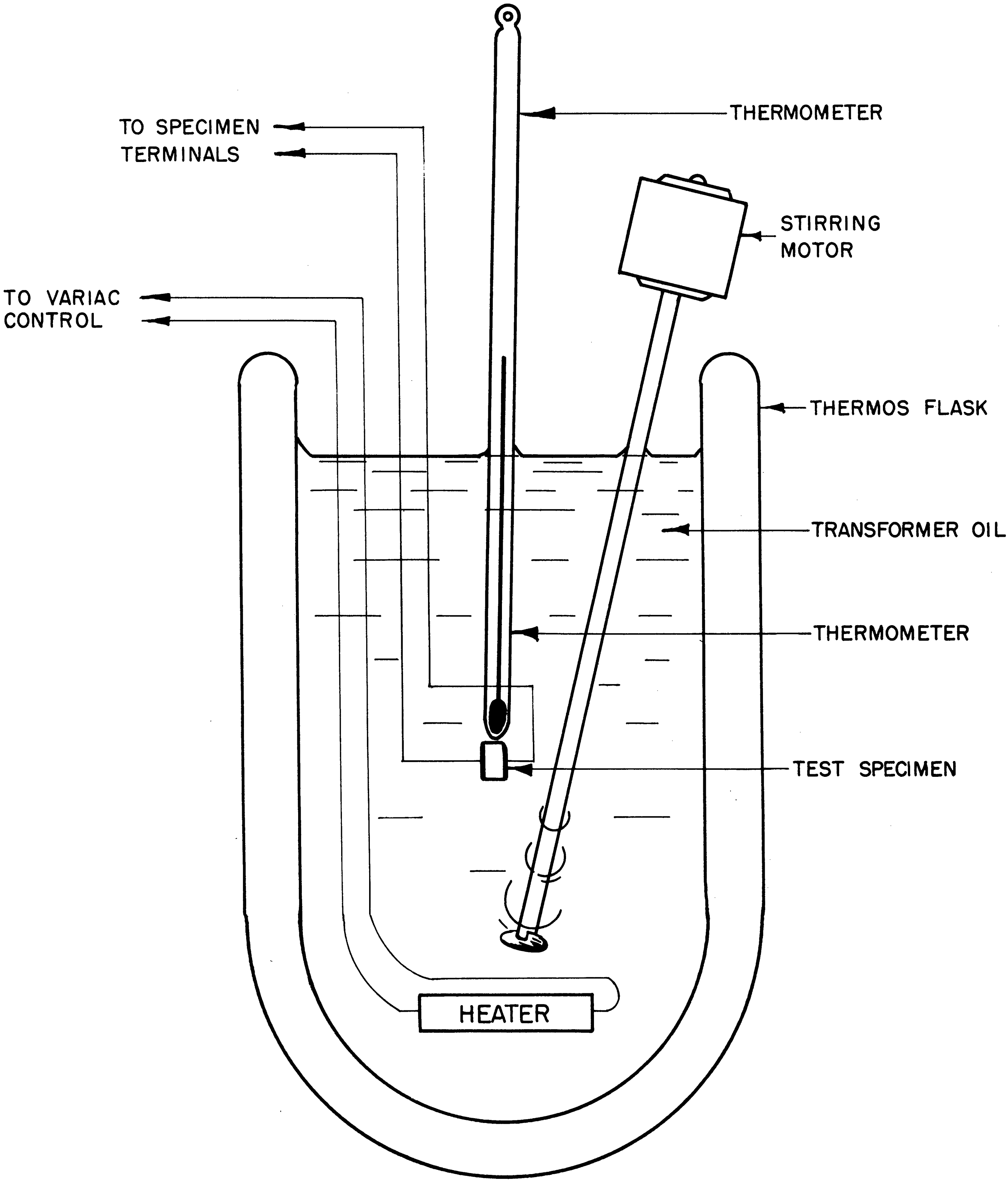


FIG 7

CONSTANT TEMPERATURE BATH FOR FERROELECTRIC SPECIMENS

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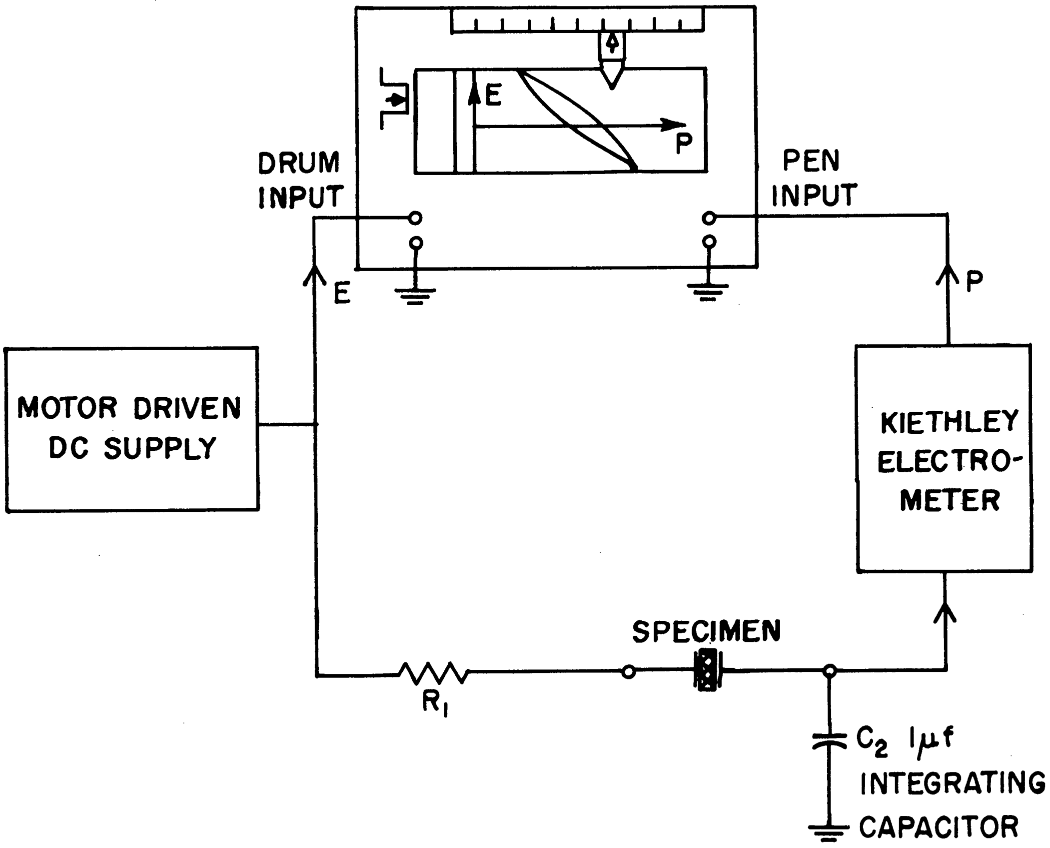

FIG. 8

BLARE UNIT MODIFIED
FOR SLOW PE LOOP PLOTTING

## 7. BUTTERFLY AND P-E LOOPS

A series of Butterfly loops for various commercial capacitors is shown in Fig. 9. All of these capacitors are of the ferroelectric type, but it can be seen that the two upper curves show little change with applied voltage. In this plot, the dielectric constant was not calculated for the capacitors, but for specimens of known dimensions the dielectric constant may be obtained from Eq. 1.

Figure 10 shows corresponding C-E and P-E loops plotted on the same voltage axis for a typical commercial material. In this case the plot was done at low speed, a complete cycle taking approximately 100 seconds. One difficulty in obtaining satisfactory P-E loops at very low speed is the inherent leakage resistance of the capacitor. Although this resistance is generally of the order of thousands of megohms or greater, giving very small leakage currents, the charging currents at low speeds are also very small. In this case the integrator error may be appreciable, and the resulting P-E loop distorted.

## 8. $\epsilon$-T-E SURFACES

By cycling a sample between zero field and a positive value, a one sided, or half-butterfly loop is obtained. This type of plot is useful in designing voltage tunable devices. If a number of plots of this sort are made at successively increased temperatures, a temperature family is obtained. A convenient way of presenting these data is on a three dimension plot, or $\epsilon$-T-E surface. A typical $\epsilon$-T-E plot is shown in Fig. 11. This plot indicates the changes in dielectric constant as both applied electric field and temperature are varied. The dielectric material is a barium-strontium titanate ceramic, and is used in voltage tunable devices at high frequencies. It is interesting to

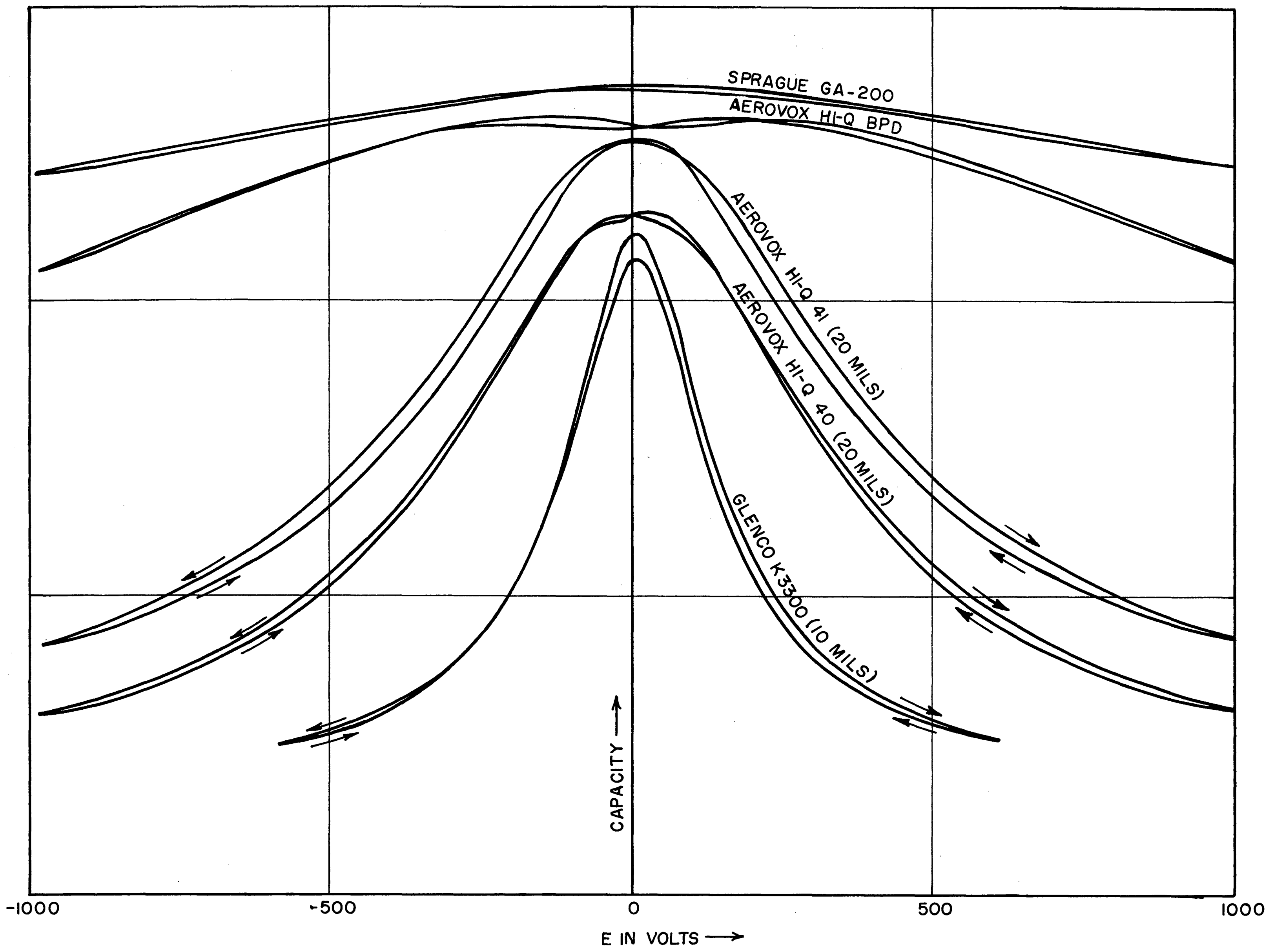

FIG. 9

BUTTERFLY (C-E) LOOPS FOR VARIOUS COMMERCIAL CAPACITORS

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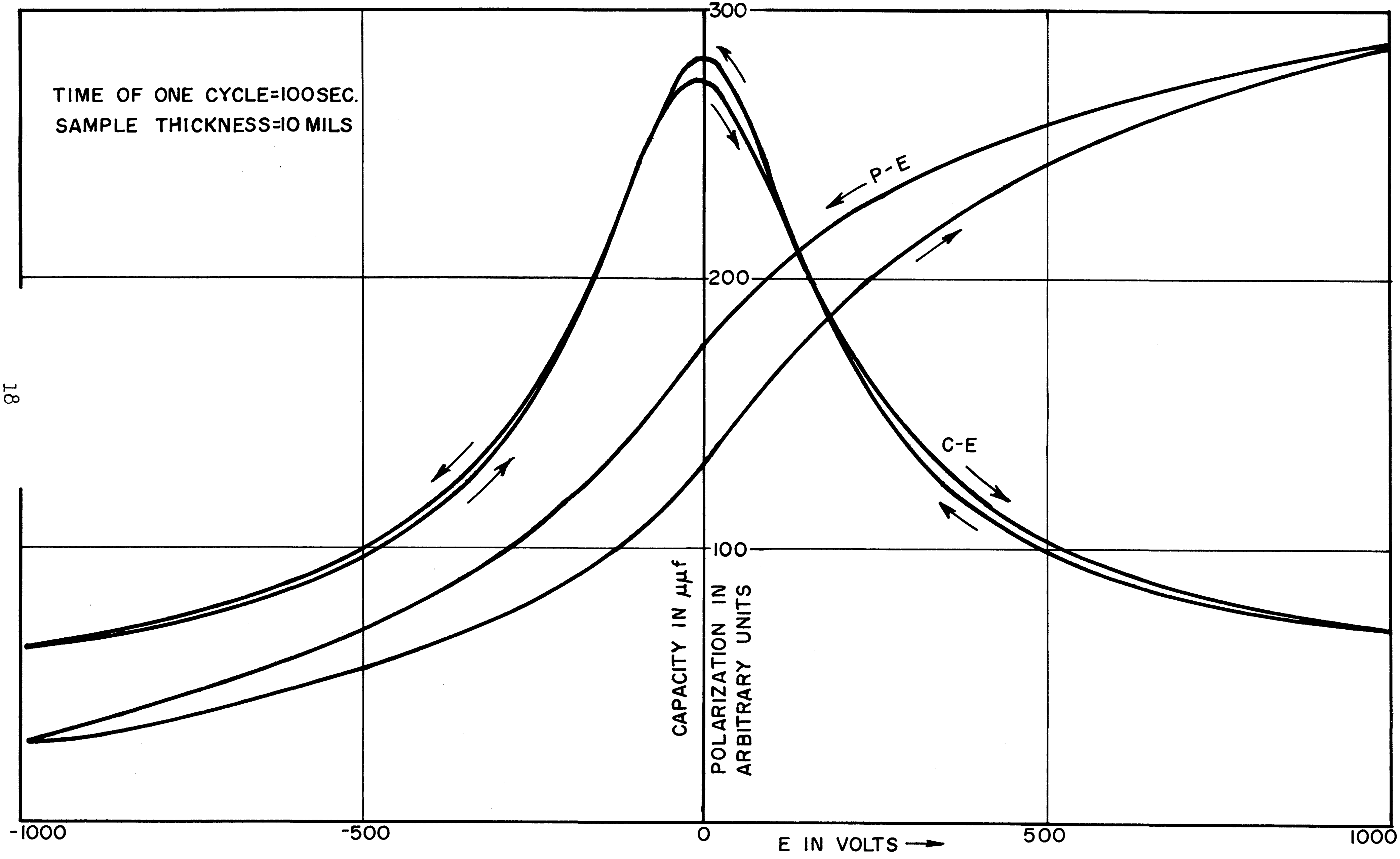
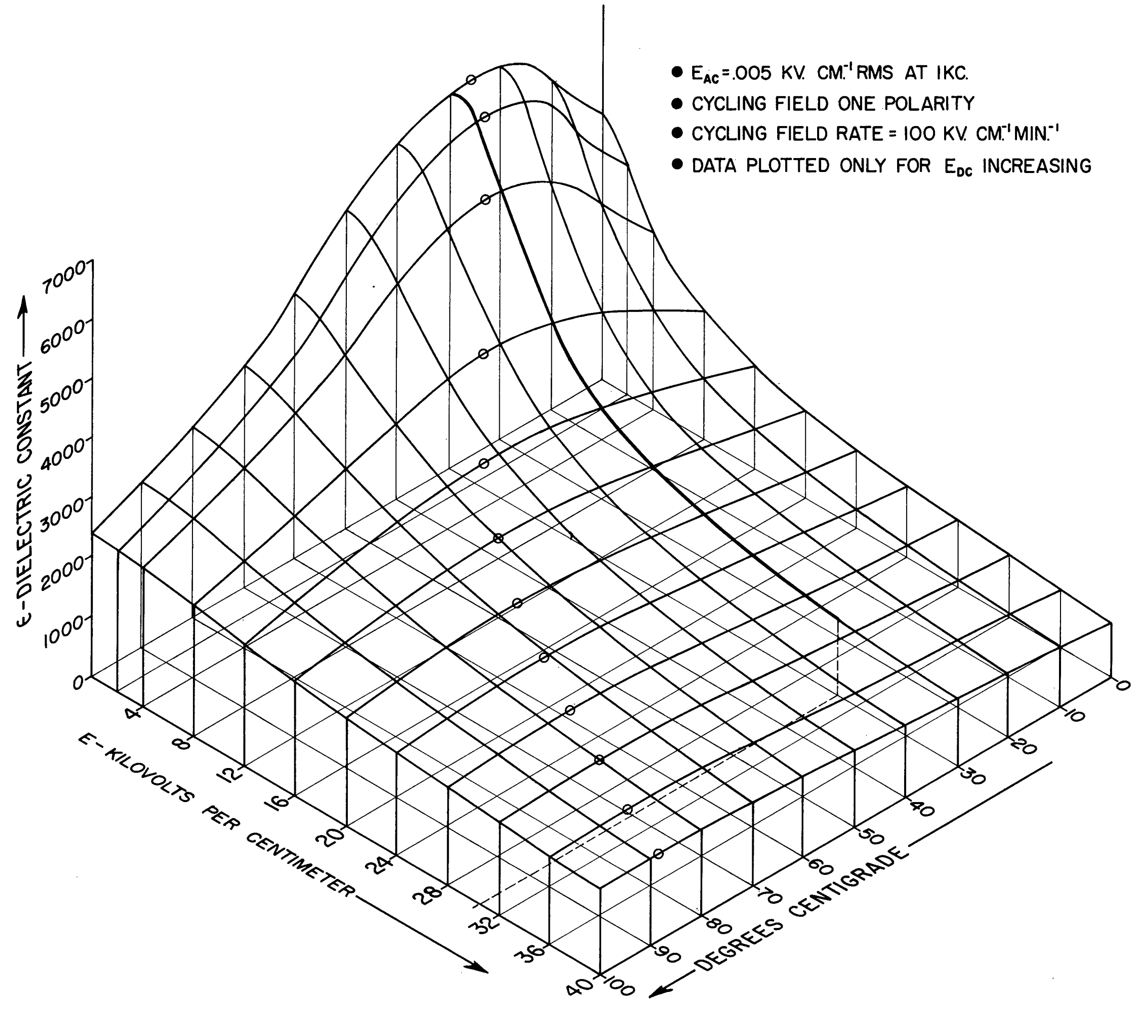


FIG. 10

BUTTERFLY C-E AND P-E LOOPS FOR GLENCO K3300



- $E_{AC}$ = .005 KV. CM.$^{-1}$ RMS AT 1KC.
- CYCLING FIELD ONE POLARITY
- CYCLING FIELD RATE = 100 KV. CM.$^{-1}$ MIN.$^{-1}$
- DATA PLOTTED ONLY FOR $E_{DC}$ INCREASING

FIG. 11

ε-T-E SURFACE
FOR AEROVOX "HI-Q" 40

note that the small signal dielectric constant of many of these materials is essentially constant with frequency up to several hundred megacycles[1]. Therefore, the 1000 cycle data indicated by the chart is useful over a wide range of frequencies.

To avoid confusion, only the curves for electric field <u>increasing</u> were used in drawing the $\epsilon$-T-E surface. However, the difference between the increasing curve and the decreasing curve can be quite small for many materials in the one-sided, or half-butterfly plot.

In taking the data, the temperature was changed in successively increasing steps, thus avoiding discrepancies caused by the thermal aging effect discussed in Section 5.

$\epsilon$-T-E surfaces for 21 types of ferroelectric ceramic materials have been obtained. These data were published in a previous report[2].

## 9. MAGNETIC BUTTERFLY LOOPS

When a magnetic specimen is subject to combined ac and dc magnetic fields, denoted by $\Delta H$ and $H_o$, the material responds with a combined ac and dc flux density, denoted by $\Delta B$ and $B_o$. The incremental[3] permeability $\mu_\Delta$ is defined by the ratio $\Delta B/\Delta H$, and is a function of both $H_o$ and $\Delta H$. The small signal, or reversible permeability, $\mu_r$, is the limit of $\mu_\Delta$ as $\Delta H \rightarrow 0$.

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[1] See, for instance, H. Diamond and L. W. Orr, "Interim Report on Ferroelectric Materials and their Applications," Electronic Defense Group Technical Report No. 31, p. 58, University of Michigan, July, 1954.

[2] L. W. Orr, $\epsilon$-T-E Surfaces of Ferroelectric Ceramics," EDG Technical Report No. 53, University of Michigan, October, 1955.

[3] For a full discussion of kinds of permeability, and typical variation of $\mu_\Delta$ as a function of both $H_o$ and $\Delta H$, see L. W. Orr, Permeability Measurements in Magnetic Ferrites," EDG Technical Report No. 9, University of Michigan, September, 1952.

A magnetic Butterfly loop for a material generally refers to the variation of $\mu_r$ vs $H_o$ when the latter is cycled symmetrically about zero field. Such a loop is useful in determining the small signal behavior of an inductor using the material as its core.

A further distinction in the kind of permeability must be made. Referring only to the reversible permeability, its variation with $H_o$ depends upon the direction of the applied field. If the field is applied in a direction parallel to the ac measuring field, one obtained the "parallel field" Butterfly loop. However, if $H_o$ is applied perpendicular to the direction of the ac field, one obtains quite a different form of butterfly loop.

Using a toroidal magnetic specimen having a suitable winding, both kinds of loops may be obtained with BLARE. The parallel field Butterfly loop is obtained using an arrangement indicated by Fig. 12.

Figure 13 shows the circuit of the variable dc current supply and the method of connecting a ferrite torodial inductor. The supply gives a maximum current of 2 amperes, and the slow variation is obtained by means of a temperature-limited diode (actually, four 5U4 rectifiers). The diode is furnished with a varying heater power by means of the BLARE motor-controlled variac and a suitable step-down transformer. The source of dc current through the diode is furnished by a second power supply shown in the lower part of Fig. 13.

As the dc current is slowly varied through the inductor, a steady 1000 cycle current is also applied. The ac voltage developed across the inductor is proportional to its impedance and is therefore a measure of the permeability of the core. This voltage is fed to the BLARE amplifier and detector, and finally to the pen input of the recorder. The drum input of the recorder is obtained from the voltage drop across the 0.25 ohm resistor. The drum

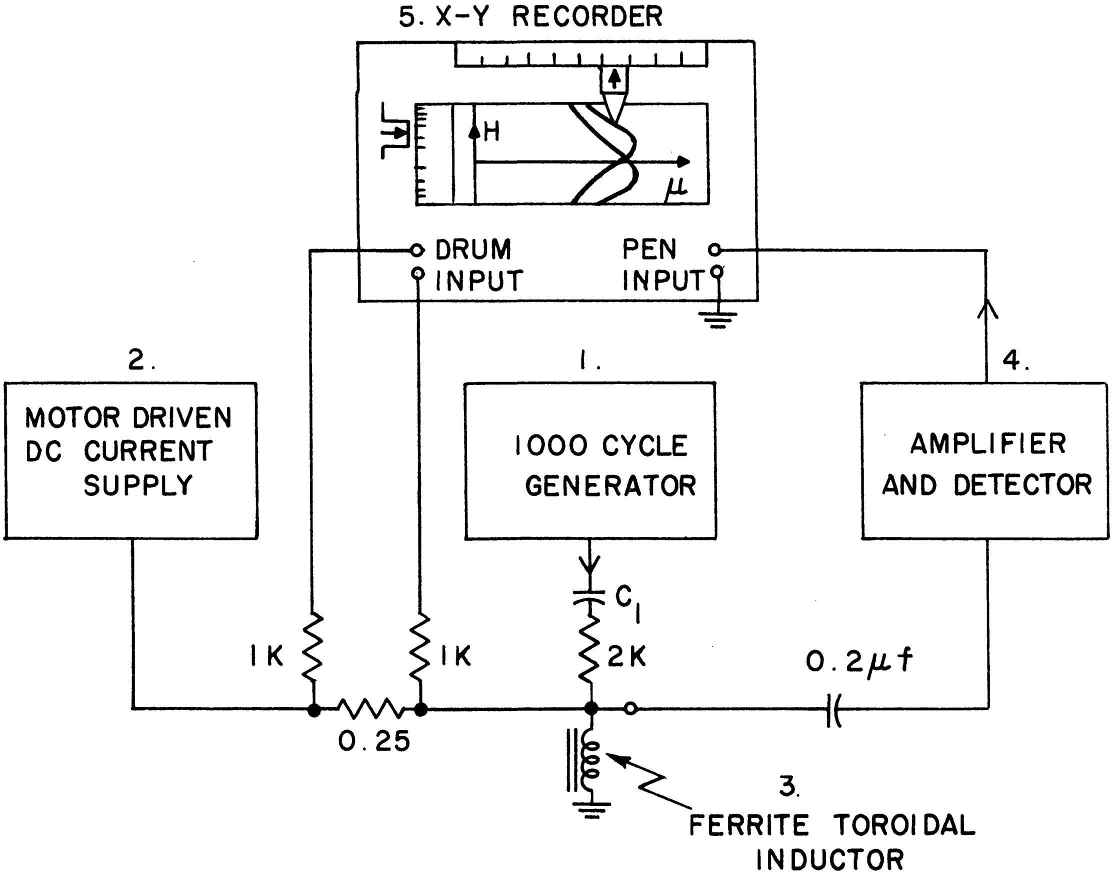

FIG 12

BLOCK DIAGRAM OF BLARE MODIFIED FOR MAGNETIC BUTTERFLY LOOP RECORDING

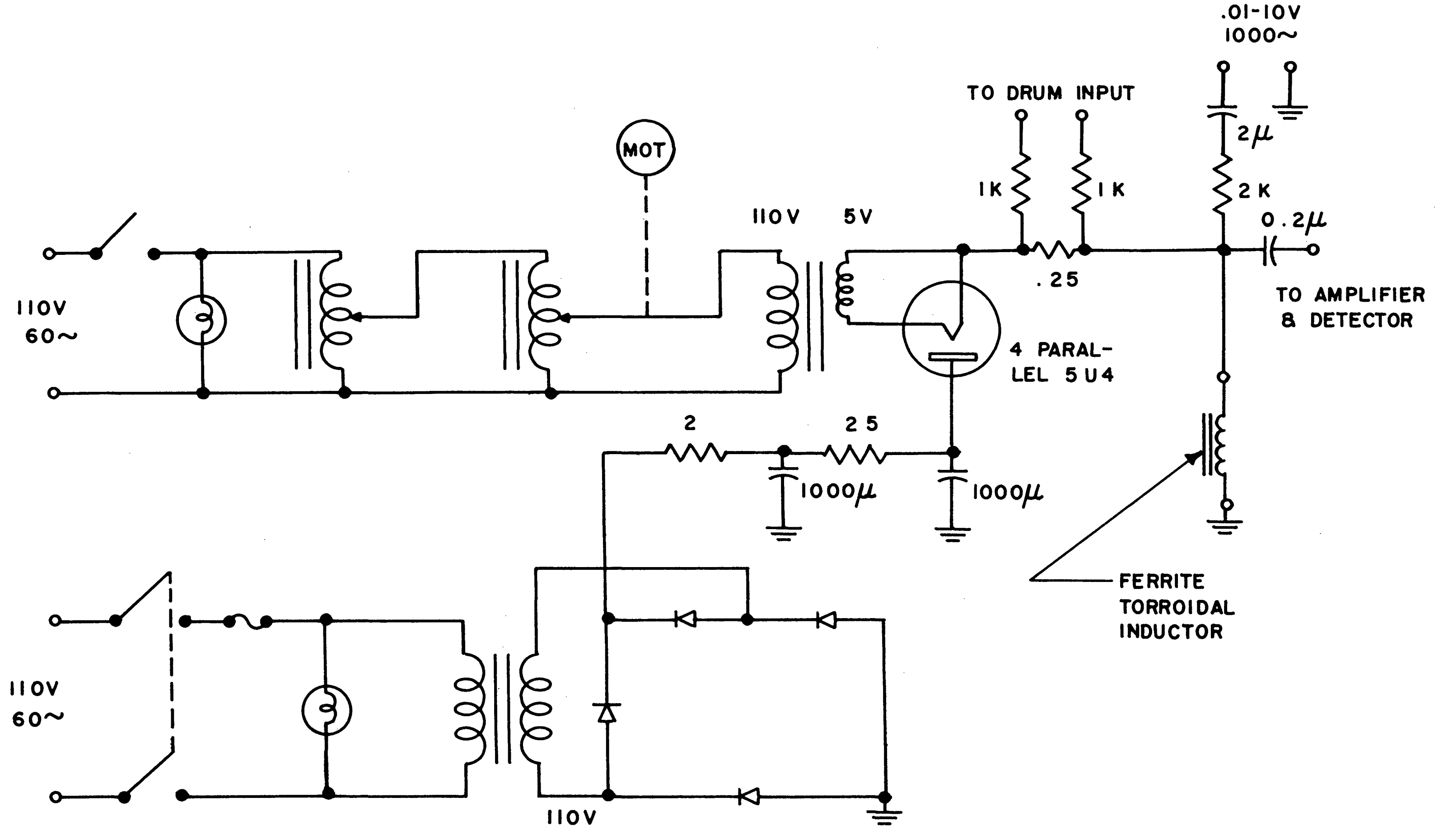

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FIG 13

VARIABLE CURRENT (0-2a) SUPPLY

displacement is therefore proportional to the applied dc field. The two 1K ohm resistors are used for isolation purposes.

When the automatic features of BLARE are employed, one wing of the butterfly loop is obtained. To obtain the second wing of the loop, the inductor leads and the drum input leads are both reversed at times of zero dc current. A typical parallel Butterfly loop for a ferrite toroid is shown in Fig. 14.

To obtain a perpendicular loop, the dc magnetic field, $H_0$, is applied by an external electromagnet in a direction perpendicular to the ac measuring field, $\Delta H$, as shown in Fig. 15.

In this case the variable current source is used to drive the field winding of the electromagnet, while the toroid is placed between the poles as shown, and the impedance of the winding is measured as described previously, using the 1000 cycle drive. To obtain both wings of the loop, the field winding leads and drum input leads are both reversed at times of zero current.

Care must be taken to maintain the plane of the toroid parallel to the pole faces; otherwise the $H_0$ field will not be everywhere perpendicular to the measuring field. Because of the strong field, it is necessary to clamp the toroid core in position.

When proper precautions are made, an impedance variation such as indicated by Fig. 16 is obtained. This is the perpendicular butterfly loop for the same toroid core as in Fig. 14.

It is possible to reduce the impedance data to permeability changes when required. It should be noted, however, that the permeability of most magnetic materials, including ferrites, is not constant but falls off with increasing frequency. Thus magnetic Butterfly loop data taken at 1000 cycles cannot be transposed to rf frequencies with anything like the same assurance as was possible with ferroelectric data.

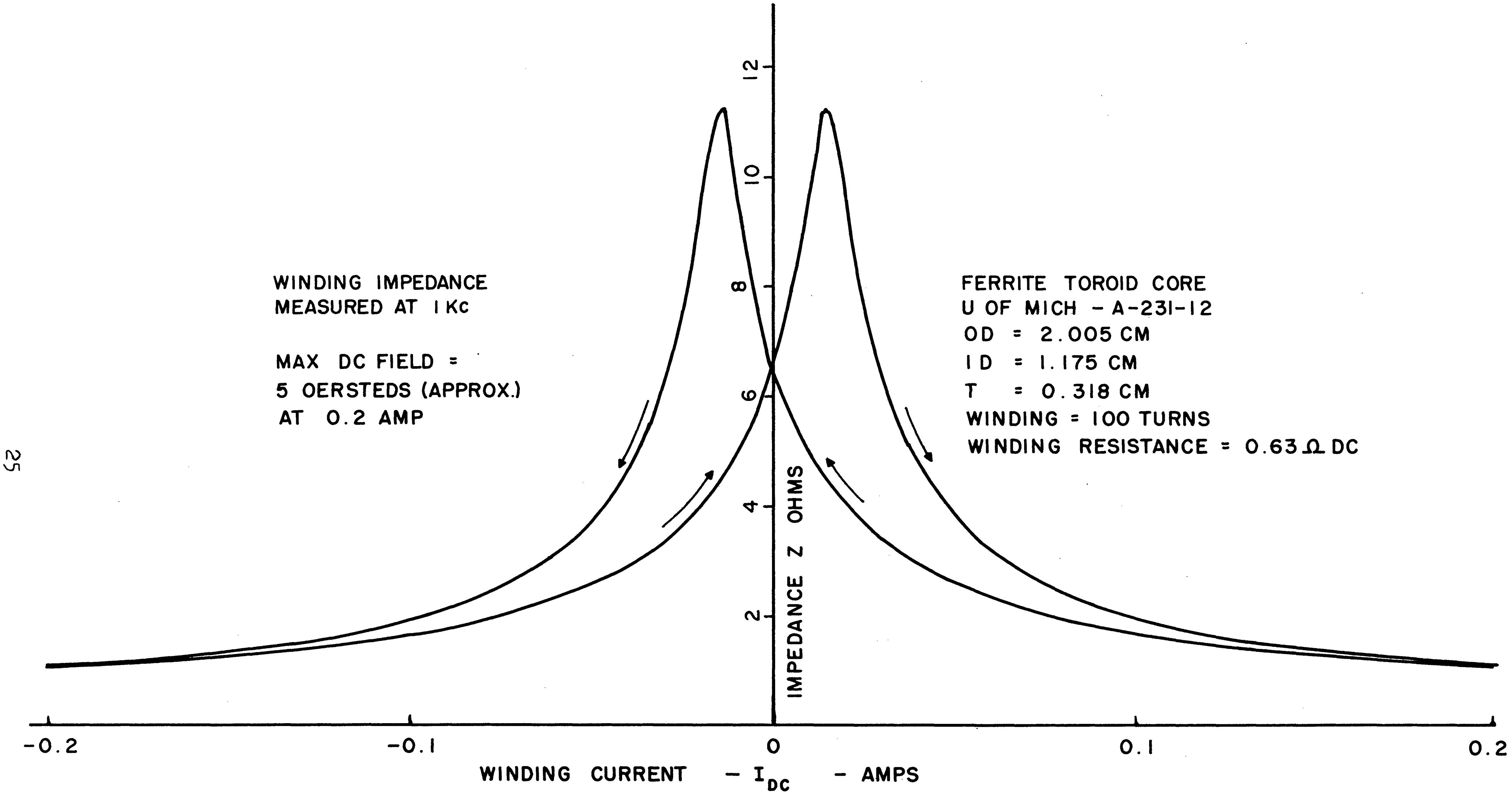

FIG 14

IMPEDANCE BUTTERFLY LOOP - PARALLEL FIELDS

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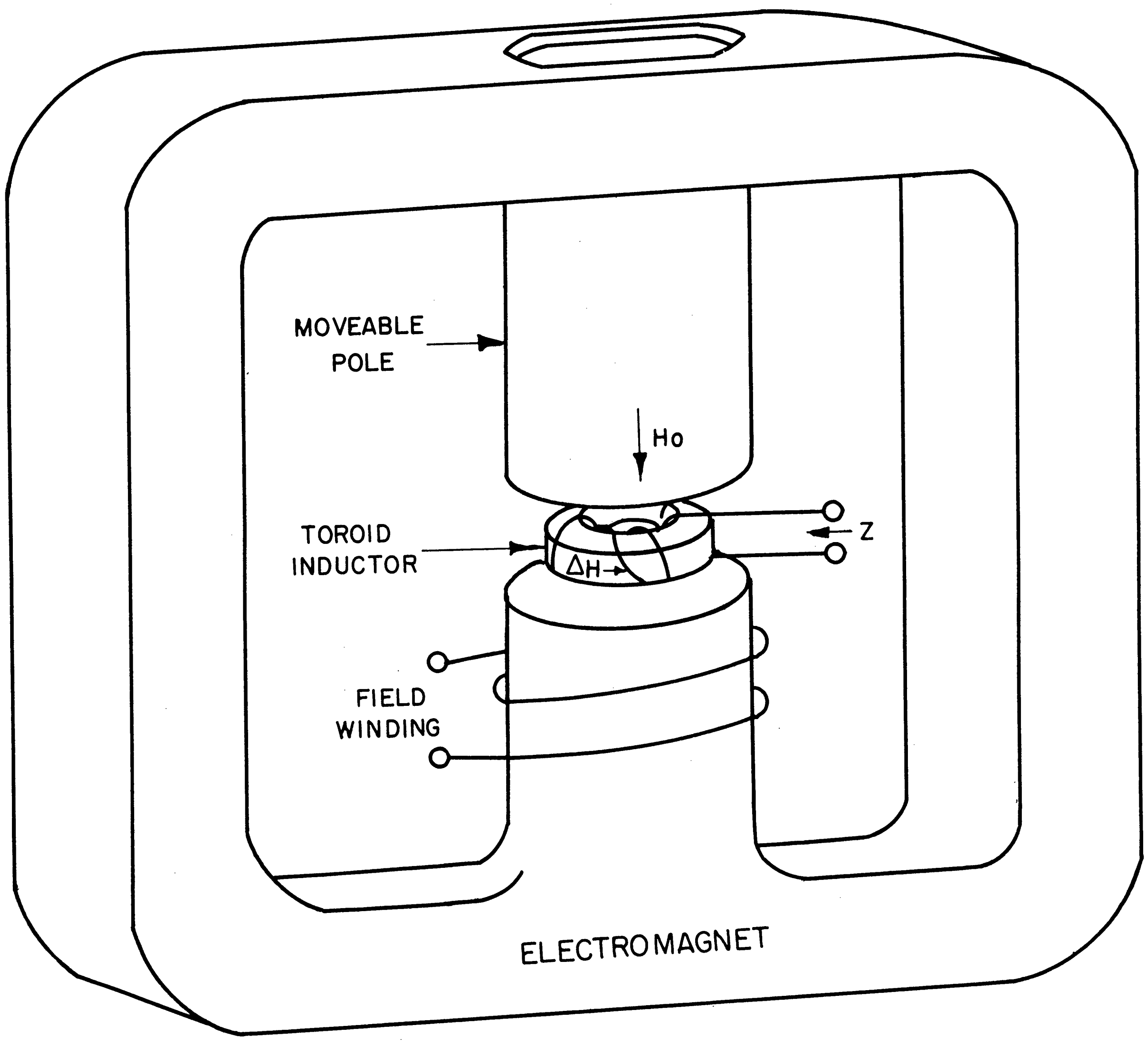
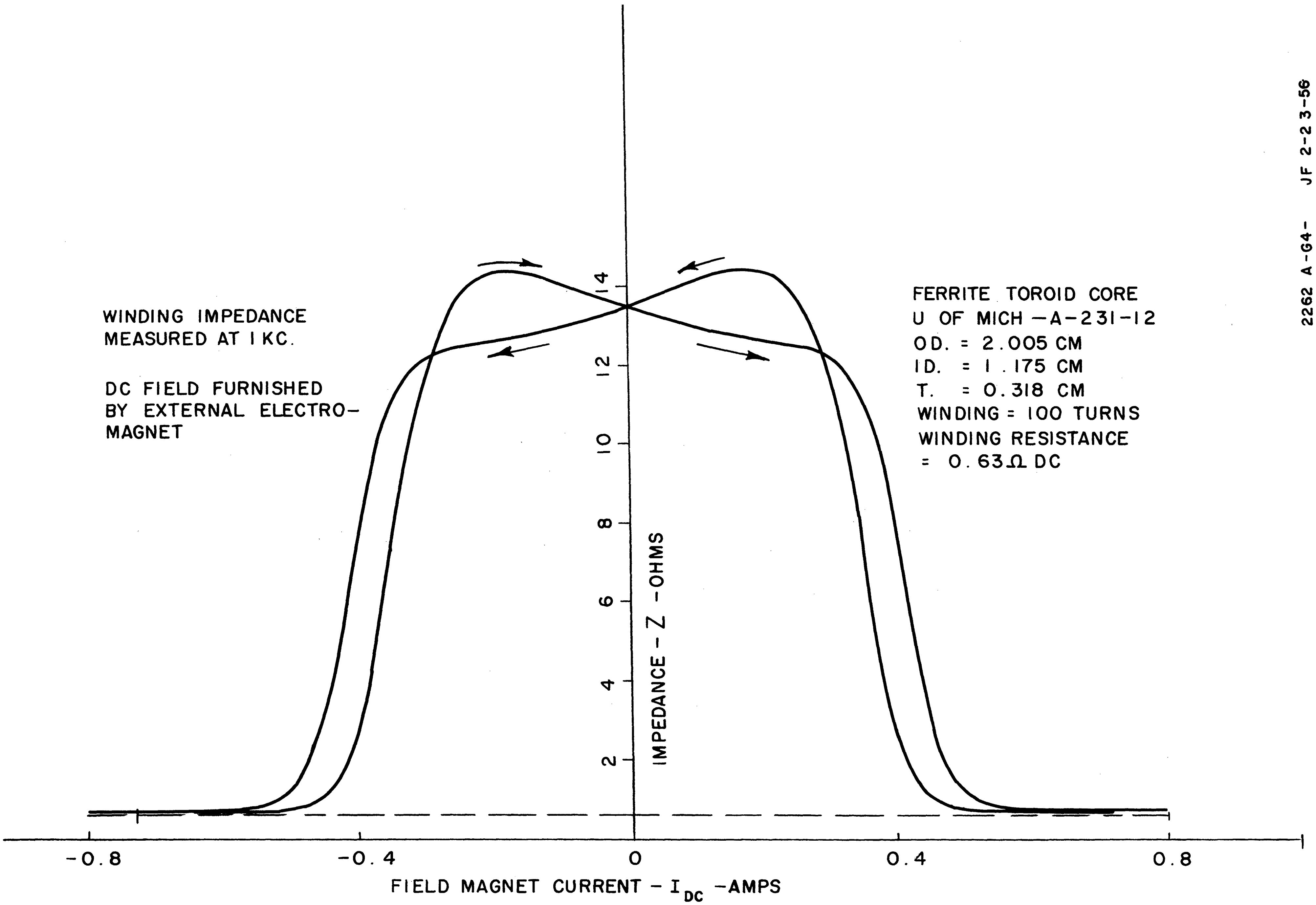


FIG 15 TOROID SPECIMEN ARRANGED FOR PERPENDICULAR MAGNETIC FIELDS



WINDING IMPEDANCE
MEASURED AT I KC.

DC FIELD FURNISHED
BY EXTERNAL ELECTRO-
MAGNET

FERRITE TOROID CORE
U OF MICH – A-231-12
OD. = 2.005 CM
ID. = 1.175 CM
T. = 0.318 CM
WINDING = 100 TURNS
WINDING RESISTANCE
= 0.63Ω DC

FIG 16 IMPEDANCE BUTTERFLY LOOP — PERPENDICULAR FIELDS

2262 A-G4- JF 2-2 3-56

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