Report CCG 71-57, NBS 2210472

NBS IR 74-476 (R)

STABLE PRESSURE TRANSDUCER

J. H. Colwell

Pressure and Vacuum Section
Heat Division
National Bureau of Standards
Washington, D.C. 20234

July 1974

Annual Report for Period July 1973 - June 1974

Prepared for CCG-Army/Navy/AF

Abstract

This report describes the continuing development of a capacitive pressure transducer which utilizes a solid dielectric material. A way of combating the pronounced temperature dependence of this device, based on the use of materials that have the same temperature dependence but different pressure dependences in opposite arms of the measuring bridge, is discussed. It is proposed that particular cuts from anisotropic materials be used to get the proper capacitive properties for the gauge. The thermal relaxation phenomena and pronounced temperature gradients observed in the pressure vessel are described. The design of the capacitance bridge and automating circuitry to be used with the capacitance pressure gauge is presented.

FORM CD-14 (5-10-71) Prescr. by DAO 214-2 — U.S. DEPT. OF COMM.

TRANSMITTAL SLIP

DATE: 8/26/74

TO: W. R. Kelley's Secretary

REF. NO. OR ROOM, BLDG.:

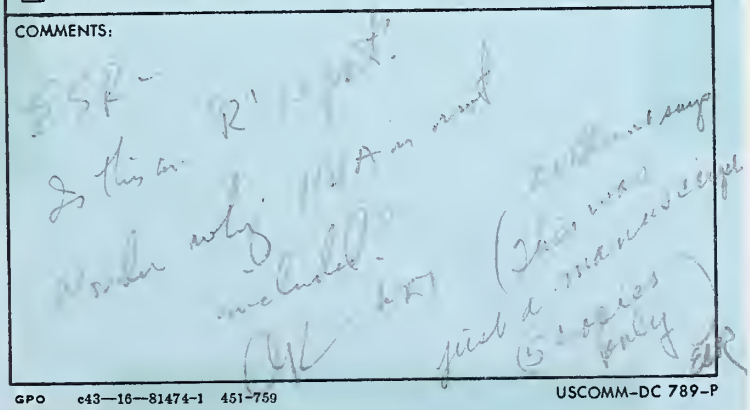
FROM: Dr. Colwell's Secretary

REF. NO. OR ROOM, BLDG.: 221.07

ACTION

- ☐ NOTE AND FILE
- ☐ NOTE AND RETURN TO ME
- ☐ RETURN WITH MORE DETAILS
- ☐ NOTE AND SEE ME ABOUT THIS
- ☐ PLEASE ANSWER
- ☐ PREPARE REPLY FOR MY SIGNATURE
- ☐ TAKE APPROPRIATE ACTION
- ☑ PER OUR CONVERSATION
- ☒ PER YOUR REQUEST
- ☐ FOR YOUR APPROVAL
- ☐ FOR YOUR INFORMATION
- ☐ FOR YOUR COMMENTS
- ☐ SIGNATURE
- ☐ INVESTIGATE AND REPORT

COMMENTS:



GPO c43—16—81474-1 451-759 — USCOMM-DC 789-P

INTRODUCTION

The objective of this project is to develop a pressure transducer which can be used as a transfer standard in disseminating the absolute pressure values determined by controlled clearance piston gauges in standards laboratories. We are currently interested in pressures up to 70 or 140 MPa (10,000 or 20,000 psi), a range where the stated accuracy of the piston gauges is of the order of $1:10^4$. The pressure transducer should therefore be capable of precision in excess of $1:10^4$ over this range but another requirement of a transfer standard is that it will be able to maintain its calibration for an extended period of time while in constant use. The latter requirement has led to the idea of a capacitance gauge using a solid dielectric capacitor. Such a device using a hard, and preferably crystalline, dielectric material with electrodes deposited directly on its surface would not be expected to suffer any perminent deformation with repeated cycling under hydrostatic pressures and would therefore have the required long term stability. Andeen, Fontanella, and Schuele have demonstrated the feasibility of such a gauge using single crystals of CaF_2 as a dielectric medium. With this gauge capacitance changes of about $3:10^7$ must be resolved to determine pressures to a precision of $1:10^4$ at 70 MPa. Using the three terminal method, modern A.C. bridge techniques and accurate ratio transformers, capacitance measurements to $1:10^7$ are readily achievable. Tests have not indicated any instabilities in the capacitors, so a gauge using CaF_2 crystals does meet the basic requirements. The capacitance of the CaF_2 gauge is, however, strongly temperature dependent; 1 mK changes the capacitance by $3:10^7$, the required resolution of the gauge. It is possible to thermostat the gauge within the 1 mK range but it is not convenient and only transfers the long term stability requirements to the thermostating thermometry.

The problem is compounded by the considerable amount of heat which is generated in the pressure vessel whenever the pressure is changed thus requiring long periods to reestablish thermal equilibrium. A search has been made for materials having a smaller temperature to pressure dependence of capacitance but no dramatic improvements have been found. The search is continuing in this laboratory but the nature of the measurements make this process tedious. As an expedient, toward getting a working device, other ways of overcoming the temperature dependence of the capacitance measurement are being sought. The bridge method required for the capacitance measurements offers other ways of cancelling out the temperature dependence of the pressure determination. One technique is to put capacitors of the same material in opposite sides of the bridge so that their capacitances are cancelling each other. If one capacitor is pressurized and the other kept at ambient pressure while keeping the two in thermal contact, the temperature dependence will cancel to first order and any additional thermostating required should be trivial. James Miller of the U.S. Army Missile Command is pursuing developments along this line. The principle difficulty of this method is that it requires establishing thermal equilibrium between a sample within a pressure vessel and one outside which can require a considerable length of time. In our laboratory we are currently concentrating on a variation of this same general idea. We are attempting to find two materials which will have the same temperature dependence of capacitance yet having considerably different pressure dependencies. Again, the capacitors would be placed in opposite arms of the bridge but both would be inside the pressure vessel. In this way, the temperature dependence of the device would cancel as above but the two capacitors could be in intimate thermal contact, thereby reducing the thermal equilibation time between them. To utilize this technique we are now faced with the problem of finding the

two materials with the proper capacitive properties. Since several anisotropic materials have considerably different temperature dependence along the different crystalline axes we are hoping that we will be able to match two samples by cutting a sample from the anisotropic crystal at an intermediate angle between the major axes.

Toward this end we have begun collecting data on anisotropic material and are continuing to look at various other materials that may be of possible use in the device. Several experiments have been carried out to gain knowledge about the thermal equilibration problems within pressure vessels. Work on the prototype capacitance bridge for use with a capacitive pressure transducer is described.

SAMPLE ELECTRODES AND SAMPLE HOLDERS

The deposition of electrodes on the dielectric samples has been a problem throughout this work both from the standpoint of forming the insultaing gaps for the guard electrodes and because of poor adherence of the electrodes to the sample. For several months we have been using vacuum deposited aluminum coatings exclusively. Samples are cleaned by washing with soap and water, rinsing with alcohol and freon, and finally ion bombarding in the evaporator for upwards to 1 hour prior to coating. We have started using 99.999% aluminum wire as our source material. This wire outgasses very little on melting so that a good vacuum can be maintained throughout the coating process which probably accounts for the excellent mirror-like coatings obtained. All coatings are checked by pulling with Scotch tape and very few have failed the test.

We are continuing to form the electrode gaps by masking the samples with wedge-shaped steel rings during the deposition process. Magnets behind the sample support in the evaporator are used to hold both the sample and masking

ring in place. We are using hardened steel for the masking rings which makes them much less susceptible to damage than the mild steel rings formerly used. In addition it is possible to lap a much finer edge on the hardened steel rings so that we have been able to consistently achieve gaps as narrow as 8 μm (0.0003").

One drawback in using masking rings with a fixed source and a fixed sample is that there is invariably some area of the sample that is shadowed sufficiently by the ring that the coating is quite thin. We have overcome this problem by adapting the evaporator so that the sample is rotated about its axis during the evaporation process. The evaporator filament is positioned so that it is at a 45° angle to the sample axis, in this way all areas in the vicinity of the ring get a uniform coating. A disadvantage of this procedure is that only one sample can be coated at a time thus greatly increasing the sample preparation time.

The sample holders for mounting the dielectric samples in the pressure vessel have been modified so that electrical contacts are made with small gold-plated nickel deposited bellows, see Fig. 1. The bellows are 1/8" diameter and 3/16" long and can be compressed up to 0.070". The advantage in using the bellows is that their spring constants are very consistant (2.3g/0.001") so that the force on the contacts can be predetermined by adjusting the amount the bellows will be compressed when the sample holder is assembled. This adjustment is made by varying the thickness of the Teflon washers which insulate the contacts from the body of the sample holder. The bellows of the high voltage contact is usually compressed about 0.040" (92 g load) and that of low voltage contact 0.020" (46 g load). In this way there is sufficient load (∿ 46 g) on the sample to ensure that good electrical contact between the guard ring of the sample and the grounded case of the sample holder

is maintained. With this arrangement we have had no trouble with contacts opening during pressure cycling.

SAMPLE SELECTION

In our continuing survey of the temperature and pressure dependence of capacitance of various materials we have been concentrating on materials that are mechanically hard and chemically inert, have a low dielectric loss component, and are available in an appropriate form. An additional qualification to the last condition must be a reasonable cost, which does keep the number of materials to be tested within bounds. In Table 1 we have tabulated the temperature and pressure dependence of the capacitance of several materials for which data are available. The third column gives the ratio of the temperature to pressure dependence which, for our purpose, we may consider as a quality factor. As can be seen, CaF_2, which we consider our benchmark material, was not a poor initial choice. The ratio for X-cut quartz is much better, but the magnitude of the pressure dependence of this material is so small that the measurement sensitivity will be marginal. Pyrolytic boron nitride is by far the best of any material studied and is still considered for possible use, but this material has a layered structure and the surface is rather easily damaged. We have put several anisotropic materials on the list to show the range that their temperature and pressure dependences cover. From this list there are not many combinations of materials that could be used in the opposite arms of the capacitance bridge to achieve cancellation of the temperature dependence. The one attractive pair is the a-cut paratellurite (TeO_2) and an intermediate cut, probably about a 45°, of sapphire (Al_2O_3). This combination would have a pressure sensitivity approximately equal to CaF_2. To look for other combinations we are preparing

Table 1

The Temperature and Pressure Dependence of the Capacitance of Select Materials

Material	$\frac{\partial \ln C}{\partial T}_P \times 10^6/K$	$\frac{\partial \ln C}{\partial P}_T$ $10^{12}/Pa$	$\frac{(\partial C/\partial T)}{(\partial C/\partial P)} \frac{MPa}{K}$
CaF_2	263	- 38	- 6.9
BaF_2	231	- 50	- 4.6
NaCl	356	-118	- 3.0
TlBr	-330	-218	1.51
BN	- 52	123	- 0.42
$KTaO_3$	-2700	-182	15.0
$SrTiO_3$	-3820	-248	15.4
Quartz-fused	11	4.6	2.4
Quartz-Z-cut	30	- 1.7	-17.6
Quartz-X-cut	10	9.5	1.05
Al_2O_3-a	110	- 10	-11
Al_2O_3-c	190	- 11	-17
TiO_2-a	-650	- 35	18.5
TiO_2-c	-990	- 72	13.8
TeO_2-a	156	29	5.4
TeO_2-c	244	- 49	- 5.0
MgO	111	- 20	5.5
$CaCO_3$-a	(400)		
$CaCO_3$-c	(400)		

to look at MgF_2 and $CaCO_3$, and to reexamine Quartz, Al_2O_3, and TeO_2. We are going to measure isotropic materials such as, gadolinium gallium garnet, magnesium aluminate spinel, and several glasses to try to find other cases where the temperature dependence of the material will fit within the range of an anisotropic material.

We have repeated, with new samples, the earlier measurements on quartz made in this laboratory. The equilibrium temperature and pressure dependences measured were close to the earlier values, but a strange relaxation phenomenon was observed. When the pressure was changed and the thermal equilibrium had been reestablished in the pressure vessel, there still occurred a very long relaxation process which required up to 24 hours before equilibrium was reached. In changing the pressure between 0 and 70 MPa, the total relaxation was about 40% of the total-initial change and always back toward its previous equilibrium value. The effect was largest in the Z-cut crystal, it was observed in 45°-cuts containing the Z-axis, but was not noticeable in X or Y cut crystals. It is thought that the phenomenon may be caused by the piezoelectric properties of quartz, possibly due to charge generation on the surface of the sample when it is pressurized. Tests, however, such as shorting the two sides of the capacitor had no effect on measured capacitance. We have no explanation of this phenomenon but the experience makes one wary of piezoelectric materials.

THERMAL EQUILIBRATION IN THE PRESSURE VESSEL

In the introduction we mentioned some of the thermal equilibration problems encountered in the pressure vessel. Here we describe some observations made on our system—some were by design, others incidental to other measurements. In Fig. 2 is pictured the group of five sample holders that

are usually used in a pressure run. They are mounted on the pressure vessel closure through which the electrical leads are brought. This assembly fits inside a pressure vessel which is 1 1/2" I.D. and 3 5/8" O.D. The well in the vessel is 15" deep so that it is only half filled by the sample assembly; the lower half has been filled with a brass cylinder to reduce the oil volume in the system. In operation the pressure vessel is immersed in a thermostated water bath.

It is interesting, first of all, to calculate the temperature changes which occur in the system when it is pressurized. If we had an instantaneous, i.e. adiabatic, compression of the system to 70 MPa (10,000 psi) the oil in the system would undergo the largest change, warming some 7 to 10 K. The brass sample holders (and brass cylinder) would warm about 0.4 K. The steel vessel uniformly cools about 0.09 K under the combined influence of hoop and longitudinal stretching and radial compression. The sample itself undergoes a temperature change which would be superimposed in the observed capacitance change due to the pressure increase—in CaF_2 the instantaneous temperature increase would be 1.15 K.

It is possible to solve the expressions for the thermal relaxation of such a system but hardly practical. We can write an approximate relationship for the relaxation which should be useful for discussion purposes,

$$\log \frac{T - T_e}{T_o - T_e} = X \frac{\lambda A}{CV\rho\ell} t + Y. \qquad (1)$$

In this expression T is the temperature at time t, T_e and T_o are the equilibrium and arbitrary initial temperatures. X and Y are constants which depend on the geometry. λ, A, and ℓ are the thermal conductivity, cross sectional areas, and

conductance path length of the component of the system which impedes conduction the most, whereas C, V, and ρ are the specific heat, volume, and density of the component with the largest heat capacity.

Upon compression, the oil in the system produces about an order of magnitude more heat than the solid components and is also the poorest conductor so it is obvious from the start that its volume should be minimized. It is also apparent that for any given geometry the relaxation time will increase as the square of the overall size.

In our apparatus, pressurizing to 70 MPa (in approximately 1 minute) raises the temperature of the samples between 1 and 2 K. The system takes about 45 minutes to return to equilibrium (i.e to within 1 mK of the thermostated temperature) showing a relaxation time of 5-6 minutes. With the brass cylinder in the pressure vessel the amount of heat produced is cut in half but the relaxation time is slightly longer so that it takes the same length of time to reach equilibrium.

We are currently interested in how quickly two samples within the pressure vessel will equilibrate, two experiments were carried out for this purpose. In the first, two CaF_2 samples, each mounted in individual sample holders, were placed adjacent to each other in the pressure vessel assembly. The sample holders were firmly bolted together by nuts on the screw rod supports so they would normally be considered to be in intimate thermal contact. The experiments were run with the CaF_2 samples in opposite arms of the capacitance bridge so that the capacitance difference was observed. Upon pressurizing the system to 70 MPa large temperature gradients were set up between the two cells. The capacitance change reached $\sim$ 80 ppm (equivalent to a temperature difference of 0.3 K) and took 35 minutes to relax to the equivalent of 1 mK difference between the samples. On

decompression the behavior was different in one respect, the maximum displacement (in the opposite direction) was only 40 ppm but it relaxed back through zero displacement and approached equilibrium from the opposite side. The total equilibration time was again 35 minutes. Heating the pressure vessel via the thermostating water bath at a rate of 0.5K/min produced a steady state capacitive difference of 12 ppm (equivalent to 50 mK).

In an attempt to minimize the temperature gradients observed in the above system, the double sample holder shown in Fig. 3 was fabricated and the same set of experiments were carried out. The results showed a large improvement but not as much as one would expect. On compression to 70 MPa the capacitance difference reached 3.5 ppm (14 mK) but took 15 minutes to return within 1 mK of equilibrium. On decompression (the displacement was in the same direction) the difference reached 3 ppm and took 9 minutes to return to equilibrium. Upon heating at 1/2K/min the displacement reached 0.8 ppm.

It is obvious from these experiments that temperature gradients within a pressure cell can be very pronounced. In order to bring two samples into thermal equilibrium we can improve their thermal contact. Two samples in the bridge network could be physically joined at their common low voltage contact. Another approach would be to make the environment within the pressure cell symmetrical, so that the same gradients exist at both samples.

One result of these two experiments was a demonstration of the precision possible with the capacitance gauge. In comparing the two capacitors after thermal equilibration was attained, the ratio of capacitance was the same to within our limit of detection, $5{:}10^8$, for pressures from 0 to 140 MPa. This was true in both experiments although there was a difference of 3 ppm in the capacitance ratio between the two experiments. In both experiments there was a change in the ratio of about 0.8 ppm on changing the temperature from 20 to 40 °C.

AUTOMATIC CAPACITANCE BRIDGE

A limited-range capacitance bridge is being constructed which was designed specifically for use with the dielectric capacitance pressure gauge. It will be capable of measuring the ratio of two capacitors which differ by less than 10% and which also have reasonably small loss tangents. After an initial adjustment, using a phase compensated resistor, the bridge is intended to be completely automatic.

The design of the bridge proper, shown in Fig. 4, is in principle the same as that presently being used as a research tool in our laboratory. The bridge transformer forms the 1:1 ratio arm of the bridge and supplies the voltage for the ratio transformer used in balancing the bridge. The output voltage of the ratio transformer is injected into one of the ratio arms of the bridge through the two stage transformer. The power for this injected voltage comes from an operational amplifier so that there is no loading of the ratio transformer and the bridge impedance is kept small. It is the turns ratio on the two stage transformer and the voltage supplied to the ratio transformer that limits the range of capacitor ratios that can be measured. A switch on the primary of the two stage transformer allows the selection of a total range of either 1 or 10%. The switch on the input to the ratio transformer allows this range of bridge ratios to be taken on either the positive or negative sides or centered about the ratio of 1. Any difference in loss between the measuring capacitor and the reference capacitor is compensated by injecting an in-phase, i.e. resistive component, signal into the detector arm of the bridge. The magnitude of this signal is controlled by multiplying the D.C. in-phase signal of the phase sensitive detector by an A.C. signal from the bridge transformer.

The exact phase alignment of the bridge will be manually adjusted by balancing out the signal from a pure resistance.

Fig. 5 is a block diagram of the logic circuitry as presently planned for the automation of the capacitance bridge. The design is limited to some extent by the incorporation of a 5 decade programmable ratio transformer (PRT). We require a 7 digit reading and would desire 8 digits, so it is necessary to limit the range of capacitance ratios that the instrument can cover. In the 1% range the first two significant digits are preset by the range control, in the 10% range only the first significant digit is preset. The next five significant digits are determined by the ratio transformer and two additional digits are obtained by the digital conversion of the analog voltage resulting from any remaining bridge imbalance.

In operation the imbalance current from the capacitance bridge is amplified by an FET preamplifier and by three programmable gain amplifier sections. These are followed by two lock-in amplifiers, one phased to detect the resistive loss component of the bridge and the other phased at 90° to detect the capacitive imbalance. The rectified loss component is further filtered and used to control the automatic loss feedback network in the bridge. This signal can be monitored by a front panel meter.

The rectified capacitance imbalance signal is fed to the gain control module and to two voltage-to-frequency converters (VFC). The signal is inverted before one of the VFC's, so that one VFC delivers a pulse rate proportional to positive signals and the other to negative signals.

The output of the two VFC's are fed into a bank of 14 up-down counters by way of a multiplex unit that switches in and out various counters. After each period of counting the contents of the counters are dumped into latches with

hold that count while the next is in progress. The signals from the latches are routed through buffers to the BCD output for external processing and are routed to the seven-segment decoders which generate a 10 digit LED display. The signal from latches 4', 5, 6, 7, and 8 are routed through drivers to the programmable ratio transformer and provide the updated bridge setting. Only decades 9 through 13 are zeroed before each count so that the new count is added to or subtracted from the existing transformer setting.

The clock steps down the bridge frequency to approximately 10 second and 1 second intervals which are used as counting times. The clock is used to turn the counters on and off, and while the counters are off enables the gain control and range control, transfers information from the decade counter to the latches, zeros counters 9 through 13 and turns on the counters for the next interval.

The gain control looks at the output of the bridge and sets the amplifier gain decades to keep the input to the VFC's less than 10 volts but greater than 1/2 volt. It is operative only when the counters are off. When the amplifier gain changes the counting gain or count time must change to keep the overall gain constant. The counter gain is changed by the multiplexers switching between the last five decade counters. On the highest amplifier gain the clock is set for 10 second counts; on all other gains it is set for 1 second counts.

The range control switches between the following deviation capabilities of the bridge: 0 to 1%, $\pm$ 1/2%, -1 to 0%, 0 to 10%, $\pm$ 5%, and -10 to 0%. The switching is done automatically seeking the highest sensitivity, but can be set manually. Range control switching takes place only when the counter is off and only when the programmable ratio transformer is asked to go beyond its limit (as detected by a carry of borrow signal from counter 4'). Counters 1 2, 3, 4, 4' are reset each time a range control switching occurs.

Decade counters 9 and 10 do not control the bridge. They are zeroed before each count and provide the two analog decades of bridge sensitivity beyond the ratio transformer setting.

The fourth decade is split, 4 going to the readout and 4' controlling the PRT. This was necessary to permit the use of the bridge ranges $\pm$ 1/2% and $\pm$ 5%. On these ranges, when the bridge ratio (and readout) is exactly 1.000, the PRT must be set to 50000, the 5 appearing in the fourth readout decade.

The precision desired from this bridge, $1{:}10^7$, is pushing the limits of the programmable ratio transformer. These devices are frequency dependent, the ratios depending to some extent on winding impedance, core size, shunt capacitance, etc. The ratio transformer was designed for 400 Hz operation but we would like to work at a higher frequency and had intended to use 1592 Hz ($\omega = 10^4$), the frequency of our research bridge. We have had the NBS Electrical Measurements Section determine the accuracy of the ratio transformer as a function of frequency to establish the size of the errors involved; the results are shown in Fig. 6. Ratio settings of 0.2, 0.5, and 0.8 are plotted versus frequency squared. Theoretically, errors in ratio settings reach maxima at the 0.2 and 0.8 settings plotted in the figure.

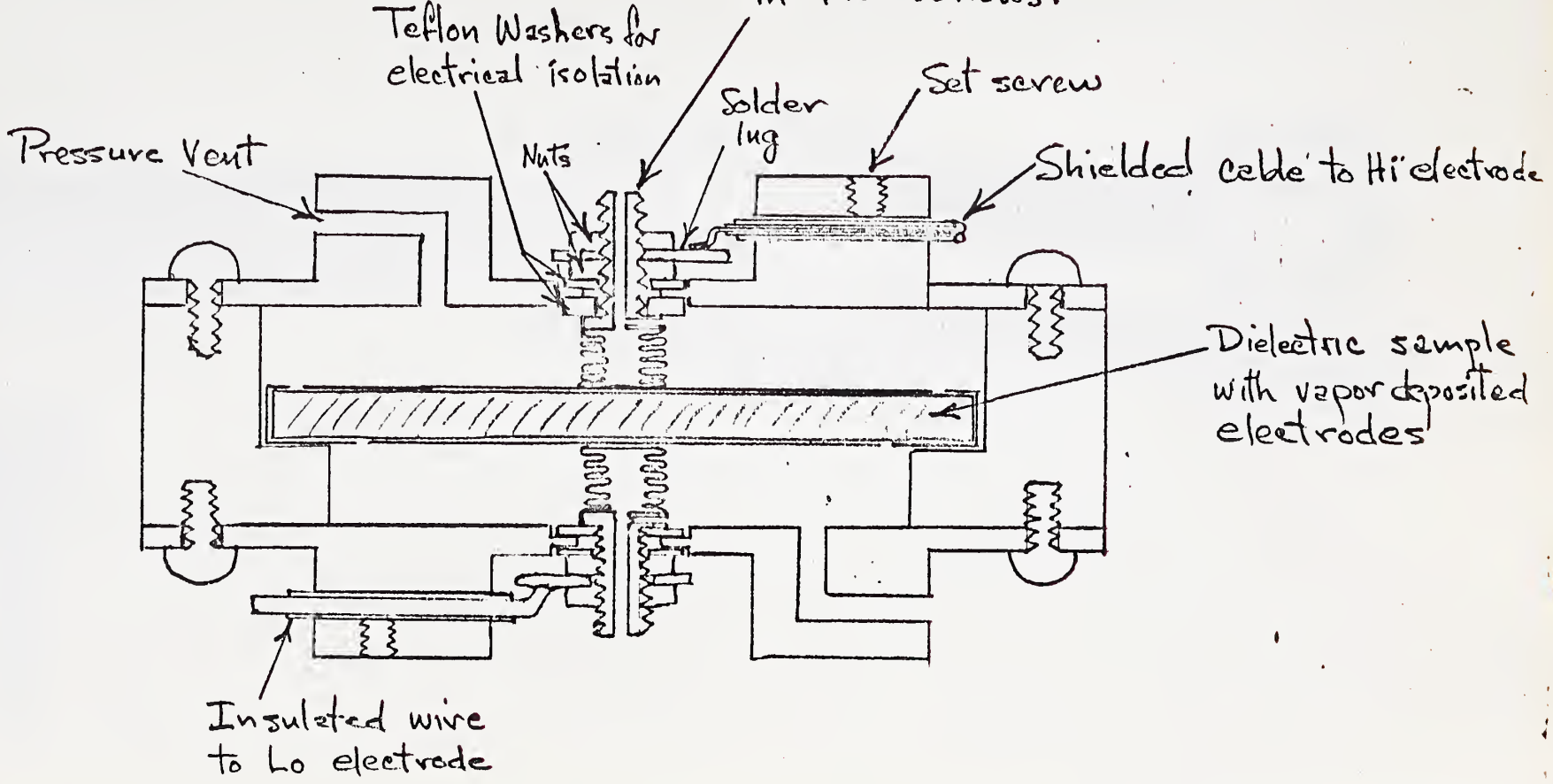
We have decided to operate the bridge at 1,000 Hz where the accuracy of all ratios is within $2{:}10^7$, and, in addition, the ratios all err in the same way except for the two lowest settings. As a result, bridge precision of $1{:}10^7$ should be achievable.

The design of all components of the capacitance bridge has been completed and nearly all the hardware for its construction has been assembled. The toroidal transformers have been wound. The oscillator has been constructed and masks for the printed circuit boards of the other components are being laid out.

Fig-1

Sample Holder

Electrical contacts consist of small nickel deposited bellows which are soldered to 4-40 screw studs. Holes through the screw studs allow pressure equalization in the bellows.



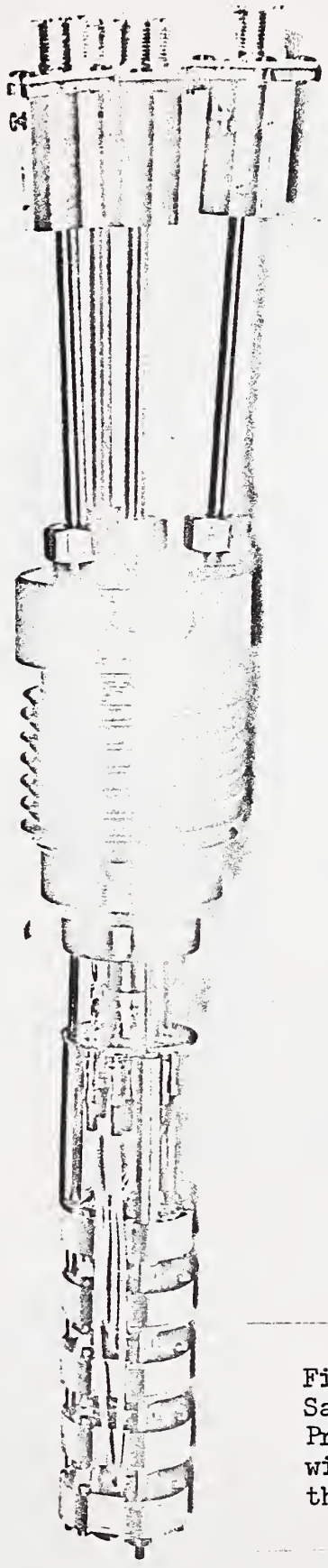


Fig. 2
Sample Holders and
Pressure Vessel Closure
with Electrical Feed-
throughs.

Fig. 3

Double Sample Holder

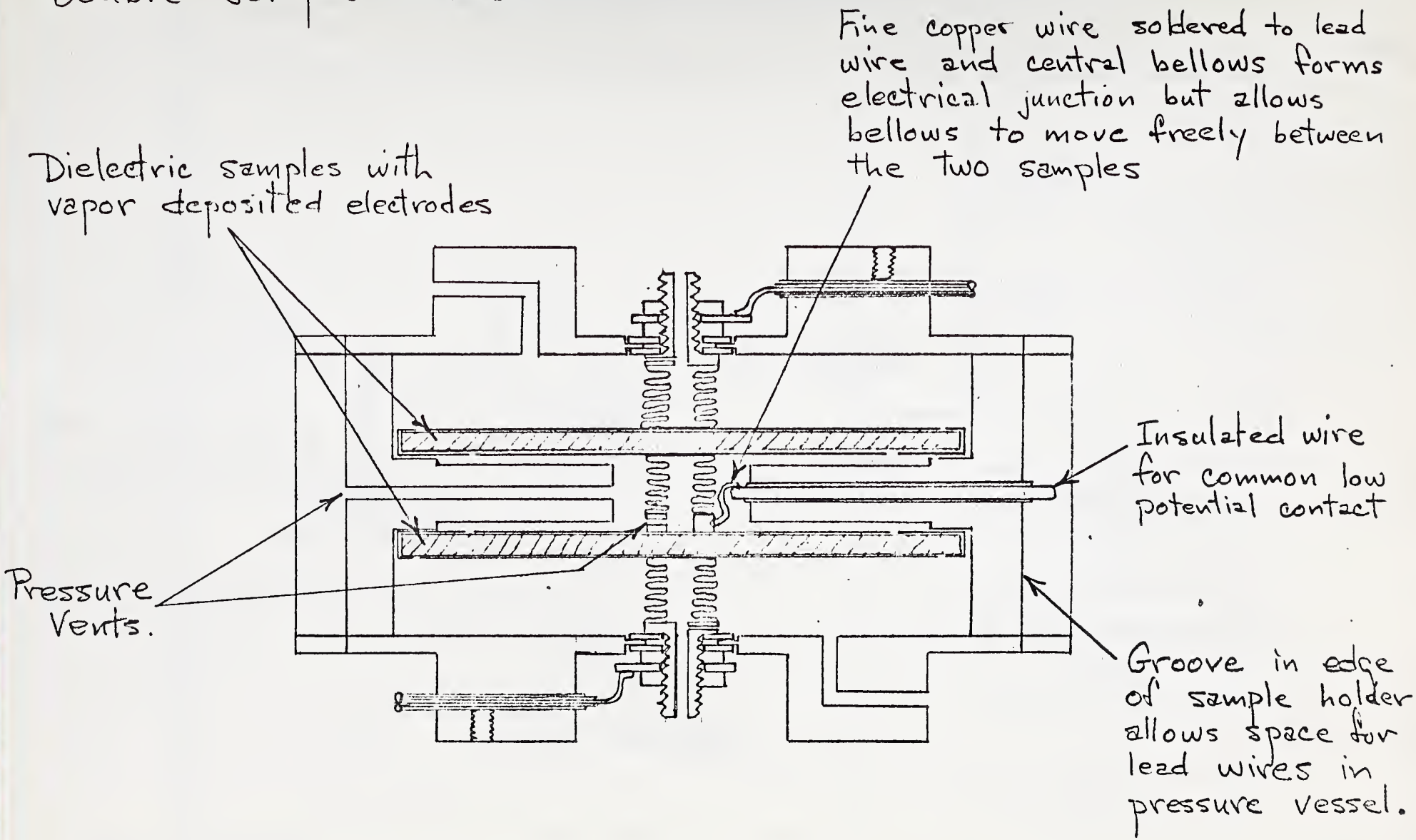


Fig 4

CAPACITANCE BRIDGE

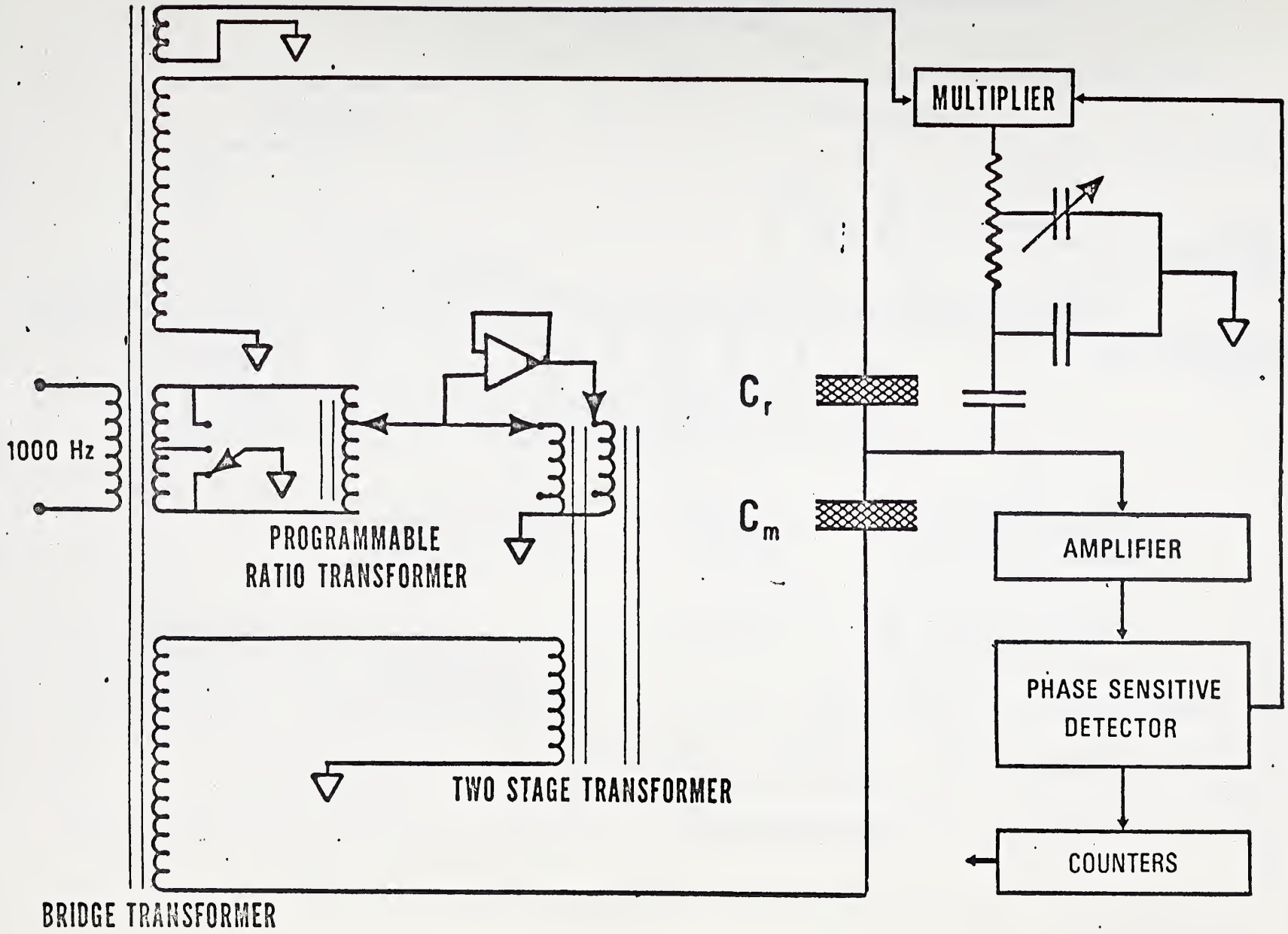
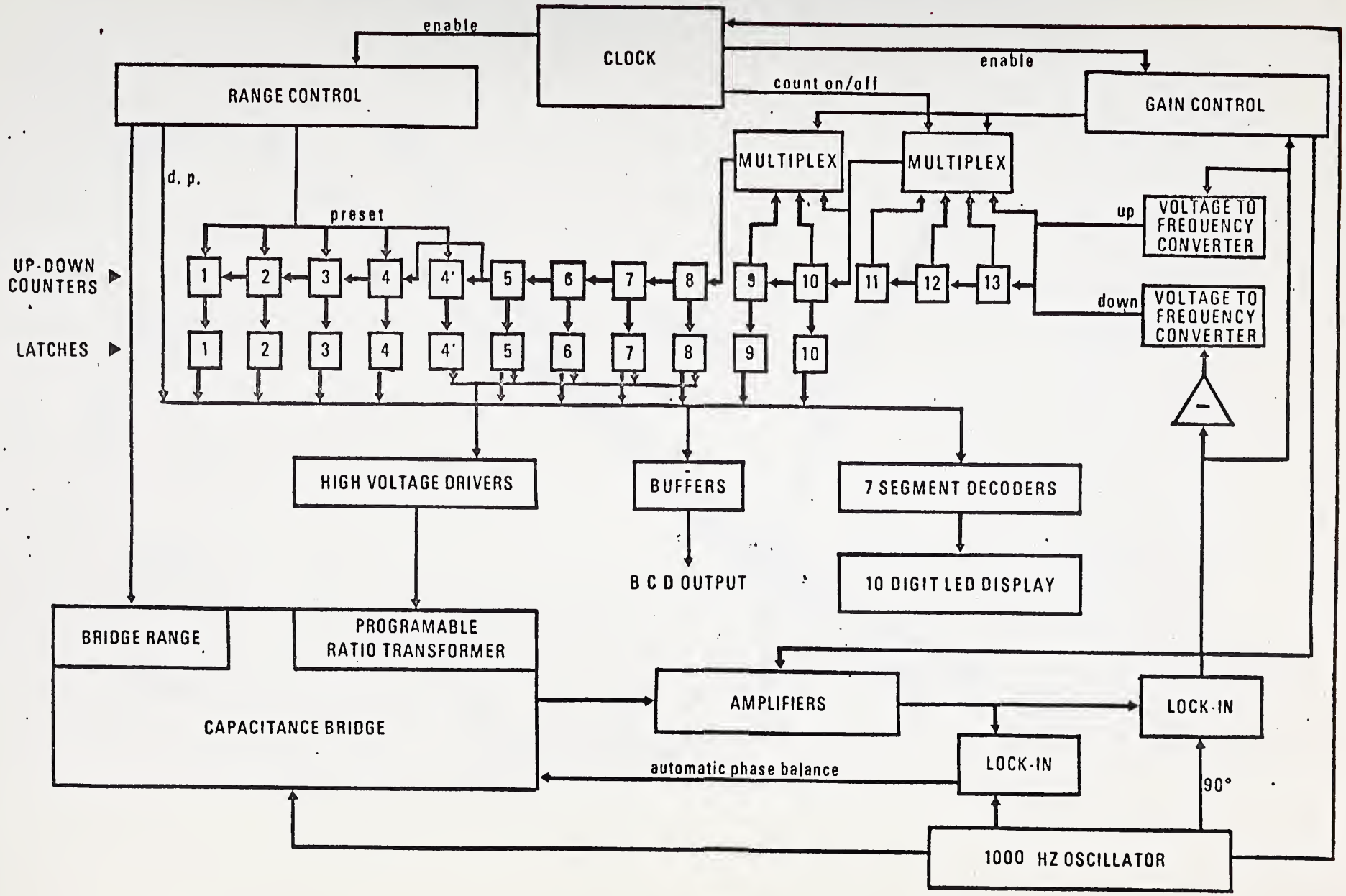
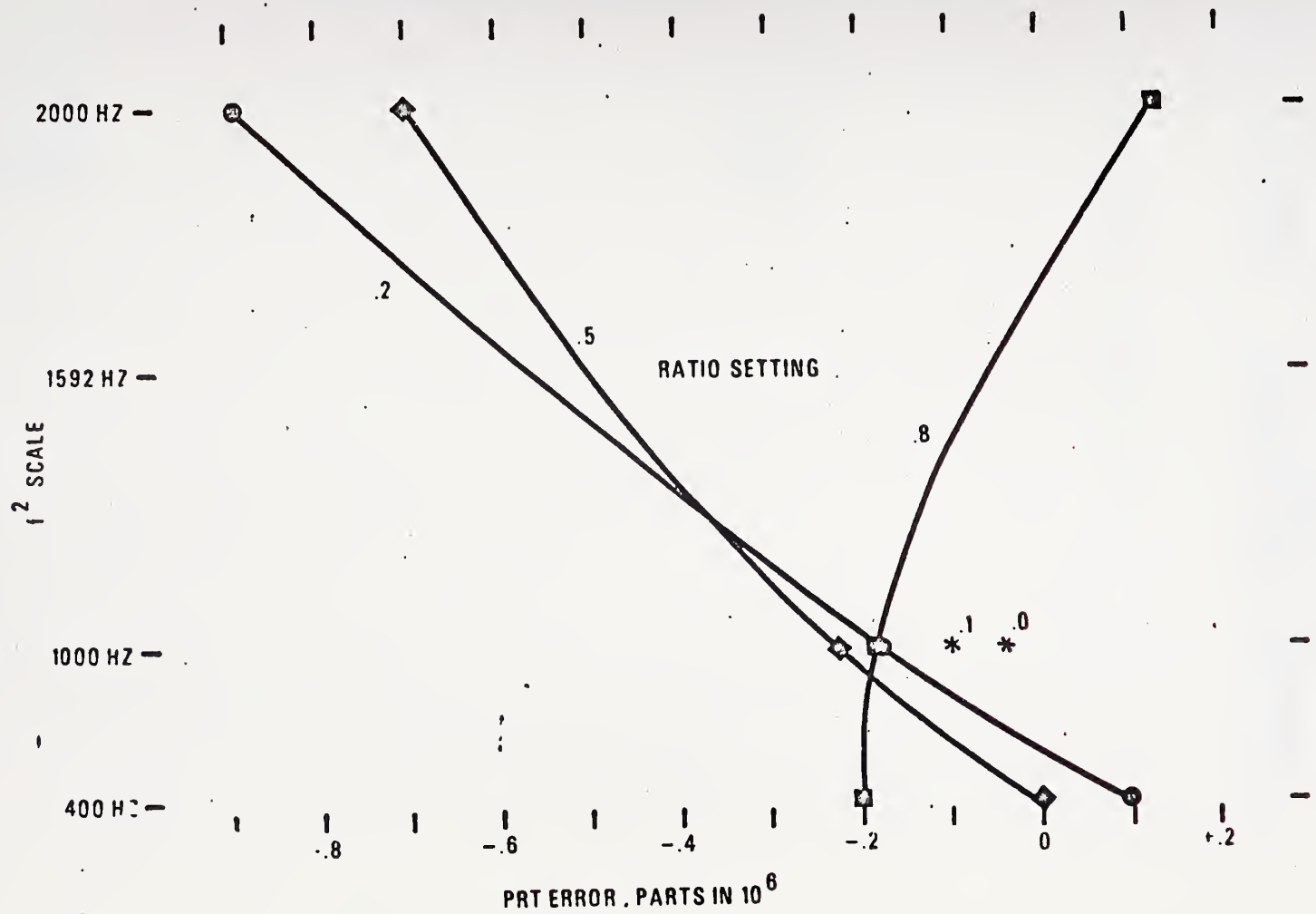


Fig 5

AUTOMATIC CAPACITANCE COMPARATOR




2000 HZ
1592 HZ
1000 HZ
400 HZ
f² SCALE
.2
.5
.8
RATIO SETTING
.1
.0
-.8
-.6
-.4
-.2
0
+.2
PRT ERROR . PARTS IN 10⁶