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Contract NONR 3437(00)

Semiannual Technical Summary Report

Period Ending 20 February 1964

'STABLE DENSE COLD PLASMA'

Advanced Research Projects Agency
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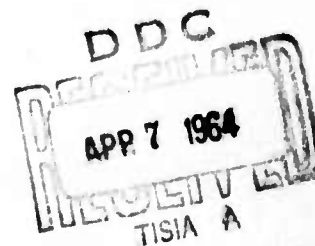
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31 March 1964

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INTRODUCTION

Activities on our dense cold gaseous plasma program during this reporting period have centered around two aspects, the preparation of gaseous plasmas of higher electron densities, and the measurement of electron concentrations in both the liquid and gaseous states.

I. PURIFICATION AND LIFETIME

The objective of this phase of the program is to enable us to attain higher operating temperatures and/or longer lifetimes than attained to date, i.e., 190°C for short periods and 52 minutes at 145°C , respectively, in order to obtain more gaseous data on the cold plasma. Two approaches have been taken in the past reporting period. One is a modification of the ammonia purification system to improve the purity of the ammonia as well as to make it easier to prepare the sodium-ammonia solution. The latter is accomplished by splitting the time-consuming purification of ammonia over a period of two days with low temperature storage over metallic sodium overnight.

A second approach is being initiated, namely, an independent method of determining the effects of (i) the various purification procedures for sodium and ammonia, (ii) the glass cleaning procedures, (iii) the compositions of the various glasses, and (iv) the seal-off. The method chosen, that of I. Wawshawsky, measures the hydrogen liberated at low temperatures by, e.g., traces of water adsorbed in the glass surfaces. The presently employed procedure of testing effectiveness of purification, etc., by behavior at high temperatures is more time consuming; in addition, it is more difficult to assign the sources of failures to particular causes. For this reason, we have designed and are setting up a modification of Wawshawsky's procedure. In this system, we shall evaluate, independently of the present conductivity

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technique, the effects of the various procedures, treatments, and compositions on the stability of sodium ammonia solutions. The revisions of the method of purifying the ammonia during the latter part of the reporting period follows: The initial purification is now accomplished by passing tank ammonia of 99.99 percent purity through a U-tube containing activated charcoal to absorb organic matter (e.g., hydrocarbons, aromatics, and their derivatives). The ammonia is then condensed over sodium in a dry box and kept there overnight at -78° . The purpose of the sodium is to reduce oxidizing impurities (mostly water). Even at -78°C , there is always some reaction between the sodium and the ammonia, a process which liberates hydrogen. With reasonably good ammonia as a starting point, a minimal amount of hydrogen will be formed. It is possible that dangerous amounts of hydrogen may be built up if impurities are present which can catalyze the sodium-ammonia reaction. Therefore, as a safety measure, we have installed a trap containing 60 grams of spongy palladium to absorb the hydrogen as it is formed. This amount of palladium is sufficient to absorb the hydrogen liberated from about 10 cm^3 of sodium. Before a new batch of ammonia is purified, the palladium is heated to about 100° under vacuum, and in this manner, rejuvenated. To date, our experience has been that the amount of hydrogen build-up overnight, using the palladium, is negligible.

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2. ELECTRICAL CONDUCTIVITY MEASUREMENTS

Conductivity measurements have been extended to the concentrated solutions in an attempt to produce gaseous plasmas with much higher conductivity than obtained previously, namely, greater than 100 mhos/cm. In addition, studies of the effects of pressure on electrical conductivity of liquid solutions at constant temperature have been initiated. Also, conductivity-temperature data are now being measured at constant pressure throughout most of the liquid range. A theoretical study of the electrical conductivity measuring technique has been carried out, and the behavior of the conductivity measuring system has been accounted for quantitatively. The method therefore has been reduced to an absolute method of measuring resistance so that, in principle at least, it is no longer necessary to standardize by comparison with known resistances.

Measurements of conductivity vs temperature along the vapor pressure curve have been extended to the more concentrated solutions and the results are shown in Figure 1. The temperature coefficients of conductivity of these solutions are smaller than those of the more dilute solutions. In all cases, there was a peak at about 160°C. A possible explanation for the peak is evaporation of NH_3 into the volume of gas above the liquid solution, thus making the solution more concentrated. Either decomposition or decrease in volume of the liquid due to further evaporation of NH_3 , thereby opening up the toroidal circuit, may account for the final drop in measured conductivity. We did not produce conducting gases even at 190°C for the concentrated solutions. It is for this reason that our procedures are being modified to get to the higher temperatures more rapidly since we would like to produce gaseous plasmas of the concentrated solutions. To do this, we have modified not only the purification procedure but also the experimental conductivity system to reduce

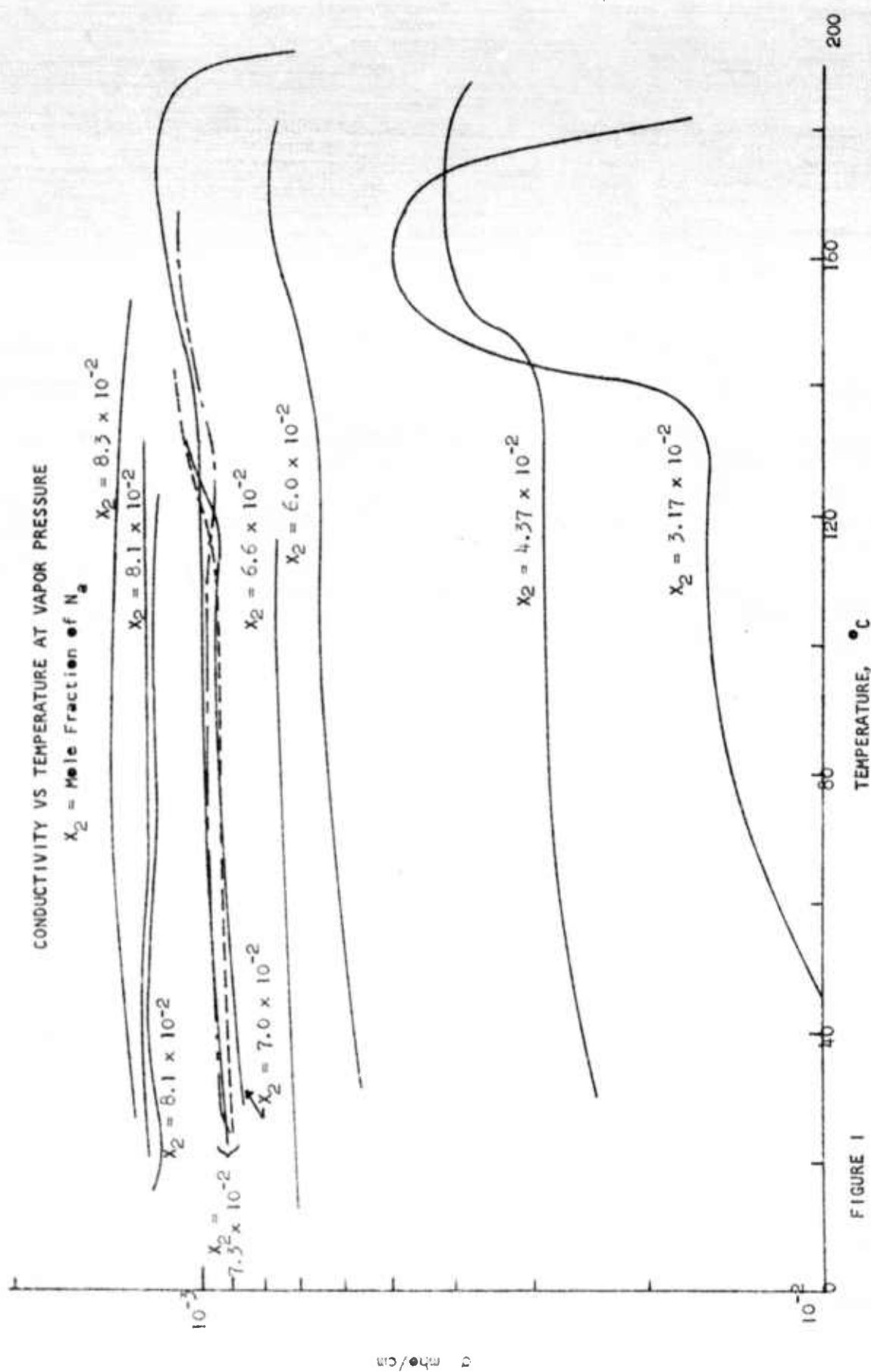


FIGURE 1

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operational difficulties at higher temperatures.

In the case of measurements of conductivity vs pressure at constant temperature, a hysteresis effect has been observed as shown in Figure 2. The source of this hysteresis has not yet been established. To determine whether the hysteresis is due to solubility of nitrogen in the liquid sodium ammonia solutions, we have modified the conductivity cells to increase the diffusion path length. In the new cells, the pressurizing gas will have to diffuse further through liquid before affecting conductivities.

In Figure 3, the result of a conductivity vs temperature at constant pressure measurement is compared to a similar measurement at the vapor pressure. These results appear to be valid, but we are withholding judgment on this until we learn more about the cause of the hysteresis effect noted above.

3. HALL EFFECT

The measurement of the Hall Effect in sodium ammonia solutions is of importance in that it offers us an independent method of determining electron densities and electron mobilities. Attempts reported in the literature to measure these, using classical chemical techniques, have encountered unresolved difficulties. The Hall measurement, as we are setting it up, should not only solve this problem but also enable us ultimately to extend the Hall measurements throughout the whole liquid range as well as into the gas.

A Hall measuring technique compatible with the stringent requirements imposed by the chemical behavior of sodium ammonia solutions has been designed. Initially, a unit will be fabricated that will operate in air rather than in the external pressurization bomb. Its range of usefulness will be from about -80 to $+20^{\circ}\text{C}$. Once the operating

CONDUCTIVITY VS PRESSURE

Mole Fraction of $N_2 = 3.6 \times 10^{-2}$

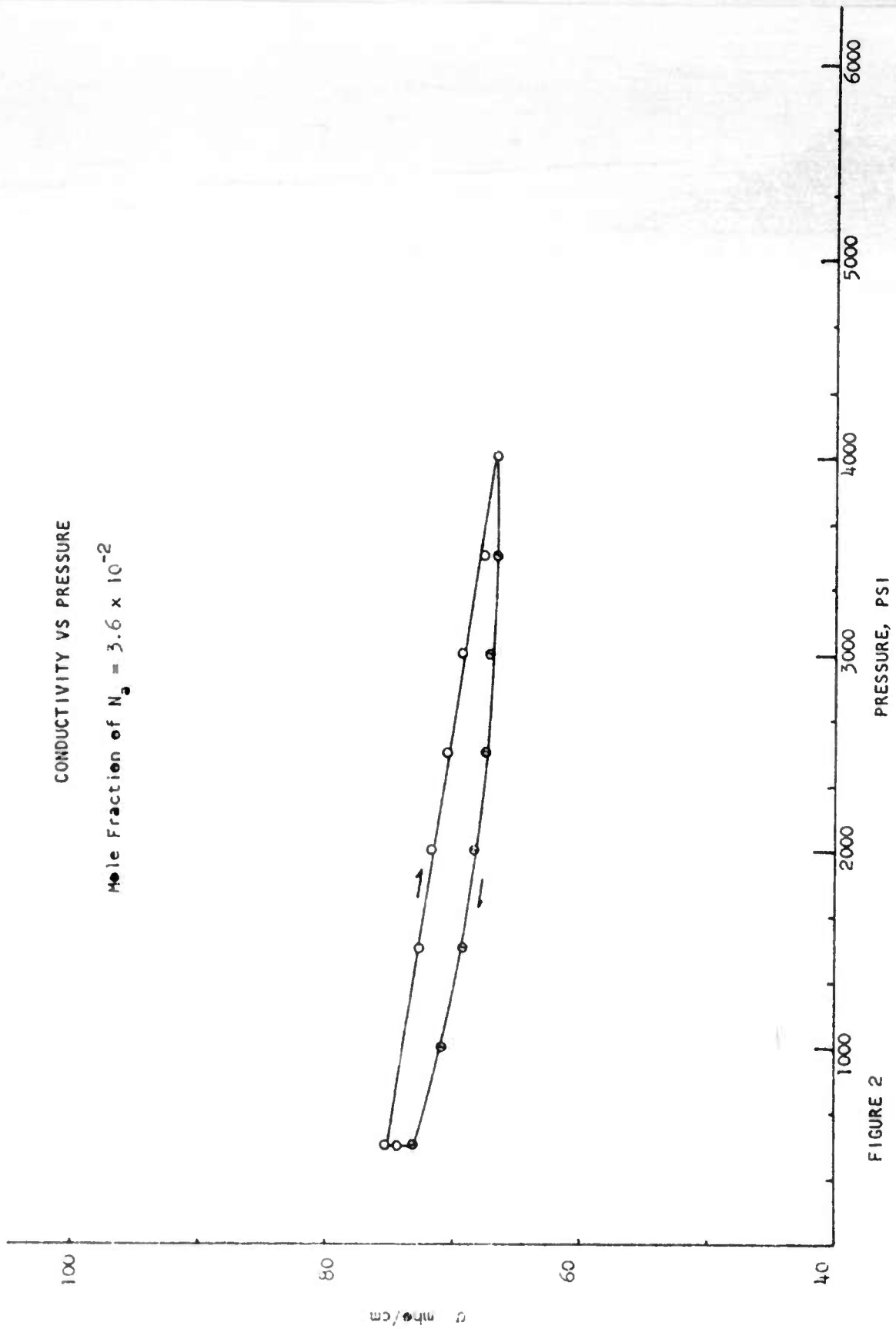


FIGURE 2

CONDUCTIVITY VS TEMPERATURE

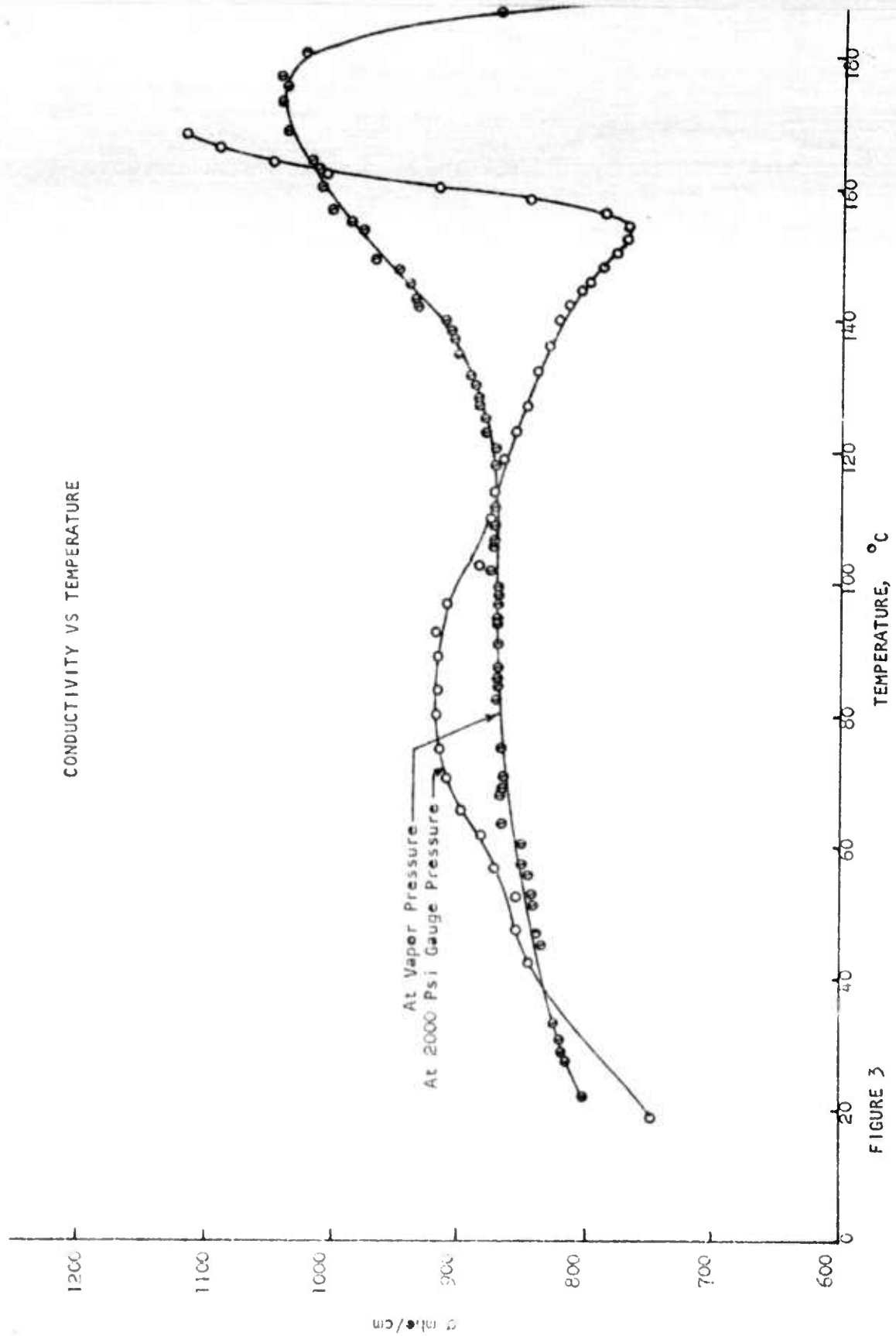


FIGURE 3

characteristics have been well established, the device may be re-designed for measurement to high temperature inside a bomb. Since the Hall cell will be fabricated from glass, the perpendicular alignment of the arms measuring the output signal cannot be controlled accurately enough for useful results. Therefore, a "boat" technique will be used to separate the Hall signal from the misalignment voltage and other background.

The magnetic field giving rise to the Hall effect will be 5000 gauss in the prototype system and its frequency will be 60 cps. The sample voltage will be at a frequency of 460 cps so that the Hall signal will occur at a frequency of 400 cps. This signal will be passed through filters tuned to this frequency to remove the 60 cps, 460 cps, and 520 cps background as well as harmonics generated by saturation effects in the magnet. The filtered signal will be fed to a phase-sensitive detector which will provide further discrimination against unwanted signals.

4. DENSITY MEASURING SYSTEM

To determine the variation in density with temperature change of a sample, an oven has been fabricated to house the sample while the temperature is changed over a wide range. The change in density of the sample will result in the rise of the level of the sample in a tube of known diameter. The height of this rise will be measured by means of a cathetometer.

A heavy support table has been made and on this an oven has been fabricated. The oven is separated from the table by supporting legs and the sample in the oven is supported separately by invar rods. A cathetometer, mounted on the table, is used to measure the rise in fluid level inside the oven. A fluorescent light is placed on the far

side of the oven to illuminate the meniscus. Three layers of pyrex glass, coated with an infrared reflecting film, serve to reduce heat loss of the sample while also serving as a barrier should the glass tube containing the sample rupture. As further protection, the optical path is bent 90° to keep the cathetometer and the operator out of direct line of sight.

A nichrome heating wire provides the heat to raise the oven temperature while a solenoid controlled liquid CO_2 injection system provides cooling. A fan circulates the surrounding gas while it is being heated or cooled and baffles serve to provide more uniform flow across the sample. Although the CO_2 cooling system requires an on-off controller, the heating element can be proportionally controlled for accurate temperature control.

In addition to density measurement, a conductivity head is also provided so that simultaneous conductivity-density-temperature measurements can be made.

This unit is currently being tested to determine heating and cooling rates. An additional CO_2 solenoid valve is being added to provide both high and low cooling rates. In addition, a second heating coil is being added and the system is being connected to a source of 220 VAC to provide a more rapid heating rate.

The present heating and cooling rates give a 50°C rise or drop from room temperature in the first two minutes. The lowest temperature attainable with the CO_2 , -84°C , is reached in about 9 minutes which is over 10°C per minute. In 9 minutes of heating, the temperature can be raised from room temperature to 150°C while 200°C takes about 27 minutes. The additional heating coils will improve this heating rate considerably. When heating from -84°C , a rate of 90°C for the first two minutes can be attained. This is the same rate that can be obtained when cooling from $+200^\circ\text{C}$.

PLANS FOR THE NEXT QUARTER

Evaluation of the effects produced by the various surface treatments and glass compositions will be initiated, using Warshawsky's technique. Based on these results and the use of a variety of initial fractional fillings of the conductivity cells, further attempts will be made to measure the electrical conductivities of the more concentrated gases.

Before proceeding with the main set of measurements of the pressure effects, the sources of the hysteresis mentioned earlier will be studied. When the hysteresis is either removed or better understood, the effects of pressure on conductivity at constant temperature will be studied. In addition, the conductivity will be measured as a function of temperature at constant pressure.

The density measuring system will be used initially from -80° to $+20^{\circ}\text{C}$ with samples designed for use inside the bomb. The electrical conductivity will be measured concurrently. A special set of samples will be prepared for measuring density as a function of temperature along the vapor pressure curve to elevated temperatures. In addition, our preliminary theory of the temperature coefficient of density will be compared with the experimental data.

It is planned to complete the fabrication of the Hall effect system in this period. This system is designed for operation from -80° to $+20^{\circ}\text{C}$. The high temperature Hall effect system will be built at a later date.

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