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AN APPARATUS FOR THE COMBINED MEASUREMENT OF SEEBECK COEFFICIENT AND ELECTRICAL RESISTIVITY

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ABSTRACT

An apparatus has been designed which allows simultaneous measurements of Seebeck coefficient and electrical resistivity of semiconducting thermoelectric materials from room temperature up to 1300K. This apparatus was necessitated by requirements to make thermoelectric measurements on materials, i.e., the SiGe alloys, whose properties exhibit thermal hysteresis. To this end, a previously described high-temperature Seebeck coefficient apparatus[1] has been modified to allow for measurement of the electrical resistivity. In addition to the two point-contact probes, represented by the thermocouples on the ends of the sample, two more point contacts were introduced along the length of the sample in order to conduct 4-probe measurements. Mathematical expressions were developed which relate the voltage-current characteristics to the sample size and probe placement. This apparatus has been tested and proven satisfactory by comparing measurements on SiGe alloys with those made in more conventional apparatus.

INTRODUCTION

In the course of investigations at JPL of a wide variety of thermoelectric materials for power generation, separate apparatus have been set up for the measurement of Seebeck coefficient and 4-probe electrical resistivity. The performance and reliability of those apparatus are well-established and well-proven by comparative measurements with other laboratories over a number of years. However, with our more recent emphasis on the Si-Ge thermoelectric materials a need existed to combine these two apparatus so that simultaneous measurements of Seebeck coefficients and electrical resistivity could be made. This was necessitated because of the propensity of Si-Ge alloys to exhibit dopant precipitation at high temperatures with the concomitant thermal hysteresis of thermoelectric properties.

In the previously described Seebeck apparatus[1], the sample electrical contacts were essentially point contacts on the ends of the cylindrical samples. In order to combine this measurement with the 4-probe electrical resistivity apparatus, two more point contacts (voltage probes) were placed along the length of the sample and the point contacts at the sample ends were utilized as current contacts. Thus, it was necessary to develop mathematical expressions for point, as opposed to area, current contacts and relate the current-voltage characteristics to the sample size and probe spacing. A description of the design of the apparatus follows.

APPARATUS DESIGN

In the original design of the Seebeck coefficient measurement apparatus[1] the sample was sandwiched between two quartz light pipes. A temperature gradient was established in the sample by these light pipes conducting heat from tungsten lamps to the sample ends. Niobium-tungsten thermocouples embedded in the ends of the light pipes and pressed on to the sample ends measured both the temperature gradient in the sample and the Seebeck voltage generated by this temperature gradient.

Figure 1 is a schematic diagram of an isothermal heater housing, sample holder, fuzed quartz, light pipes, thermocouple instrumentation and the Nb probes and Nb lead wires for resistivity measurement. The sample is supported by two spring loaded light pipes and light pipe support assembly. The light pipes were fabricated from a 1/2" diameter fused-quartz rod. Four small holes (0.75 mm diameter) were drilled in the ends of the light pipe to accommodate the thermocouple wires (W, Nb). The Nb wire was outermost to insure that the Nb metal contacted the sample. To shield the thermocouple junction from direct illumination from the light source, two holes (1.5 mm diameter) were drilled parallel and close to the thermocouple holes to deflect the light from the junction.

For resistivity measurement two Nb rods (i.e., voltage probes) were placed on top of the sample (0.25 inch apart). The ends of the rods are sharpened to a fine point, to reduce the surface contact. The rods were loaded with weights to insure good electrical contact. Small Nb wires (.010" diameter) were welded to the rods for electrical connections. The voltage leads and the thermocouple wires were brought out from the furnace in single-bore alumina tubes for electrical insulation.

Four heater elements were fabricated from 1/4 in. thick alumina plate, 0.05 in. wide, 1/8 in. deep grooves were machined into one side of the plate 1/4 in. apart, and .030

in. diameter tantalum wire was placed in the grooves for resistance heating. Each heater element wire was connected to an alumina stand-off outside the heater housing and all heater elements were connected in series to insure even current flow through all four heater elements. The inside walls of the heater housing were covered with ten (10) layers of tantalum foil and bolted to the walls by means of tungsten bolts to insure an isothermal enclosure. The box-like heater housing was made of 1/4 in. thick copper plate with water-cooled tubes soldered to the outside walls with a door-like front plate opening for easy access to the sample, thermocouples and the instrumentation.

The whole assembly was placed inside a 24-inch vacuum system which was connected to a 6-inch diffusion pump system.

A schematic diagram of the Seebeck and resistivity measurement system is shown in Figure 2. The top of the sample was instrumented with two Nb probes which were connected to a DVM for voltage measurement. The two sides of the sample were instrumented with two pairs of thermocouple (W and Nb) wires. The W legs of the thermocouple were directly connected to the Seebeck amplifier (A-2). The Nb legs of the thermocouple were connected to a relay. In the relax-mode of the relay, the Nb wires were connected to the Seebeck amplifier (A-1) by way of the relaxed contact of the relay. The output of A-1 is connected to a X-Y recorder for Seebeck coefficient measurements. In the energized-mode of the relay the Nb wires were connected to the load current supply (I_L) by way of the energized contact of the relay.

The schematic diagram also shows the system heater and the heater power supply, the light pipes, the 600W light bulbs and the lamp power supplies.

Resistivity Measurements

The system heater was used to bring a sample to a predetermined temperature. At this point the resistivity measurement was made, using a 4-probe measurement technique. Even though the furnace is isothermal, a small temperature gradient is always present across the sample which generates some voltage, E_{oc}. This voltage is monitored and recorded. After the E_{oc} is recorded the relay is energized and a small current (100-200mA) is passed through the sample and the R_s resistor. The R_3, or shunt, resistor (1 ohm, 1%) is used for the accurate measurement of the current flow in the circuit. Due to the resistance of the sample, a voltage drop is generated across the sample and this voltage drop is monitored by the two Nb probes placed on top of the sample. Knowing the

open circuit voltage (E_{oc}), the load voltage (E_L) and the load current (I_L), the resistance of the sample can be calculated. Knowing the area (A) of the sample and the distance (l) between the two Nb probes, the resistivity of the sample can be determined. Due to the size of the sample, the placement of the probes and the point contacts for the load current input, I_L to the sample, a correction factor is applied for the final calculation of the sample resistivity (P_c) (Figure 3).

Seebeck Measurement

After the sample resistivity measurement is completed a small amount of current is applied to each light bulb by turning on the lamp power supplies, slightly raising the sample temperature. Current is increased to one lamp and approximately the same amount is lowered to the other lamp. Thus, one end of the sample temperature (T_H) is raised, while the other end is decreased (T_C). However, the average temperature (T_{AVG}) remains the same.

Amplifier circuit

A schematic diagram of the amplifiers is shown in Figure 4. Two pairs of thermocouple (Nb and W) wires lead from the sample to the amplifiers. Figure 4 also shows the primary thermoelectric voltages ΔV_1, ΔV_2, ΔV_3 used to determine the absolute Seebeck coefficient of the sample, S(T), where T is the sample temperature. V_{Nb} and V_W are the Seebeck voltages generated by the two thermocouple arms. Because of the very low Seebeck voltage of the Nb leads at any temperature, V_{Nb} can be assumed to be negligible. Therefore, the input to the A-1 amplifier is the Seebeck voltage (V_S) generated by the sample: $\Delta V_1 = V_S$. The input to the A-2 amplifier is the sum of the Seebeck voltage generated by the sample (V_S) and the Seebeck voltage generated in the tungsten leads (V_W), i.e., $\Delta V_2 = V_S - V_W$. The output of amplifier A-1 is fed to the Y-axis of the X-Y recorder where it represents the Seebeck voltage of the sample. The outputs of Amplifiers A-1 and A-2 are algebraically summed at the input of Amplifier A-3, where the two voltages ΔV_1 and ΔV_2 are subtracted from each other. This difference is proportional to the temperature gradient ΔT across the sample, i.e., $\Delta V_3 = (\Delta V_1 - \Delta V_2)$. The output of Amplifier A-3 is fed to the X-axis of the X-Y recorder where it represents the temperature gradient across the sample.

EXPERIMENTAL RESULTS

In order to test the performance of the apparatus a comparison was made of the electrical resistivity of an n-type Si-Ge/GaP sample measured in our standard 4-probe (large area current contacts) system versus the resistivity values obtained using the combined Seebeck-resistivity (point current contacts) system. The results are shown in Figure 5-1 to be in excellent agreement.

A comparison of the Seebeck coefficient values obtained on the same sample in both the standard Seebeck apparatus and the combined apparatus is shown in Figure 5-2, and again, the agreement was excellent.

CONCLUSIONS

A combined high-temperature Seebeck coefficient and electrical resistivity apparatus has been constructed which has the advantages of simplicity to load (no contact fabrication) and operate, has excellent reproducibility and has been shown to give good agreement with measurements made in other apparatus proven by comparison with other laboratories. Plans are in progress to completely automate the apparatus.

ACKNOWLEDGEMENT

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REFERENCE

1. C. Wood, D. Zoltan and G. Stapfer, Rev. Sci. Instrum. 56 719 (1985)

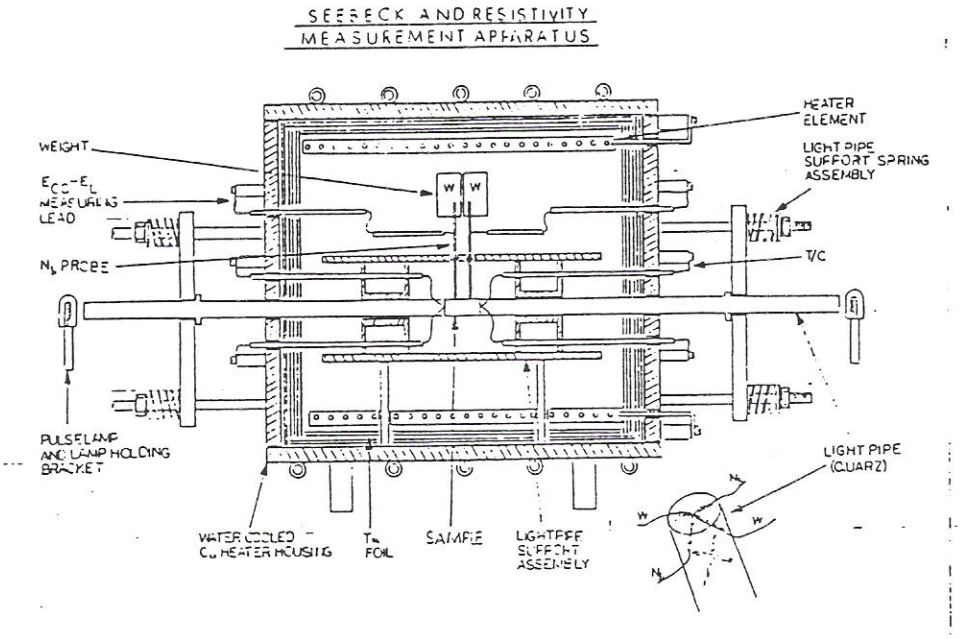


Figure 1: Schematic diagram of an isothermal heater housing, sample holder, fuzed quartz, light pipes, thermocouple instrumentation and the Nb probes and Nb lead wires for resistivity measurement.

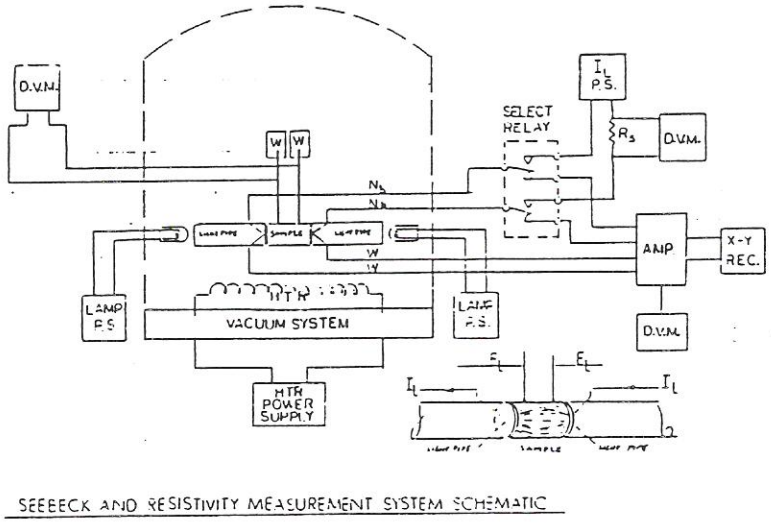
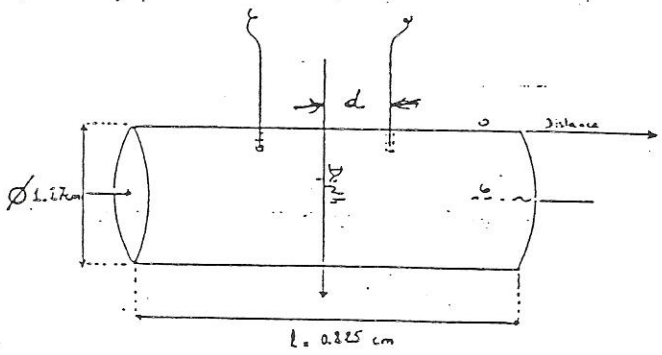


Figure 2: Schematic diagram of the Seebeck and resistivity measurement system.



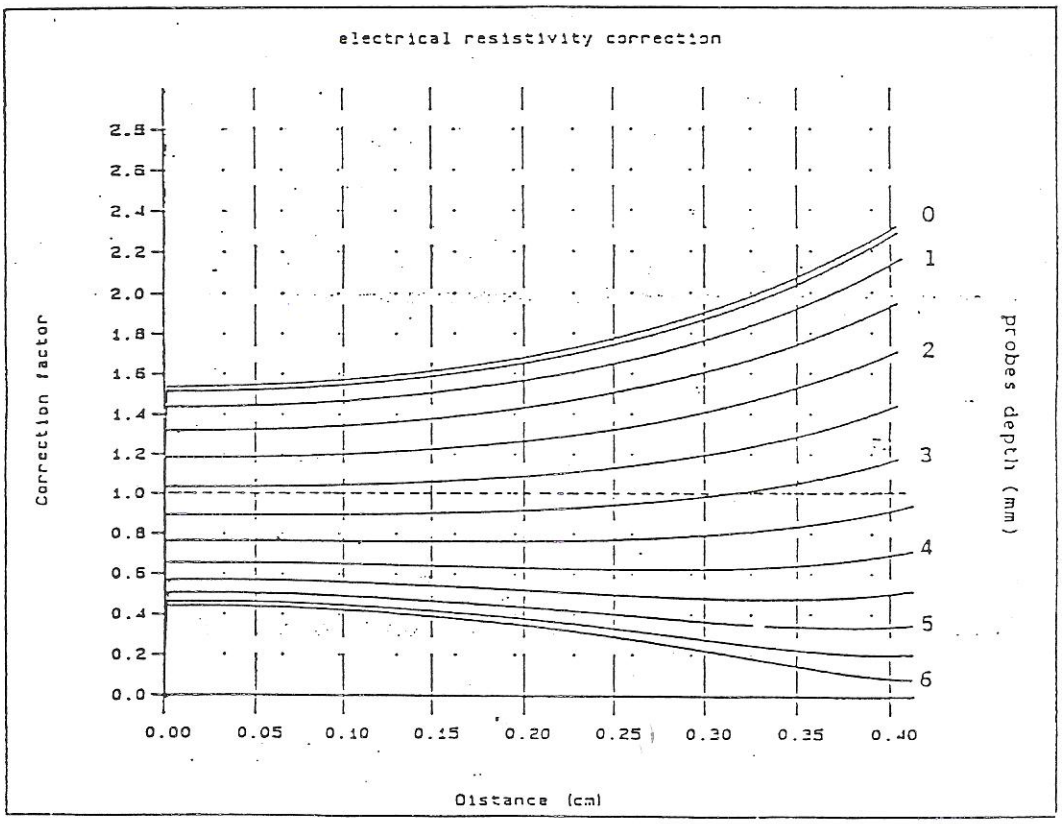


Figure 3: Correction factor for the calculation of the sample resistivity.

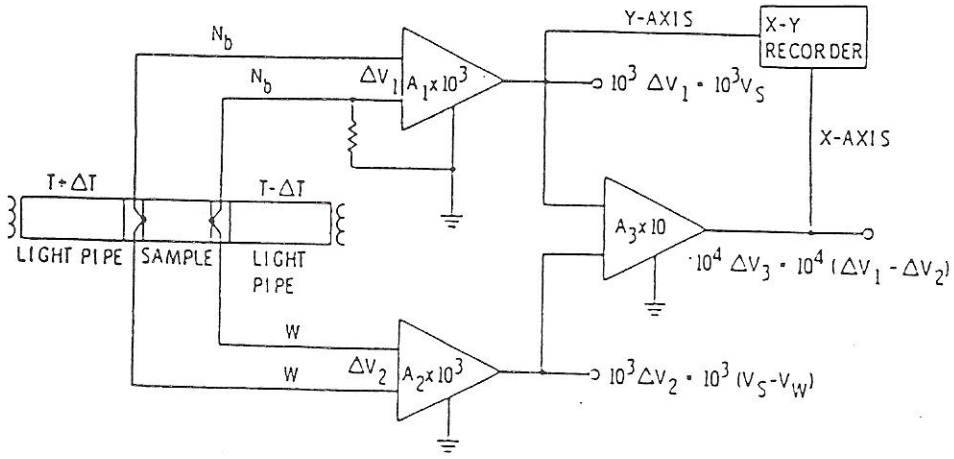


Figure 4: A schematic diagram of the amplifiers.

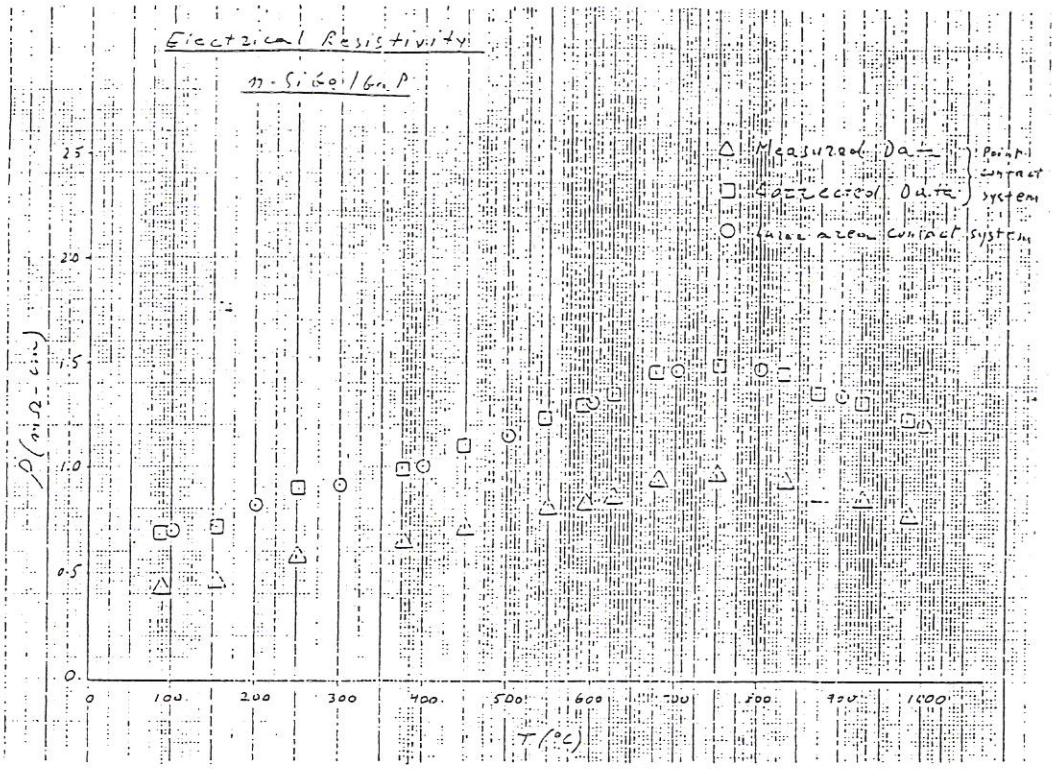


Figure 5-1: Resistivity values obtained using the combined Seebeck-resistivity system.

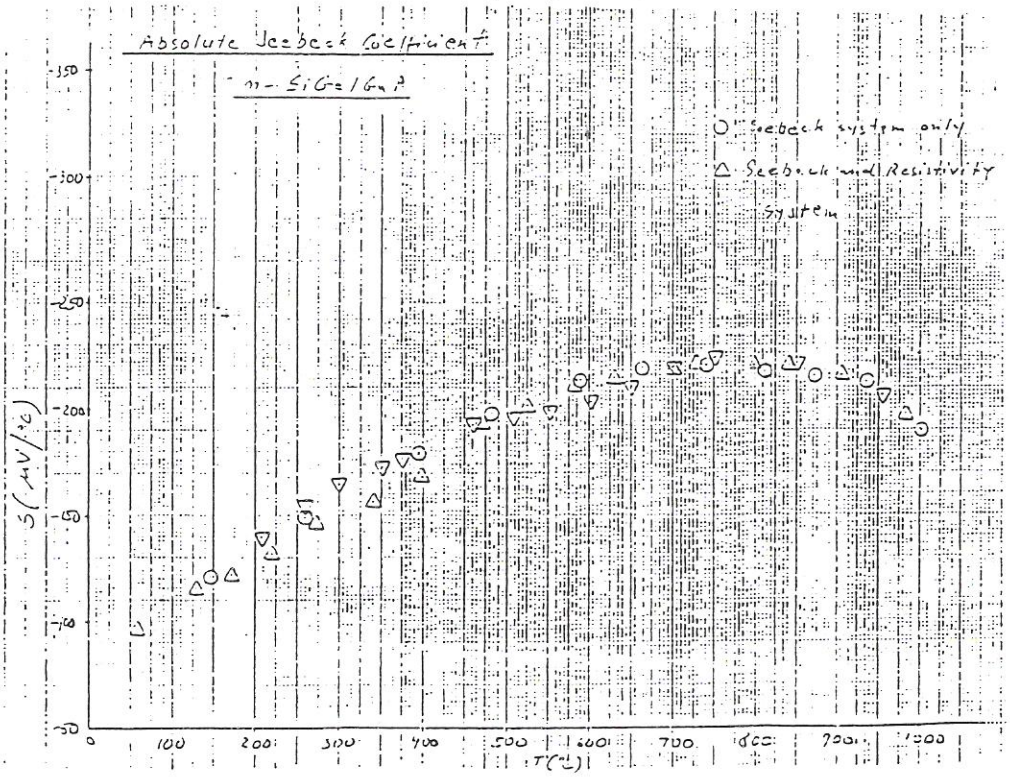


Figure 5-2: A comparison of the Seebeck coefficient values obtained on the same sample in both the standard Seebeck apparatus.