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GENERAL ELECTRIC COMPANY

LEAD TELLURIDE RESEARCH PROGRAM

R. O. Carlson and L. F. Kendall

AEC Contract No. AT(29-2)-2462

Research and Development Center
Schenectady, New York

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FINAL REPORT

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October 1968

by

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Research and Development Center
Schenectady, New York

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ABSTRACT

Single-phase PbTe doped with sodium or iodine for high temperature thermoelectric power generation was synthesized in quartz tubes or in an autoclave pressurized with argon. The PbTe was hot-extruded into rods whose densities exceeded 99.5% of theoretical. Silver and samarium were tried as substitute dopants. Compatibility tests at 600°C for 1000 hours revealed that some module construction materials such as Lava and Fiberfrax and metals like Ni, Mo, Ta, and W did not alter the electrical properties of p-type thermoelements, whereas air, water vapor, Fe, Mn, Cr, and stainless steel were highly deleterious. Six life test cells were operated with couples held in a 210°C-550°C gradient for thousands of hours. Mo or W provided a diffusion barrier at the hot end of the couples, but there was still a 3 to 9% degradation in power output over 1000 hour operation, arising mostly from an increase in the p-type thermoelement resistance.

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I. SUMMARY

The objectives of this program were to provide information on the basic physical properties of doped, single-phase, dense PbTe prepared by several novel synthesis and fabrication methods, to determine the sources of degradation observed in p-type PbTe under high hot junction temperatures, and to determine the compatibilities of the single-phase material with other materials at temperatures up to 600°C.

PbTe rods were fabricated with over 99.5% of theoretical density by extrusion from a heated steel die from material synthesized either in a sealed quartz tube or in an argon pressurized autoclave. The compositions were chosen to provide single-phase PbTe with dopant concentrations close to optimum for high temperature power generation. Sodium and iodine were used as acceptor and donor dopants, respectively; a brief study showed that silver and samarium would be possible dopant substitutions.

Two series of compatibility tests were carried out with potential contaminants and p-type PbTe sealed together in quartz tubes at 600°C for 1000 hours. Among the construction materials of our life test cells, Lava ceramic and Fiberfrax insulation had no effect on the electrical properties, but iron, stainless steel, air, and water vapor did cause large changes. The metals Ni, Mo, Ta, and W were found to be safe possible substitutes for iron and stainless steel as contact materials. Ag, Cr, and Mn were found to attack PbTe through a vapor phase reaction.

Six cells with thermocouples fabricated from extruded rods were operated for times up to 3500 hours while held in a 210°C to 550°C temperature gradient. Measurements were made of power output, electrical resistivity, and Seebeck coefficient. Molybdenum and tungsten were employed as barriers to iron diffusion at the hot junction and did reduce the degradation rate. Power output dropped about 3 to 9% over the first 1000 hours of operation with some reduction in degradation rate for more extended periods. There was generally a continuing increase in the p-type thermoelement resistance, arising from donor compensation due to diffusing impurities, such as iron crossing the end contacts or iron, chromium, or manganese coming in through the element surface after vaporization from hot parts of the test cell.

II. INTRODUCTION

The application of lead telluride for nuclear thermoelectric power systems has been hampered by lack of control of the initial material and the degradation of that material under high temperature conditions. It was the purpose of these studies to provide some answers to these problems by the preparation of highly uniform, quality controlled, single-phase material and to evaluate this material against potential contaminants and in actual operating couples.

Through Company-sponsored programs, several approaches were developed to the laboratory production of very dense PbTe with uniform and reproducible properties. The composition of the PbTe was formulated to be single phase; i.e., the potential dopants required to bring the electrical properties of PbTe to their desired range for couple operation were added only in the minimum amounts necessary for doping. By comparison with commercially available PbTe, the doped single-phase high density ingots were found to have similar electrical properties to commercial grade PbTe, but with superior mechanical properties and high temperature stability. The starting elements were chosen as the highest purities available, and the synthesis and fabrication methods insured that no loss or undesirable distribution of dopant occurred during the processing steps. Naturally, such care and control means that the resulting thermoelements are relatively expensive, but with the quality and reproducibility of end product needed for demanding space and terrestrial applications.

In parallel with an effort to enlarge the synthesis and fabrication skills for material preparation, the need to know how this single-phase, dense material would act under operating conditions led to plans for operation of couples in an ultrahigh vacuum cell to evaluate long term performance under standard test conditions. Six couples were to be fabricated and sealed off and their performance criteria of power output measured over thousands of hours.

The life test cells contained many construction materials of unknown quality as far as their behavior toward hot PbTe is concerned; therefore, a series of tests were devised in which these construction materials were enclosed in proximity with PbTe pieces, actually contacting the PbTe only if the construction material in the test cell also was in direct contact with the PbTe. The basis for evaluation was room temperature measurement of the electrical properties of the PbTe pieces before and after the high temperature treatment.

III. LEAD TELLURIDE PREPARATION

The lead telluride used on this contract was synthesized by two different methods, both developed in Company-sponsored work prior to this contract. One method utilized sealed, evacuated quartz tubes and the other utilized a graphite crucible in a pressurized autoclave. In both methods the starting ingredients were 99.999+% lead and tellurium with reagent grade lead iodide or sodium to dope the n- and p-type preparations, respectively. The iodine and sodium concentrations were chosen on the basis of our earlier studies of hot-pressed rods to provide close to optimized PbTe for thermoelectric power generation over the 210°C to 550°C range as in the life test cells. The quantities of each constituent required to give the desired composition were weighed on an analytical balance and then transferred quantitatively to either a clean, baked quartz tube coated with pyrolytic graphite or a baked-out crucible made of high purity graphite. The quartz tubes were pumped to less than 10^{-6} Torr while baking at 150°C and then sealed off; they were then heated to 1025°C in a furnace, shaken several times, and then quenched in water. The loaded graphite crucible was placed in an induction heating coil inside a water-jacketed pressure vessel. The vessel was then pumped to less than 10^{-5} Torr and backfilled with argon to about 30 atmospheres. The temperature of the crucible was raised to 1025°C and held for one-half hour while the vessel and its contents were rocked. The heated power was then turned off, and the crucible cooled by radiation and gas convection to the cold walls of the vessel.

IV. EXTRUSION AND ELEMENT PREPARATION

Shortly before we began work on this contract, we had achieved considerable success in fabricating lead telluride by extrusion. Since this method appeared to be significant improvement in the state of the art, we decided to make lead telluride thermoelements for our experiments by extrusion rather than by the vacuum hot-pressing method which we had previously developed and used. The quality of the hot-pressed elements was excellent, but the process was slow and, therefore, expensive. The extrusion process on the other hand appeared to offer economic advantages, but the process was incompletely developed and the product, relatively untried.

After some experimentation we had extruded shiny, smooth rods of n-type PbTe 1/8 inch in diameter by 30 inches long, the maximum permitted in our developmental equipment, and a few pieces 1/4 inch in diameter by two or three inches long with a little lower surface quality, but entirely acceptable. The rods were extruded through a hollow cylindrical die which was pressed against a charge consisting of lumps of PbTe contained in a die body which closely fitted the die. Both the die and the die body were made of hardened steel. The temperature of the assembly was regulated as desired by adjusting the power to a band heater which covered the outer cylindrical surface of the die body. The die assembly was contained in a hermetic housing which could be evacuated and, if desired, backfilled with argon. The load was applied to the die by means of a push rod which moved through an O-ring-sealed opening in the outer container. The various parts of the extrusion equipment are shown in Figure 1.

All of the extrusions in our early efforts were made at about 350°C in vacuum with shear dies, i.e., the axial hole through which the extruded material passed and the surface which pressed against the billet met at a right angle with at most a slight chamfer around the hole. While this design was entirely satisfactory for 1/8" diameter, successful extrusions of 1/4" diameter rods using a shear die were the exception rather than the rule. Most such rods ranged from fair, with frequent surface defects such as tears and fine shallow cracks, to very poor consisting of many small pieces filled with large cracks. The cracks in the rods classified as "fair" extended only about 0.005 to 0.01 inches beneath the surface so that sound pieces with densities exceeding 99.5% of theoretical (8.25 gcm^{-3}) could be obtained by grinding away the outer surface using a small lathe to rotate the pieces. Furthermore, extruded pieces of both p- and n-type PbTe were found to have the same electrical properties — i.e., electrical resistivities and Seebeck coefficients versus temperature — as vacuum hot-pressed pieces of the same chemical composition, as tabulated in Table I. Figures 2 and 3 plot the electrical properties of a p-type extruded piece from room temperature to 600°C, compared with two hot-pressed pieces.

Nevertheless, ways of eliminating the surface defects in the larger diameter ingots were studied in order to eliminate the grinding step. We first tried using a die in which the angle between the leading surface of the die and the axial opening was about 45°; no recognizable improvement was noted. Markedly improved surfaces were obtained, however, when the interface between the die and the lead telluride was lubricated with Teflon, the product being a core of PbTe with an extremely thin sheath of Teflon. The best rods were produced when an emulsion of Teflon, Dupont TD-30, was painted on the die surface. During subsequent tests, the electrical properties of p-type pieces extruded with

Teflon were found to be more variable than others which had not been in contact with Teflon. It is clear therefore that Teflon is not an acceptable lubricant for this application. The only other lubricating material we tried was Lexan; this was ineffective, however, and it carbonized badly during the run. Most of the extrusions were made under vacuum; a few made in argon at atmospheric pressure appeared to be unimproved by the presence of the gas.

Table II is a summary of the extrusion runs. Material for the extrusions was synthesized either in quartz tubes or in an autoclave, but the p-type extruded rods used in the compatibility tests or the life test cells were mostly synthesized by the autoclave process. Electrical data on the rods used for these tests are tabulated in Table III. Rods E-5 and E-10 were made prior to this contract period.

The methods of measuring resistivity and Seebeck coefficient are given later in this Report. The uniformity of some of the rods is quite good, with no particular trend in properties from one end of the ~5" rod to the other end. Rods E-38 and E-39 among the p-type rods are more variable than the others and they had Teflon present during extrusion. The n-type rods were less uniform; whether the variability arises from the volatility of the doping element iodine was not clear from our studies.

Two of the later extrusions, E-44 and E-45, utilized dopants other than iodine and sodium, and electrical data on these rods are summarized in Table IV. Silver (Ag) is identified in the open literature as an acceptor dopant⁽¹⁾, and Ag had been tried previously in this laboratory as a dopant in the hot-pressed rod listed in Table IV. The acceptor action of Ag at the chosen concentrations was quite weak, but the extruded rod E-44 was quite uniform along its length. An anneal at 700°C for one hour in a sealed evacuated quartz tube resulted in a large increase in acceptor concentration (ρ shifting from 4 to 0.66 mohm-cm), so that the Ag-doped rod was now fairly close to having the desired electrical behavior for thermoelectric power generation. Still, only a fraction of the Ag present was electrically active as 0.66 mohm-cm corresponded to about 1×10^{19} acceptors/cm³, whereas the Ag concentration of 0.005 atom fraction represented about 7×10^{19} silver atoms/cm³. Annealing did provide a way of making Ag a potentially usable acceptor dopant in PbTe.

The donor action of samarium (Sm) was based on our previous contract study of rare earth and transition metal impurities in PbTe.⁽²⁾ That study showed that we could introduce enough Sm to reduce the resistivity to 0.2 mohm-cm with an apparent saturation of Sm at a level of about 4×10^{19} cm⁻³. Rod E-45 is underdoped for thermoelectric usage, and the 700°C anneal did not materially increase the donor concentration. However, Sm still should prove useful as a donor impurity because its dopant concentration can be increased well beyond the level in E-45.

V. MATERIAL COMPATIBILITY TESTS

The material compatibility tests consisted of high temperature (600°C) exposure of p-type extruded PbTe thermoelements to various potential sources of chemical contamination found in our life test cell fixtures. The aim was to identify the contaminating materials from the change in electrical properties of the elements and then remove these contaminants from later test cells. The exposure temperature was 600°C (1112°F), chosen as 50° above the operating life test cell condition. The time period was 1000 hours or about six weeks, which allowed two series of tests to be conducted.

The test samples were obtained from several of the extrusion runs. The lengths were cut with a wire saw to 0.40" and then ground down from a diameter of 0.225" to about 0.18" to remove the cracked and irregular surface layer of most extruded rods. The samples were then measured for Seebeck Coefficient and resistivity as described in the next two paragraphs.

The measurement of resistivity ρ was carried out by a four-point method. Current was applied along the 0.40" length of the sample through broad-area end pressure contacts, with indium or gold foil between the PbTe and cylindrical metal contacts to reduce the contact resistances. One voltage probe was fixed by tightening down a fine screw from a concentric ring around the sample. The other probe was a long needle which could be manipulated by micrometer drive along the length of the sample. The resistance was measured on a Keithley Milliohmmeter which applied a square-shaped current pulse at a repetition rate of 100 pulses per second and measured the peak value of the alternating voltage across the voltage probes. The use of a square-wave pulser avoided the Peltier effects which would add extraneous voltages for a continuous current through the sample. The movable probe was shifted at convenient steps of 50 to 100 mils along the side of the PbTe sample so that the uniformity of the sample was checked. Both the fixed and movable probes were kept at least the distance of the sample radius away from the sample ends in order that inhomogeneities of current distribution near the contacts would not influence the measurement of bulk resistivity. Occasionally the probes were shifted 90° or 180° by rotation of the sample to further check on the sample homogeneity.

The Seebeck Coefficient S was measured by placing the cylindrical PbTe sample in the V-groove of a large copper block and then pivoting against the hot side of the sample a hot probe shaped much like a soldering iron. A copper-constantan thermocouple was located in the copper block and also near the tip of the hot probe. A switching arrangement allowed measurement on a potentiometer of the voltage of the hot probe thermocouple or of the cooler copper block thermocouple, and also the Seebeck voltage across the copper leads of each thermocouple. The whole probing arrangement was inside a large wooden box for isolation from room air drafts, and about 10 minutes was allowed for the temperature to stabilize after the hot probe was pressed against the sample. We chose a temperature difference of about 10°C to provide reasonable values for the Seebeck voltage. The measurements were repeated for several points along the sample and after a rotation of the sample to check its uniformity. If the sample were short, as for these 0.40" samples, the sample was also placed vertically on the copper block so that the hot probe hit the center of the ingot rather than the outer surface, again as a check on sample uniformity.

After electrical measurement, the 0.40" long samples were ultrasonically cleaned in alcohol, then placed in quartz tubes which had been coated with pyrolytic graphite (for most cases) and prebaked. The materials under suspicion as contaminants were baked out at elevated temperatures as appropriate to their life test conditions prior to insertion in the quartz tube with the p-type PbTe sample. Many of the tubes had an indent along their length to prevent the potential contaminant from directly contacting the sample, but in other cases the PbTe sample was directly bonded to a contaminant or wrapped in it (Fiberfrax case). The tubes were sealed off under vacuum except for the air and water vapor tests. These last two were designed to test the effects of atmospheric contamination at the levels one would expect to find in a test cell with a small leak or in a setup in which the thermal insulation was insufficiently outgassed.

The quartz tubes were made as small as possible in order to limit the free volume and special care was taken to insure that the surfaces of the tubes were isothermal. At 600°C, where the vapor pressure of PbTe is of the order of 10^{-2} Torr, temperature differences of only a few degrees over the tube surface can lead to noticeable transport of material by evaporation and recondensation on cool spots over a long period. To protect the samples against this transport effect, the exposures were conducted in a resistance-heated muffle furnace of about one cubic foot volume, in which the hot air of the interior was circulated by a fan inside the hot zone of the furnaces. Tests were conducted to determine the suitability of this furnace, and it was found that the extremes of the on-off power supply cycle amounted to only about 5°C. This cycle time was short enough that for tubes wrapped in aluminum foil there was no evidence of condensate from trial samples held for long periods at 600°C.

The first series of compatibility tests was run with p-type thermoelements only and the following materials:

1. Base element alone
2. P-type element exposed to the vapor from n-type elements
3. Element wrapped in Fiberfrax thermal insulation
4. Element exposed to vapors evolved from an iron heater block with a cartridge-type electric heater in place
5. Element with SnTe buttons attached to its ends by melting SnTe in place
6. Element brazed with SnTe to a molybdenum surface which was diffusion bonded to a stainless steel tab
7. Element exposed to the vapors evolved from fired Lava ceramic
8. Element brazed with SnTe to an iron surface which was diffusion bonded to a stainless steel tab
9. Element exposed to vapor evolved from 300 series stainless steel
10. Element exposed to vapor evolved from stainless steel sheathed thermocouple
11. Element in a tube containing air at about 0.8 mm pressure
12. Element in a tube containing an amount of water vapor sufficient to saturate the tube at room temperature

Thirteen tubes were used for the first series of compability tests, there being two control tubes. Electrical measurements were made on each sample after completion of the 1000-hour stay at 600°C. The samples were then ground down further and remeasured to determine if the observed changes in electrical properties represented true bulk effects or just surface effects.

The results of the first series of compatibility tests are given in Table V. Only the resistivity data are listed. The Seebeck Coefficient data showed, in general, similar trends to the resistivity data but the shifts were not as large as the resistivity data and are not reproduced here. The samples were divided into three groups depending on the resistivity change. For one large group, there was little or no change in resistivity, implying that the materials such as Lava, Fiberfrax, n-type PbTe, and SnTe did not contaminate PbTe. One of the two control samples probably developed large cracks in handling and so did not appear stable. A second group of samples enclosed with air, water vapor, stainless steel, and the thermocouple in stainless steel, showed about a doubling of resistivity, showing that these materials should be considered risky near PbTe at high temperature. The air and water vapor results confirmed our strong emphasis that the test cell be baked and pumped out to very high vacuum before sealoff and life testing. The third group comprising molybdenum- and iron-clad stainless steel bonded to the PbTe and an iron heater block showed enormous increases in resistivity. In all cases, the surface grinding did not materially affect the resistance after the 600°C anneal showing that the resistivity changes were indeed bulk effects.

The first series of compatibility tests was comforting in showing us that some of the construction materials like Lava and Fiberfrax are safe to use. The drastic effects from iron and stainless steel led us to consider for the second series of compatibility tests several alternative construction materials for the life test cells. These materials were molybdenum, tantalum, and tungsten, chosen as alternatives to iron for the heater block or stainless steel for the thermocouple tubing; silver as an alternative to stainless steel for thermocouple tubing; nickel, chromium, and manganese, constituents of 304 stainless steel and chosen to see if we could isolate the constituents of 304 stainless steel and steel; and stainless steel itself as a check on the first series tests.

The results of the second series of compatibility tests at 600°C for 1000 hours are given in Table VI. As in the first series, the starting p-type elements were extruded rods ground down to 0.18" diameter with one exception. Of the test samples, those with the metals Ni, Ta, Mo, and W as well as the control sample showed little or no change in resistivity, so these metals could be considered as "safe" construction materials. The sample with stainless steel showed about a doubling of resistivity in agreement with the result from the first series. The sample left intact as extruded which had a Teflon coating on it (most of the later extrusions were carried out with Teflon lubricant in the die to improve the surface quality of the extruded rod) also doubled in resistivity, indicating that the Teflon may contain some doping impurities such as fluorine which are activated at high temperature. Three samples were grossly attacked by the metal enclosed

in their tubes. Since in all cases in this second series, the metal was separated from the p-type PbTe, the reaction of these last three metals, A, Cr, and Mn must arise from a vapor transport. Mn has a relatively high vapor pressure at 600°C so its reactivity is not surprising. Ag and Cr have low vapor pressures, but evidently their reaction with PbTe to form tellurides proceeds so rapidly that Ag and Cr must constantly vaporize to try to saturate the tube volume. Since both Mn and Cr are constituents of 304 stainless steel, the contaminating action of this steel may arise from the vapor transport and reaction of these elements with PbTe. However, iron is the major constituent of stainless steel and is also a bad contaminating metal near PbTe.

VI. THERMOCOUPLE PERFORMANCE TESTS

During the lead telluride work at the Research and Development Center on Company funds, apparatus and methods were developed to provide long-term tests of thermocouples in an inert, reproducible environment. Basic to the attainment of these conditions were the use of ultra-high vacuum test cells, the use of internal fixturing materials from which volatile contaminants, mainly water vapor and oxygen, could be removed by baking at elevated temperatures while pumping, and the use of sensitive instruments for determining the residual levels of contaminants during the pump-down.

The first step in setting up a test was to braze a cold terminal to one end of the PbTe element. The terminal was a composite structure consisting of stainless steel covered with a thin layer of molybdenum; the lead telluride was brazed to the molybdenum using tin telluride in a process somewhat similar to that described by Weinstein and Mlavsky⁽³⁾ and patented by Ritchie⁽⁴⁾. Thermocouples to measure the cold junction temperatures were spot welded to the stainless steel terminals and current leads were also attached as shown in Figure 4. The two elements to be tested were set up in a spring-loaded jig shown in Figure 5. The lava block at the bottom of the assembly contained springs and followers which operated against the steel cold terminals to press the opposite ends of the elements against an Armco iron block containing a commercial electric cartridge heater in a hole along its long axis. The thermoelements were not directly in contact with iron, however; the iron block was recessed to allow the insertion of various materials for the hot junction contacts. The inserts used in one of our tests on this program were molybdenum (Mo) and two were tungsten (W); in the other three tests cups of one of these two metals were filled with tin telluride (SnTe) and then ground flush with the surrounding metal surface. The tie rods and end plates of the assembly are of stainless steel, and the end plates are thermally and electrically isolated by Lava disks.

One of the critical features of the preparation of a test was the outgassing of the electric heater, which contains parts made of underfired lava as well as partially sealed internal air spaces and can be a source of comparatively large amounts of air and water vapor. It was necessary to bake these heaters out in vacuum at about 900°C before use to insure that they would not evolve significant amounts of gas in operation at 500°C to 600°C. In order to get all of the gas out in a reasonable length of time, it was also necessary to drill a hole in the side of the heater, communicating with the dead space inside. A matching hole, visible in Figure 5, was drilled in the side of the heater block.

Figure 6 shows a jig (an earlier model than that shown in Figure 5, but mounted in the same way) mounted on the base of one of our test cells and with all of its electrical connections in place. The test cell base is a stainless steel ultrahigh vacuum flange with 18 bakeable, ceramic-to-metal sealed electrical feedthroughs welded into it. Two of the feedthroughs have 1/4" diameter conductors of electric current and heat from the thermocouple, and the jig is mounted on them; the holes for the studs can be seen in the cut-down shoulders of the lava block in the jig shown in Figure 5, and the nuts on the top ends of the studs are visible in Figure 6.

The other feedthroughs in the base have smaller conductors and are arranged in a circle; there are four nickel conductors used for heater connections and six chromel and six alumel conductors used for connections to temperature measuring thermocouples. All connections to these feedthroughs were welded. The individual element hot junction temperatures were measured by chromel-alumel couples which were protected by stainless steel sheaths and were inserted in wells drilled in the heater block. A separate, unsheathed, chromel-alumel couple was provided for temperature control by spot welding heavy-gauge wires to the iron block.

After the assembly was brought to the stage shown in Figure 6, the space around the jig was stuffed with washed (i.e., 100% fibrous) Fiberfrax thermal insulation which contains no organic binders and had previously been baked out under vacuum at 900°C. A cylindrical stainless steel cover was then placed over the assembly and sealed to the base by a mating flange and copper gasket, and the complete test cell, mounted on an ultrahigh vacuum system through a bakeable high conductance valve mounted on its top, is shown in Figure 7.

The test cell was pumped down and then baked out at 250°C while the composition of the gas pumped out of it was monitored by a mass spectrometer partial pressure analyzer. The oxygen level dropped quite rapidly, but reduction of water vapor to the background level for the system usually required a week or ten days. This is due to the low bakeout temperature; higher temperatures were impractical, however, because the copper gaskets very often softened and relaxed enough to create leaks in the seals. At the end of the bakeout procedure the pressure indicated by the discharge gauge was below 10^{-8} Torr.

After the bakeout the cell was cooled and backfilled with spectroscopic-grade argon gas; it was checked for leaks through the valve after it was closed, by means of the partial pressure analyzer. Then the cell was removed from the system and transferred to a helium-filled atmosphere glove box, where the aperture of the valve was closed off with a blank flange sealed by a copper gasket.

Instrumentation for a thermocouple test consisted of a standard type of pyrometer temperature controller attached to the heavy unsheathed thermocouple on the heater block and a digital voltmeter connected to the other instrumentation leads. Signal levels are such that all of the data taken contain a minimum of four significant figures. Figures 8A and 8B show two views of the group of 8 test cells, two of which had been running under an earlier Company-sponsored study.

The following measurements were made under equilibrium temperature conditions with the lead telluride thermocouple connected to an approximately matched external load and again on open circuit:

Electric current through the external circuit
Potential difference between thermoelement cold end caps (i.e.,
thermocouple output voltage)

Potential differences between heater block and thermoelement cold end caps
p-Type leg hot and cold end temperatures
n-Type leg hot and cold end temperatures

From the open and closed circuit potentiometer measurements listed above, the following quantities were calculated by computer:

External circuit resistance
Gross power delivered to external circuit
p- and n-Type average leg Seebeck Coefficients
EMF's developed by p- and n-type legs under closed circuit conditions
p- and n-Type leg resistances with matched load current flowing
Power which would be delivered to an exactly matched load under the closed-circuit temperature conditions (the correction for inexact load matching was usually less than 0.5%)
Power which would be developed in an exactly matched load under standard operating temperature conditions (this correction for variation of operating temperatures was usually 1 to 2%, and efforts were made to trim the temperatures to keep it within these limits)

The last quantity on the above list is referred to as "normalized power output." Since most effects due to variations of the test conditions were eliminated in the calculations, changes in the normalized power output indicate actual changes in the power-producing capacity of the thermocouple. No corrections were made to take into account the small effects due to the temperature dependence of the electrical properties of the lead telluride, however, and these contributed a noise level of about 0.5% to the curves of normalized power output vs. time.

Six test cells were placed in operation, two at a time, labeled as cells 13 through 18. The extruded rods used for the various cells were listed in Table III, with the room temperature values of Seebeck Coefficient and electrical resistivity. Also listed is the contact resistance between the PbTe rod and the cold end electrode, which was a stainless steel rivet with a molybdenum (or in one case iron) coating. The contact resistance was measured by moving one probe along the side of the PbTe rod and extrapolating the resistance curve to the contact position. The measured contact resistance in Table III is in micro-ohms. To convert to the usual expression for contact resistance, it is necessary to multiply by $\sim 0.16 \text{ cm}^2$, the contact area, which then gives $R_c \sim 3 \text{ to } 40 \text{ micro-ohm-cm}^2$. This contact resistance is in the low range of those reported by other investigators of PbTe contacts.

In the initial period of each test cell operation under a $\sim 210^\circ\text{C}$ - 550°C temperature gradient, data were recorded every day because of the relatively large changes in measured properties. In several cases, power output improved in the initial operating period, probably because the PbTe rods became better seated at the butt contacts near the iron heater block. After a few hundred hours of operation, measurements were only required about twice a week to get

enough data to provide smooth plots of the operating parameters. Use of a digital recording system and automatic terminal switching made it possible to record data on the six test cells in only a few minutes. The data were then fed by punched cards to a computer which provided a printout of the interesting parameters calculated for each cell.

The data for the test cells are listed in Tables VII through XII for cells 13 through 18, respectively. At the top of each of these Tables is given the hot terminal conditions. Thus for cell 13, the PbTe rods butted against SnTe held in Mo cups recessed in the iron heater block. For cell 14, there was SnTe in W cups. Cells 15 and 16 used Mo and W directly against the PbTe rods. Cells 17 and 18 both used W, one with and one without SnTe. These different hot terminal configurations were used to test whether Mo or W would lead to lower power degradation rates and to test the influence of SnTe.

In each of the tables of test cell data, the measured temperatures at each end of the extruded thermoelements are given for each time of data recording, together with the computed electrical data of Seebeck Coefficient, resistivity and normalized power output. Also listed is the power output relative to the peak power output, which occurred at onset of operation or shortly thereafter.

In order to see the trend of the data, the computed electrical quantities are plotted in Figures 9 through 19. Where overlap is not serious, data from several cells are plotted on the same figure to conserve space. During the operating periods of the six cells, there were two power outages which caused transient dips in power output in most of the cells and permanent degradation in cell 14. The time and duration of these power outages are listed in Table XIII, and also marked by short vertical arrows in Figures 9 through 19. The power outages occurred over weekends so that corrective action was not immediate. Because of past experience with sticking relays in the test cell heater power supply, all of the test cells had been supplied with an over-temperature safety circuit designed to prevent overheating of the PbTe thermoelements. Once a test cell had been stabilized with its 210°C-550°C gradient, the heater current was only applied intermittently, approximately 3 to 5 seconds at a time. The safety circuit was adjusted so that if the heater current remained on for longer than one to two minutes, then a relay was actuated and the power circuit opened. (The safety circuit was temporarily bypassed only for the initial setup of the temperature gradient when a careful watch was maintained to reach the desired gradient.) When the a.c. line power was interrupted in the laboratory building for about ten minutes, the test cells cooled appreciably. When power was restored, the heater current was maintained continuously to bring the heater block temperature back to 550°C. The over-temperature circuits sensed the long time duration of heater current and opened the heater circuits in most of the cells.

The power outages caused only short transient dips in power output in cells 13, 17, 18, and larger dips in cells 15 and 16, but power output recovered after a few hundred hours. A permanent large dip occurred in the

power output of cell 14, and after the second outage, cell 14 had dropped to less than 35% of its peak power. The dips in cell 14 were clearly related to large increases in the resistance of the n-type thermoelement, whereas the resistance of the p-type thermoelement and the Seebeck coefficients showed no such marked behavior at the times of the heater power outages. The probable explanation for the cell 14 behavior is the formation of internal cracks in the n-type thermoelement due to the temperature cycling.

The transient dips in power output for the other five cells were largely related to transient increases in thermoelement resistances, probably due to stress effects at the butted contacts arising from the differential expansion of the PbTe elements and the Mo or W inserts in the iron heater block. PbTe has an expansion coefficient of 19×10^{-6} whereas Mo and W are near 6×10^{-6} . The heater power outages were unfortunate and unplanned, but they were informative as to the cycling ability of spring-loaded contacts in our test cells.

The individual cell performances can be summarized as follows:

Cell 13 (Mo and SnTe at hot contacts) - fastest degradation of our set of six cells, dropping about 20% from its peak value in 3500 hours. Degradation is mostly due to the increase in p-type thermoelement resistance R_p , with smaller dips in Seebeck Coefficients S_p and S_n . The n-type thermoelement resistance R_n actually dropped during operation, offsetting to some extent the change in R_p .

Cell 14 (W and SnTe) - had the slowest degradation rate until permanently damaged by the power outages already described, which greatly increased R_n .

Cell 15 (Mo - no SnTe) - Moderate degradation, recovered from power outages slowly. R and S behavior rather like cell 13, largest change being increase in R_p .

Cell 16 (W - no SnTe) - Intermediate degradation rate to cells 13 and 15, with slow recovery from power outage. Again largest change was in R_p with relatively small changes in R_n , S_p , and S_n .

Cell 17 (W and SnTe) - Close to cell 13 in degradation rate, with small contributions from R_p , R_n , S_p , and S_n .

Cell 18 (W - no SnTe) - Intermediate degradation rate, with small shifts in all of the electrical parameters. R_n is dropping slowly offsetting the unfavorable changes in R_p , S_p , and S_n .

A study of the power output degradation for the six cells does not reveal a clearcut preference for Mo vs W or SnTe vs no SnTe at the hot junction (550°C) ends of the couples. A qualitative ranking of the test cells, taking account of effects of power outages, would be as follows:

<u>Degradation rate</u>	<u>Cell</u>	<u>Hot Electrode Construction</u>
Lowest 1.	14	W - SnTe
2.	15	Mo
3.	18	W
4.	16	W
5.	17	W - SnTe
Fastest 6.	13	Mo - SnTe

The use of Mo or W at the hot junction end is greatly superior to the use of iron (Fe) there, for under similar temperature gradient $\sim 210^{\circ}\text{C}$ - 550°C , our old cell 7 with Fe dropped over 25% in its first 1000 hours of operation whereas cells 13 to 18 range from 3 to 9% for 1000 hour operation.

Comparison of the R_p and R_n values for thermoelement resistances in the life test cells with the room temperature resistivities given in Table III shows a fair correlation; i.e., the elements with larger resistivities at room temperature showed the larger resistances averaged over the operating $\sim 210^{\circ}\text{C}$ - 550°C gradient. It is interesting to note that the greatest rate of increase of the p-type element resistance with elapsed time of operation occurred in cell 13 with the lowest element resistance, whereas for cells 17 and 18 with large element resistance, the element resistance changed more slowly or hardly at all. These changes in p-type element resistance can be ascribed to the indiffusion of donor impurities, for if the donor impurity diffuses in at the same rate in all our test cells, then the more lightly-doped elements (cells 17 and 18) will be driven close to intrinsic at the upper range of their 210°C - 550°C gradient. The more intrinsic an element, the lower its resistance will be at the high temperature end and this temperature region provides the dominant contribution to the overall resistance, as can be seen from the plot of resistivity versus temperature in Figure 3. Thus a lightly-doped p-type element will not increase as rapidly in resistance for a given addition of compensating donors as will a more heavily-doped element. Since the p-type elements in the various test cells differ by as much as 50% in initial element resistance, the degradation rate observed will be influenced by these differences, tending to mask possible differences between Mo and W.

The Mo and W were chosen primarily to act as diffusion barriers to the entry of iron donors into the elements at the 550°C ends, but our experiments do not clearly indicate which metal is the better barrier. It should be noted that most of the n-type elements showed a decrease in resistance with elapsed operating time, which could be attributed to iron donors diffusing into the elements and reducing their overall resistances. The compatibility tests have shown that large increases in p-type element resistances occur when stainless steel constituents such as Mn or Cr can be vapor-transported to a PbTe surface. Thus even if the Mo and W were completely impervious to iron, degradation could occur because of vapor transport of contaminating impurities within the cell and

diffusion into the PbTe elements, particularly near the 550°C end. Our other recent contract study on PbTe⁽²⁾ has shown that Mn and Cr share the donor activity already known for Fe.

VII. CONCLUSIONS

The rods fabricated by our hot extrusion process were shown to have the electrical properties of our earlier near-optimized hot-pressed rods and to be satisfactory for thermocouples to be used for high temperature power generation. Their single-phase composition and almost theoretical density should insure that degradation under high temperature conditions would arise only from external factors such as indiffusing impurities. The extruded rods were usable for our compatibility and performance tests after a crazed surface layer was removed. By use of a die lubricant such as Teflon, the surface quality of the extruded rod was greatly improved. However, Teflon was not satisfactory because of the variability of the electrical properties of the rods containing Teflon. Since the problem of surface crazing and deeper cracks is related to the sticking of the PbTe to the walls of the die during extrusion, an alternate approach to the use of lubricants would be to substitute another die material for the steel used in our dies. Steel was chosen initially because of its strength and machinability, but the use of Teflon lubricant showed that the die strength was not a major problem. It is possible that a ceramic die made of say, alumina, would be strong enough for the extrusion and yet provide a lubricating and non-reactive surface to guide the PbTe rod.

The trials with silver and samarium have demonstrated their possible use as acceptor and donor dopants for PbTe. Time did not permit a more detailed study to ascertain the stability of these dopants for high temperature thermocouples operation. Since iodine has a high vapor pressure, it may be difficult to control its uniformity in an autoclaved ingot, so that samarium is an attractive donor dopant to consider.

The compatibility tests have delineated some safe and some deleterious materials for close proximity to PbTe. Future life test cell designs should embody safe construction materials in the hot portions of the test cell through substitution for present iron or stainless steel parts. In addition, the vapor transport and attack of PbTe indicates that the active cell volume should be reduced to a minimum so that this contaminating mechanism can be suppressed.

The test cell data showed that Mo and W offer significant improvement over Fe as hot junction contact surfaces. The high temperature butt contacts in the spring-loaded arrangement were satisfactory for continuous operation, but were not very satisfactory when the temperature was cycled by unplanned power line failures. A brazed contact at both ends of the elements might solve this variable contact problem on cycling, but it introduces great rigidities in the test cell, which might be particularly troublesome if multiple couples are to be tested.

The observed degradation in power output of the test cells is largely related to an increase in the p-type element resistance. Since in most cells there was also a smaller decrease in the n-type element resistance, it is tempting to ascribe the degradation to the indiffusion of a donor impurity across the hot end contacts (through the Mo or W) or indiffusion of a donor impurity which is vapor transported from one of the hot regions of the cell,

especially the heater block and imbedded heater. Iron is the logical culprit for this externally caused degradation, but the compatibility tests show that other constituents of stainless steel such as Mn and Cr could contribute to the donor compensation of the p-type element. Only by constructing a new test cell without hot iron or stainless steel parts could this hypothesis for the degradation mechanism be checked, and the degradation rate possibly materially reduced. Our test cell data did not provide any clear answer as to whether W is superior to Mo at the hot contact. It made no material difference to the observed performance whether SnTe was present between the W or Mo and the PbTe rod.

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TABLE I

Comparison of Electrical Data on
Extruded and Hot Pressed Rods

<u>Type</u>	<u>Composition</u>	<u>Fabrication</u>	<u>Seebeck Coefficient $\mu\text{V}/\text{deg}$</u>	<u>Resistivity mohm-cm</u>
P	$\text{PbNa}_{0.006}\text{Te}_{1.004}$ or $\text{PbNa}_{0.0058}\text{Te}_{1.00363}$	Extrusion	+60-72	0.29-.4
P	$\text{PbNa}_{0.0058}\text{Te}_{1.00387}$	Hot Pressed	+69	0.32
N	$\text{Pb}_{1.001}\text{I}_{0.001}\text{Te}$	Extrusion	-100-180	0.37-0.6
N	$\text{Pb}_{1.001}\text{I}_{0.001}\text{Te}$	Hot Pressed	-81	0.42

Table II

Summary of Extrusion Runs

<u>Run</u>	<u>Material</u>	<u>Unusual Conditions</u>	<u>Surface Quality</u>	<u>Comments or Usage</u>
E-25	N135A	0.128" diameter	Good	
E-26	"		Fair	Density 8.262 gcm ⁻³
E-27	P116A	0.128" diameter	Fair	
E-28	"		Fair	
E-29	"		Fair	Density 8.23 gcm ⁻³ Compatibility Tests
E-30	N210		Poor	
E-31	"		Fair	Life Test Cell Elements
E-32	"		Fair +	Life Test Cell Elements
E-33	P116A	Argon	Poor	
E-34	P117		Poor	
E-35	P116A	Teflon wadding	Fair +	
E-36	"	Tapered die here on	Poor	
E-37	"	Tefcoat (Camie Co.)	Fair +	
E-38	"	Teflon (DuPont)	Good	Compatibility Tests
E-39	P117	Teflon (DuPont)	Good	Compatibility Tests Life Test Cell Elements
E-40	P117	Teflon (Vydex)	Poor	
E-41	N211	Teflon (Vydex)	Fair +	
E-42	PbSnTe			
E-43	P116A	Teflon (DuPont)	Good	
E-44	PAg2	Teflon (DuPont)	Good	
E-45	PSm7	Teflon (DuPont)	Good	
E-46	P116A	Lexan	Poor	

Usual Conditions: T ~350-400 C, Load ~8000-15000 lbs, Rate ~0.3-0.6 in/hr,
0.225" diameter, Length 4-6", Vacuum ~10 microns.

Material

P-116A (Autoclave)	PbNa _{.006} Te _{1.004}	N135A	Pb _{1.001} I _{.001} Te
P-117	PbNa _{.0058} Te _{1.0036}	N210 -	Pb _{1.001} I _{.001} Te
PAg2	PbAg _{.005} Te _{1.005}	N211	Pb _{1.001} I _{.001} Te
PSm-7	Pb _{.999} Sm _{.001} Te		

TABLE III

Room Temperature Electrical Data and
Usage of Extruded Rods
(0.225" diameter ground down to 0.18" diameter)

P-Type: Composition $\text{PbNa}_{0.006}\text{Te}_{1.004}$ except E-29, $\text{PbNa}_{0.0058}\text{Te}_{1.00363}$

<u>Extrusion Run</u>	<u>Seebeck Coefficient $\mu\text{V}/\text{deg}$</u>	<u>Resistivity mohm-cm</u>	<u>Usage of 0.4" length samples</u>	
E-5	+60-66	0.29-0.37		
	+64	0.29 No. 2	Cell 15	Rc 35 μohms
	+65	0.35 No. 3	Cell 16	10
E-10	+61-72	0.29-0.36		
	+61	0.36 No. 6	Cell 13	80
	+71	0.34 No. 5	Cell 14	85
E-29	+57-66	0.30-0.37	Compatibility tests - first series	
E-38	+51-150	0.31-0.52	Compatibility tests - second series	
E-39	+61-88	0.33-0.58	Compatibility tests - second series	
	+68	0.33 No. 5	Cell 17	100
	+72	0.47 No. 11	Cell 18	190

N-Type: Composition $\text{Pb}_{1.001}\text{I}_{0.001}\text{Te}$

E-31	-101 to -132	0.37-0.58		
	-105	0.44 No. 1	Cell 15	170
	-105	0.41 No. 2	Cell 16	35
	-108	0.58 No. B	Cell 17	85
	-103	0.45 No. D	Cell 18	20
E-32	- 71 to -250	0.39-0.66		
	-159	0.48 No. 6	Cell 13	195
	-176	0.52 No. 11	Cell 14	230

TABLE IV

Non-Sodium or Iodine Doped PbTe

<u>Rod</u>	<u>Composition</u>	<u>Initial Properties</u>	<u>After 700°C-1 Hour Anneal</u>
Hot Pressed PAg-1	PbAg _{0.01} Te _{1.005}	S = +200 ρ = 2	Data obtained prior to this contract
Extruded PAg-2 E-44	PbAg _{0.005} Te _{1.005}	S = +250 ρ = 4.0	S = +107 ρ = 0.66
Extruded PSm-7 E-45	Pb _{0.999} Sm _{0.001} Te	S = -114 ρ = 5.4	S = -95 ρ = 3.9

S - Seebeck Coefficient, Microvolts per Degree C.

 ρ - Electrical Resistivity, Milliohm - Cm.

TABLE V . RESULTS OF FIRST SERIES OF COMPATIBILITY TESTS AT 600°C

Sample Characteristics		Potential Contaminant In Tube	Electrical Properties ρ -milliohm-cm			Relative Stability
			Initial	After 1000 hrs	After Surface Grinding	
1)	E-10 #1	None	0.32	0.38	0.36	Little effect on ρ
2)	E-29 #12	None	0.33	0.48 cracks?	1.6	
3)	E-29 #2	Brazed with SnTe	0.34	0.35	0.34	
4)	E-29 #3	Fiberfrax	0.31	0.38	0.35	
5)	E-29 #6	N-PbTe	0.30	0.36	0.35	
6)	E-29 #1	Lava	0.32	0.32	0.39	
7)	E-29 #11	1 mm air	0.36	0.61	0.56	Doubling of ρ
8)	E-29 #8	Water	0.34	0.72	0.63	
9)	E-29 #4	Stainless steel	0.32	0.65	0.4-1.7 cracks?	
10)	E-29 #5	Thermocouple in Stainless steel	0.31	0.73	0.72	
11)	E-29 #9	Brazed to Mo-clad Stainless steel	0.34	1.52	1.44	4 to 10X Increase in ρ
12)	E-29 #10	Iron Heater block	0.37	1.82	1.25	
13)	P-90 #6	Brazed to Fe-clad Stainless steel	0.38	3.6	3.4	
E-10 PbNa _{.006} Te _{1.004} - P type extrusion						
E-29						
P-90 PbNa _{.005} Te _{1.005} - P type hot pressed						

TABLE VI . RESULTS OF SECOND SERIES OF COMPATIBILITY TESTS AT 600°C

	<u>Sample Identification</u>	<u>Potential Contaminant in Tube</u>	<u>Electricity Resistivity ρ</u> milliohm-cm		<u>Relative Stability</u>
			<u>Initial</u>	<u>After 1000 Hours</u>	
1)	E-38 #2	None	0.55	0.61	} Little or no change in resistivity
2)	E-38 #3	Ni	0.62	0.58	
3)	E-38 #9	Ta	0.38	0.45	
4)	E-38 #11	Mo	0.32	0.39	
5)	E-39 #9	W	0.48	0.58	
6)	E-39 #10 (not ground down)	Teflon Coating	0.33	0.62	} Approximate doubling of resistivity
7)	E-39 #8	Stainless Steel	0.49	1.14	
8)	E-39 #2	Ag	0.47	— Reaction with sample - surfaces eroded.	
9)	E-38 #7	Cr	0.37	— Reaction with sample - surfaces eroded.	
10)	E-38 #8	Mn	0.37	— PbTe almost gone - very extensive reaction with Mn.	

TABLE VII

TEST CELL NO. 13 DATA (Mo and SnTe)

Elapsed Time (Hours)	Temperatures (°C)		Ave. Seebeck Coefficients		Resistances		Power Output P($\Delta T=340^{\circ}\text{C}$)	Relative to .1274
	P leg	N leg	S_p	S_n	R_p	R_n		
26	549.5 - 213.0	550.0 - 207.3	236.1	231.6	28.1	21.7	0.1269	0.9961
50	549.0 - 210.0	550.5 - 204.9	235.8	231.9	28.3	21.3	0.1274	1.0000
68	548.2 - 211.2	550.2 - 206.1	235.2	231.7	28.3	21.2	0.1273	0.9992
92	547.2 - 212.2	549.4 - 207.1	235.1	231.8	28.5	21.1	0.1268	0.9953
170	547.2 - 211.1	549.2 - 206.1	233.7	230.9	28.9	20.6	0.1258	0.9875
194	547.0 - 212.0	549.4 - 207.1	233.7	231.1	29.0	20.6	0.1256	0.9859
219	547.5 - 212.4	549.6 - 207.2	233.7	231.0	29.3	20.6	0.1247	0.9790
242	547.1 - 212.4	549.1 - 207.1	233.3	230.6	29.4	20.5	0.1245	0.9773
266	547.5 - 210.8	549.2 - 205.4	232.3	230.4	29.3	20.4	0.1242	0.9748
337	547.2 - 210.1	549.1 - 204.8	231.6	229.9	29.6	20.2	0.1233	0.9679
361	547.7 - 210.6	549.2 - 205.3	231.7	229.9	29.8	20.2	0.1229	0.9649
387	547.2 - 209.0	548.7 - 203.6	231.2	229.6	29.8	20.1	0.1228	0.9636
427	547.1 - 209.4	548.7 - 204.0	230.6	229.5	29.9	20.1	0.1222	0.9591
505	547.9 - 209.7	549.7 - 204.2	230.2	229.2	30.2	20.1	0.1212	0.9516
526	548.1 - 211.5	550.2 - 205.9	230.8	229.2	30.5	20.1	0.1206	0.9463
548	547.6 - 212.5	549.8 - 206.9	230.7	229.2	30.5	20.1	0.1205	0.9457
576	547.6 - 211.8	549.6 - 206.3	230.5	229.1	30.6	20.0	0.1204	0.9451
602	547.9 - 212.7	550.2 - 207.0	230.5	229.2	30.7	20.1	0.1199	0.9415
669	548.0 - 212.4	550.2 - 206.8	230.0	228.7	30.9	20.0	0.1193	0.9362
693	547.9 - 213.2	550.2 - 207.7	230.2	229.1	31.0	20.1	0.1191	0.9349
740	547.4 - 212.6	549.4 - 207.0	229.5	228.1	31.1	19.8	0.1187	0.9316
767	547.8 - 212.5	550.0 - 207.0	229.3	228.6	31.1	20.0	0.1185	0.9299
835	546.9 - 212.7	549.2 - 207.3	228.9	228.3	31.2	19.8	0.1182	0.9277
867	547.3 - 212.5	549.6 - 206.9	228.9	228.4	31.4	19.9	0.1177	0.9235
889	547.1 - 212.4	549.2 - 206.9	228.5	228.2	31.4	19.8	0.1176	0.9228
933	547.8 - 212.6	550.1 - 207.2	228.4	228.3	31.6	19.9	0.1169	0.9178
1004	548.1 - 212.7	550.1 - 207.2	227.9	228.1	31.7	19.9	0.1163	0.9133
1056	547.5 - 212.6	549.7 - 207.2	227.8	227.8	31.8	19.8	0.1161	0.9112
1104	548.1 - 212.9	550.2 - 207.3	227.5	227.8	32.0	19.8	0.1156	0.9071
1178	548.0 - 212.1	550.0 - 206.5	227.7	227.5	32.0	19.7	0.1150	0.9030
1226	548.5 - 212.5	550.5 - 206.9	226.7	227.4	32.2	19.7	0.1147	0.9003

TABLE VII TEST CELL NO. 13 DATA (Mo and SnTe)

Elapsed Time (Hours)	Temperatures (°C)		Ave. Seebeck Coefficients		Resistances		Power Output P(ΔT=340°C)	Relative to .1274
	P leg	N leg	S _p	S _n	R _p	R _n		
1274	548.5 - 212.3	550.8 - 206.6	226.3	227.5	32.2	19.8	0.1143	0.8970
1344	548.6 - 212.4	550.7 - 206.8	226.0	227.4	32.4	19.7	0.1138	0.8930
1394	548.8 - 212.3	550.8 - 206.8	225.7	227.2	32.5	19.7	0.1135	0.8907
1441	548.5 - 211.8	550.4 - 206.4	225.3	226.9	32.6	19.6	0.1130	0.8871
1514	549.0 - 212.8	551.0 - 207.3	225.2	227.1	32.8	19.7	0.1125	0.8834
1562	548.9 - 212.0	550.9 - 206.4	224.6	226.8	32.8	19.7	0.1121	0.8802
1610	549.1 - 211.8	551.4 - 206.3	224.5	226.7	32.9	19.7	0.1119	0.8783
1681	548.9 - 212.7	551.5 - 207.3	224.6	226.9	33.0	19.7	0.1116	0.8761
1730	548.1 - 211.9	550.1 - 206.5	223.9	226.5	33.1	19.6	0.1113	0.8732
1844	548.1 - 211.4	550.3 - 206.0	223.2	226.3	33.2	19.6	0.1105	0.8677
1892	548.1 - 211.9	550.1 - 206.7	223.3	226.3	33.3	19.5	0.1105	0.8670
1940	548.8 - 211.7	550.8 - 206.3	222.7	226.4	33.4	19.6	0.1099	0.8628
1973	548.9 - 210.4	551.4 - 205.6	217.8	222.8	32.8	19.3	0.1077	0.8456
2022	548.5 - 210.3	551.0 - 205.4	221.4	225.4	33.6	19.7	0.1083	0.8498
2069	549.1 - 210.9	551.7 - 206.4	221.6	225.3	33.2	19.3	0.1100	0.8633
2142	547.6 - 210.7	550.4 - 205.9	221.4	225.5	33.8	19.6	0.1079	0.8470
2191	549.0 - 209.6	551.3 - 204.8	220.7	225.5	33.9	19.7	0.1073	0.8420
2239	549.4 - 210.4	551.8 - 204.9	220.4	225.3	34.0	19.6	0.1071	0.8405
2312	548.5 - 209.8	550.6 - 204.4	220.1	225.0	34.0	19.4	0.1071	0.8407
2360	548.8 - 209.6	551.0 - 204.1	219.7	224.9	34.0	19.5	0.1068	0.8384
2407	549.0 - 209.6	551.3 - 204.0	219.5	224.9	34.1	19.5	0.1064	0.8355
2479	549.2 - 211.1	551.3 - 205.8	219.6	225.1	34.3	19.5	0.1063	0.8345
2528	548.9 - 210.6	551.3 - 205.1	219.2	225.0	34.3	19.5	0.1060	0.8324
2575	549.1 - 211.4	551.3 - 206.1	219.3	225.3	34.4	19.5	0.1059	0.8314
2647	548.3 - 210.3	550.6 - 205.0	218.8	224.9	34.4	19.5	0.1057	0.8294
2738	548.6 - 211.0	550.9 - 205.6	218.6	224.7	34.5	19.4	0.1053	0.8265
2767	548.8 - 209.6	551.3 - 204.5	217.9	224.3	34.6	19.6	0.1043	0.8186
2810	548.9 - 208.7	551.2 - 203.9	217.7	224.5	34.6	19.5	0.1044	0.8198
2860	548.9 - 209.4	551.1 - 204.4	217.6	224.7	34.6	19.5	0.1044	0.8198
2931	549.4 - 209.1	551.5 - 204.2	217.4	224.5	34.7	19.4	0.1042	0.8182

TABLE VII

TEST CELL NO. 13 DATA (Mo and SnTe)

Elapsed Time (Hours)	Temperatures (°C)		Ave. Seebeck Coefficients		Resistances		Power Output $P(\Delta T=340^{\circ}\text{C})$	Relative to .1274
	P Leg	N Leg	S_p	S_n	R_p	R_n		
3033	549.3 - 209.0	551.3 - 204.1	217.0	224.5	34.8	19.4	0.1038	0.8149
3124	549.0 - 209.3	551.6 - 204.5	216.8	224.5	34.9	19.5	0.1036	0.8131
3201	549.2 - 209.4	551.6 - 204.5	216.5	224.4	34.9	19.5	0.1032	0.8103
3268	549.1 - 209.3	551.7 - 204.4	216.5	224.3	34.9	19.4	0.1033	0.8111
3364	549.1 - 209.5	551.3 - 204.7	216.2	224.1	35.1	19.3	0.1029	0.8079
3438	548.5 - 209.7	551.1 - 204.8	216.0	224.1	35.0	19.3	0.1028	0.8071

TABLE VIII TEST CELL NO. 14 DATA (W and SnTe)

Elapsed Time (Hours)	Temperatures (°C)				Ave. Seebeck Coefficients		Resistances		Power Output P(ΔT=340°C)	Relative to 0.1182
	P leg		N leg		S _p	S _n	R _p	R _n		
2	542.0	- 194.3	542.6	-194.9	239.7	224.7	31.8	19.6	0.1209	-
21	541.5	- 195.5	542.0	-196.1	240.8	226.1	34.0	18.2	0.1203	-
26	548.0	- 205.6	549.0	-206.2	243.6	228.0	35.2	18.9	0.1182	1.0000
98	548.0	- 206.6	548.8	-207.1	244.9	228.0	36.8	19.0	0.1156	0.9780
122	547.8	- 207.2	548.5	-207.6	245.0	228.2	36.8	19.0	0.1155	0.9772
147	548.0	- 206.8	548.6	-207.5	244.8	228.1	36.7	19.0	0.1157	0.9788
171	547.8	- 206.8	548.5	-207.4	244.8	228.0	36.7	19.1	0.1156	0.9778
195	548.5	- 206.6	549.0	-207.4	244.5	227.9	36.5	19.1	0.1158	0.9798
266	547.4	- 206.8	548.0	-207.5	244.1	227.5	35.8	19.0	0.1169	0.9888
290	548.0	- 207.7	548.6	-208.3	244.1	227.7	35.8	19.2	0.1167	0.9876
315	547.3	- 206.7	547.9	-207.3	243.7	227.3	35.5	19.1	0.1172	0.9915
356	546.8	- 206.9	547.3	-207.6	242.9	227.2	35.1	19.1	0.1174	0.9933
434	547.7	- 207.3	548.2	-207.9	242.4	227.1	34.9	19.2	0.1175	0.9938
455	547.9	- 206.9	548.6	-207.5	242.2	226.8	34.8	19.2	0.1173	0.9927
477	547.5	- 207.2	548.2	-207.9	241.9	226.9	34.7	19.2	0.1173	0.9927
505	547.6	- 206.7	548.1	-207.5	241.6	226.8	34.6	19.2	0.1176	0.9951
531	547.7	- 207.4	548.4	-208.1	241.6	226.8	34.6	19.2	0.1175	0.9941
597	548.4	- 207.1	548.9	-207.9	240.8	226.5	34.4	19.3	0.1174	0.9928
622	548.0	- 207.4	548.5	-208.2	240.6	226.5	34.3	19.2	0.1175	0.9941
669	547.1	- 206.9	547.7	-207.6	240.2	226.4	34.2	19.3	0.1174	0.9931
696	547.7	- 207.3	548.3	-208.1	240.0	226.2	34.2	19.2	0.1172	0.9912
764	546.8	- 206.9	547.2	-207.7	239.4	226.0	34.0	19.2	0.1172	0.9912
795	547.8	- 207.6	548.3	-208.2	239.5	226.0	34.1	19.3	0.1168	0.9878
817	547.5	- 207.5	548.0	-208.1	239.0	225.7	34.0	19.3	0.1168	0.9881
861	548.0	- 207.7	548.3	-208.4	239.1	225.7	34.1	19.3	0.1165	0.9856
933	548.3	- 207.8	548.9	-208.5	238.4	225.6	34.0	19.4	0.1161	0.9824
985	547.7	- 207.4	548.3	-208.2	238.2	225.5	34.0	19.3	0.1162	0.9832
1033	547.4	- 207.2	548.0	-207.9	237.8	225.1	33.9	19.3	0.1160	0.9817
1107	547.4	- 207.1	548.1	-207.9	237.9	224.8	33.9	19.3	0.1158	0.9794
1155	547.3	- 206.7	547.8	-207.5	237.1	224.9	33.9	19.3	0.1156	0.9783

TABLE VIII TEST CELL NO. 14 (W and SnTe)

Elapsed Time (Hours)	Temperatures (°C)		Ave. Seebeck Coefficients		Resistances		Power Output P($\Delta T=340^{\circ}\text{C}$)	Relative to 0.1182
	P Leg	N Leg	S _p	S _n	R _p	R _n		
1203	547.3 - 206.9	547.8 - 207.6	236.7	224.8	33.9	19.3	0.1153	0.9753
1272	547.2 - 207.0	547.5 - 207.6	236.4	224.5	33.9	19.3	0.1149	0.9721
1322	547.6 - 207.0	548.2 - 207.8	236.2	224.4	34.0	19.3	0.1146	0.9697
1370	547.3 - 206.7	547.5 - 207.4	235.8	224.3	33.9	19.2	0.1147	0.9702
1442	548.1 - 207.5	548.4 - 208.3	235.6	224.4	34.1	19.3	0.1141	0.9652
1491	547.5 - 206.8	547.9 - 207.6	235.2	224.0	34.0	19.3	0.1138	0.9631
1539	547.7 - 206.7	547.9 - 207.4	235.1	223.9	34.1	19.3	0.1136	0.9609
1609	547.6 - 206.6	548.1 - 207.7	234.7	223.8	34.1	19.3	0.1133	0.9588
1658	548.1 - 207.2	548.6 - 208.0	234.6	223.7	34.2	19.3	0.1129	0.9553
1772	547.5 - 206.9	547.7 - 207.7	233.9	223.4	34.3	19.3	0.1125	0.9515
1820	547.3 - 207.0	547.8 - 207.9	234.1	223.6	34.3	19.4	0.1123	0.9504
1869	548.8 - 207.0	549.1 - 207.8	233.7	223.4	34.4	19.4	0.1118	0.9456
1902	549.4 - 207.2	550.0 - 208.0	232.8	222.0	34.8	19.8	0.1091	0.9228
1951	548.4 - 206.2	548.8 - 207.1	233.0	222.8	35.0	20.6	0.1075	0.9097
1998	549.9 - 207.4	550.2 - 208.0	233.0	222.9	35.2	21.2	0.1062	0.8983
2071	547.8 - 206.8	548.2 - 207.6	232.8	222.7	35.3	21.8	0.1047	0.8857
2120	549.2 - 206.0	549.4 - 206.6	232.2	222.1	35.4	22.2	0.1034	0.8749
2167	549.5 - 205.8	549.8 - 206.8	232.1	222.3	35.5	22.5	0.1026	0.8679
2240	548.6 - 205.9	549.0 - 206.6	231.8	222.1	35.4	23.0	0.1017	0.8601
2289	549.0 - 205.1	549.4 - 205.9	231.5	222.0	35.6	23.3	0.1007	0.8519
2335	549.6 - 205.4	549.8 - 206.1	231.2	222.0	35.6	23.5	0.1001	0.8473
2408	549.1 - 206.0	549.5 - 206.8	231.3	221.9	35.8	23.9	0.0992	0.8395
2456	549.1 - 205.8	549.3 - 206.5	230.9	221.9	35.8	24.1	0.0988	0.8356
2504	549.3 - 206.4	549.5 - 207.2	230.7	221.9	35.8	24.3	0.0981	0.8302
2575	548.7 - 205.8	548.8 - 206.6	230.7	221.5	35.9	24.5	0.0975	0.8249

TABLE VIII

TEST CELL NO. 14 DATA (W and SnTe)

Elapsed Time (Hours)	Temperatures (°C)		Ave. Seebeck Coefficients		Resistances		Power Output $P(\Delta T=340^{\circ}\text{C})$	Relative to 0.1182
	P Leg	N Leg	S_p	S_n	R_p	R_n		
2667	548.8 - 206.4	549.3 - 207.2	230.5	221.4	36.0	25.0	0.0965	0.8163
2742	548.8 - 203.5	549.3 - 204.6	230.0	221.4	36.6	55.8	0.0636	0.5383
2785	549.0 - 203.1	549.5 - 204.1	229.8	221.3	36.7	63.2	0.0588	0.4975
2836	548.9 - 203.0	549.4 - 204.2	229.6	221.3	36.7	70.9	0.0545	0.4613
2907	549.3 - 202.5	550.0 - 203.7	229.5	221.1	36.8	80.8	0.0498	0.4213
3009	549.1 - 201.9	549.8 - 203.2	229.1	221.0	37.0	91.6	0.0452	0.3826
3100	549.4 - 202.3	549.7 - 203.7	228.9	221.0	37.2	93.2	0.0443	0.3746
3244	549.5 - 202.6	550.1 - 203.9	228.4	221.1	37.5	88.7	0.0462	0.3909
3339	549.3 - 203.2	549.9 - 204.4	228.3	220.9	37.6	80.4	0.0493	0.4174
3413	549.2 - 203.5	549.5 - 204.8	227.9	220.7	37.7	74.8	0.0516	0.4365

TABLE IX

TEST CELL NO. 15 DATA (Mo ELECTRODE-NO SnTe)

Elapsed Time (Hours)	Temperatures (°C)		Ave. Seebeck Coefficients		Resistances		Power Output P ($\Delta T=340^{\circ}\text{C}$)	Relative to 0.1217
	P leg	N leg	S _p	S _n	R _p	R _n		
18	549.6 - 212.6	547.9 - 206.0	229.1	241.7	31.0	21.7	0.1215	-
22	548.3 - 213.2	546.9 - 206.4	229.2	241.6	30.8	21.8	0.1217	-
66	547.8 - 214.7	546.0 - 207.7	229.3	241.7	31.5	21.1	0.1216	-
137	547.7 - 215.7	546.6 - 208.5	229.8	241.7	32.3	20.9	0.1207	-
163	548.1 - 215.8	546.8 - 208.5	229.7	241.5	32.5	20.7	0.1205	0.9899
165	547.9 - 216.0	546.5 - 208.8	229.5	241.3	32.3	20.6	0.1210	0.9943
190	547.7 - 215.9	546.4 - 208.6	229.7	241.6	33.6	20.7	0.1202	0.9879
214	547.7 - 215.7	546.4 - 208.5	229.5	241.2	32.7	20.5	0.1203	0.9887
237	548.2 - 215.8	546.5 - 208.4	229.4	241.4	32.7	20.5	0.1203	0.9882
311	547.5 - 216.2	546.2 - 208.6	229.4	241.1	33.3	20.4	0.1193	0.9799
335	548.0 - 216.0	546.6 - 208.4	229.2	241.3	33.2	20.4	0.1194	0.9814
360	547.5 - 216.0	546.0 - 208.4	229.4	240.9	33.2	20.2	0.1198	0.9847
384	547.3 - 216.1	546.0 - 208.5	229.5	241.1	33.4	20.2	0.1193	0.9803
407	547.5 - 216.1	546.0 - 208.4	229.4	240.9	33.1	20.2	0.1200	0.9858
477	547.4 - 216.2	546.0 - 208.6	229.1	240.6	33.5	20.0	0.1191	0.9789
499	547.8 - 216.1	546.2 - 208.5	229.1	240.8	33.6	20.1	0.1189	0.9767
527	547.7 - 216.5	546.3 - 208.9	229.5	240.6	33.6	20.0	0.1192	0.9792
551	547.5 - 216.0	546.1 - 208.4	229.2	240.6	33.6	20.0	0.1191	0.9782
574	547.9 - 216.3	546.5 - 208.7	229.2	240.3	33.9	19.9	0.1185	0.9735
647	548.1 - 216.6	546.5 - 208.9	229.2	240.5	34.2	19.9	0.1179	0.9684
671	547.3 - 216.3	545.9 - 208.4	229.1	240.4	34.0	19.9	0.1183	0.9719
695	547.2 - 215.8	545.7 - 208.2	229.0	240.2	34.0	19.8	0.1183	0.9720
743	547.7 - 215.9	546.3 - 208.1	229.0	240.0	34.2	19.8	0.1178	0.9678
814	546.6 - 215.2	545.0 - 207.6	228.9	239.6	34.5	19.6	0.1173	0.9639
904	546.2 - 215.4	545.0 - 207.4	229.0	239.7	34.4	19.6	0.1175	0.9656
977	546.2 - 215.5	545.0 - 207.5	229.0	239.3	34.7	19.5	0.1169	0.9607
1025	545.6 - 215.5	544.2 - 207.4	229.1	239.3	34.8	19.5	0.1166	0.9583
1073	546.0 - 215.3	544.7 - 207.5	228.8	239.3	34.9	19.5	0.1163	0.9554
1106	545.6 - 214.1	544.3 - 206.4	226.3	238.2	38.7	19.6	0.1070	0.8790
1155	544.6 - 213.1	543.4 - 205.4	227.6	238.4	37.0	19.4	0.1112	0.9140
1202	544.2 - 213.2	543.1 - 205.6	227.6	238.4	37.1	19.4	0.1111	0.9132
1275	543.5 - 213.9	542.2 - 206.4	227.7	238.6	37.2	19.4	0.1110	0.9117

TABLE IX

TEST CELL NO. 15 DATA (Mo ELECTRODE-NO SnTe)

Elapsed Time (Hours)	Temperatures (°C)		Ave. Seebeck Coefficients		Resistances		Power Output P ($\Delta T=340^{\circ}\text{C}$)	Relative to 0.1217
	P leg	N leg	S_p	S_n	R_p	R_n		
1324	544.5 - 213.6	543.4 - 205.7	227.6	238.4	36.8	19.4	0.1117	0.9178
1372	544.4 - 213.4	543.4 - 205.5	227.6	238.3	37.0	19.3	0.1113	0.9149
1445	544.6 - 213.6	543.2 - 205.7	227.5	238.3	37.2	19.3	0.1109	0.9116
1493	544.9 - 213.2	543.6 - 205.3	227.3	238.3	37.1	19.3	0.1112	0.9136
1539	544.7 - 213.1	543.5 - 205.1	227.2	238.2	37.0	19.3	0.1111	0.9133
1613	543.7 - 213.1	542.7 - 205.3	227.2	238.0	37.0	19.2	0.1113	0.9146
1661	543.6 - 213.0	542.3 - 205.2	227.3	238.0	37.2	19.1	0.1110	0.9124
1708	543.7 - 213.8	542.5 - 205.8	227.4	238.0	37.1	19.1	0.1112	0.9140
1780	543.9 - 213.4	542.6 - 205.6	227.3	238.0	37.3	19.1	0.1108	0.9108
1871	544.2 - 213.9	542.9 - 206.1	227.2	238.1	37.7	19.1	0.1100	0.9041
1947	544.1 - 213.3	543.2 - 205.6	227.3	238.0	38.4	19.1	0.1090	0.8954
1990	543.9 - 213.2	542.7 - 205.3	226.9	237.8	37.8	19.0	0.1099	0.9031
2040	542.5 - 213.0	541.5 - 205.1	227.0	237.8	37.7	19.0	0.1100	0.9037
2111	543.4 - 213.0	542.1 - 205.2	226.8	237.6	37.9	18.9	0.1096	0.9006
2213	544.6 - 213.3	543.4 - 205.3	226.9	237.7	38.4	19.0	0.1087	0.8933
2304	543.9 - 213.3	543.0 - 205.7	226.7	237.6	38.4	19.0	0.1086	0.8927
2381	544.0 - 213.6	542.9 - 205.6	226.8	237.8	38.5	19.0	0.1084	0.8911
2448	544.4 - 213.7	543.5 - 206.0	226.6	237.7	38.4	18.9	0.1087	0.8934
2544	543.9 - 213.6	542.9 - 205.7	226.5	237.4	38.6	18.9	0.1082	0.8892
2618	544.2 - 213.9	543.3 - 206.5	226.6	237.9	38.8	19.0	0.1079	0.8862

TABLE X TEST CELL NO. 16 DATA (W ELECTRODE-NO SnTe)

Elapsed Time (Hours)	Temperatures (°C)		Ave. Seebeck Coefficients		Resistances		Power Output P ($\Delta T=340^{\circ}\text{C}$)	Relative to 0.1255
	P leg	N leg	S _p	S _n	R _p	R _n		
2	564.4 - 209.7	566.1 - 206.0	236.3	245.4	35.2	23.4	0.1181	-
23	548.5 - 205.7	550.0 - 201.8	233.4	243.2	31.3	21.0	0.1252	0.9984
25	548.4 - 211.9	550.1 - 208.1	235.3	244.0	31.5	21.2	0.1255	1.0000
91	548.0 - 213.8	549.8 - 209.8	235.6	243.5	32.3	20.5	0.1254	0.9992
122	547.2 - 214.0	549.0 - 210.0	235.8	243.2	32.4	20.3	0.1256	1.0008
144	548.8 - 214.7	550.3 - 210.5	235.4	243.1	32.6	20.2	0.1250	0.9960
188	549.7 - 215.1	551.1 - 210.9	235.8	242.8	33.0	20.0	0.1246	0.9928
260	549.3 - 215.2	551.0 - 211.0	235.7	242.4	33.4	19.8	0.1239	0.9876
285	549.6 - 215.1	551.3 - 210.8	235.2	242.0	33.6	19.8	0.1230	0.9801
312	548.7 - 214.2	550.3 - 210.4	235.1	242.0	33.5	19.5	0.1238	0.9861
336	549.4 - 214.0	551.0 - 210.2	235.5	242.3	33.9	19.7	0.1225	0.9762
359	548.2 - 213.5	549.8 - 209.8	234.9	241.9	33.8	19.5	0.1231	0.9809
433	548.4 - 213.6	549.9 - 210.1	234.8	241.8	34.3	19.4	0.1220	0.9723
457	548.8 - 213.5	550.4 - 209.7	235.0	241.9	34.5	19.5	0.1216	0.9691
481	548.7 - 213.5	550.0 - 209.9	234.9	241.6	34.6	19.3	0.1215	0.9680
506	548.6 - 213.5	550.1 - 209.8	234.9	241.7	34.7	19.4	0.1211	0.9652
530	548.4 - 213.4	549.8 - 209.7	234.7	241.5	34.8	19.3	0.1210	0.9638
599	548.3 - 213.4	549.6 - 209.8	234.8	241.4	35.2	19.2	0.1202	0.9580
621	548.6 - 213.4	550.0 - 209.8	234.6	241.4	35.3	19.2	0.1199	0.9557
650	548.5 - 213.4	549.7 - 209.7	234.6	241.4	35.4	19.2	0.1197	0.9534
673	548.2 - 213.1	549.5 - 209.6	234.3	241.3	35.4	19.1	0.1195	0.9524
697	548.4 - 213.1	549.7 - 209.5	234.5	241.2	35.6	19.1	0.1192	0.9500
769	548.9 - 213.6	550.0 - 210.0	234.4	241.1	35.9	19.0	0.1186	0.9452
793	548.5 - 213.3	549.7 - 209.7	234.3	241.1	35.9	19.1	0.1185	0.9438
818	548.3 - 213.2	549.6 - 209.5	234.2	241.0	36.0	19.0	0.1183	0.9430
865	548.8 - 213.2	550.0 - 209.5	234.4	241.1	36.2	19.0	0.1180	0.9401
936	548.3 - 212.7	549.4 - 209.2	233.8	240.7	36.3	18.9	0.1175	0.9366

TABLE X TEST CELL NO. 16 DATA (W ELECTRODE-NO SnTe)

Elapsed Time (Hours)	Temperatures (°C)		Ave. Seebeck Coefficients		Resistances		Power Output P ($\Delta T=340^{\circ}\text{C}$)	Relative to 0.1255
	P leg	N leg	S _p	S _n	R _p	R _n		
985	547.1 - 212.3	548.1 - 208.7	233.8	240.6	36.5	18.8	0.1172	0.9337
1027	547.9 - 212.3	548.3 - 208.8	234.0	240.6	36.8	18.8	0.1167	0.9301
1099	547.2 - 212.5	548.7 - 208.9	233.7	240.5	36.9	18.8	0.1163	0.9270
1147	547.2 - 212.5	548.2 - 208.8	233.9	240.7	37.1	18.8	0.1161	0.9248
1195	547.2 - 212.4	548.4 - 208.8	233.5	240.5	37.2	18.7	0.1158	0.9230
1229	547.2 - 211.6	548.3 - 208.0	231.1	239.5	42.4	19.4	0.1033	0.8229
1278	546.7 - 211.2	547.9 - 207.5	232.5	240.0	40.4	19.4	0.1077	0.8578
1325	546.8 - 211.5	548.0 - 207.9	232.5	239.9	40.0	19.3	0.1084	0.8637
1398	545.5 - 211.9	546.8 - 208.3	232.8	240.1	39.7	19.3	0.1093	0.8707
1447	547.0 - 211.2	548.3 - 207.5	232.2	239.8	39.5	19.3	0.1093	0.8706
1494	546.8 - 211.3	548.3 - 207.5	232.3	239.7	39.4	19.3	0.1095	0.8727
1567	546.2 - 211.3	547.6 - 207.6	232.1	239.7	39.4	19.2	0.1094	0.8715
1615	546.9 - 211.0	548.3 - 207.2	232.0	239.6	39.4	19.2	0.1094	0.8719
1662	547.0 - 211.2	548.6 - 207.4	231.7	239.4	39.3	19.2	0.1093	0.8712
1735	546.2 - 211.2	547.7 - 207.4	231.6	239.3	39.3	19.1	0.1093	0.8710
1783	546.2 - 210.8	547.6 - 207.1	231.6	239.5	39.3	19.2	0.1094	0.8719
1830	546.4 - 211.4	547.6 - 207.6	231.6	239.5	39.4	19.2	0.1093	0.8707
1902	546.0 - 211.6	547.3 - 207.8	231.5	239.3	39.4	19.1	0.1093	0.8706
1994	546.3 - 212.0	547.4 - 208.1	231.6	239.2	39.6	19.1	0.1087	0.8660
2069	546.3 - 211.9	547.9 - 207.9	231.7	239.4	38.2	19.4	0.1111	0.8852
2112	546.4 - 211.7	548.0 - 207.7	231.2	239.2	37.8	19.3	0.1117	0.8900
2162	546.3 - 211.9	547.6 - 207.8	231.5	239.2	38.0	19.3	0.1113	0.8871
2233	546.6 - 211.4	547.8 - 207.4	231.0	239.2	38.1	19.3	0.1110	0.8841
2335	546.6 - 211.0	548.2 - 207.2	231.1	239.4	38.4	19.4	0.1104	0.8793
2426	546.6 - 211.6	548.0 - 207.9	231.0	239.3	38.7	19.3	0.1099	0.8758
2503	546.6 - 211.7	548.3 - 207.9	230.8	239.1	38.8	19.4	0.1094	0.8720
2571	547.5 - 212.4	549.2 - 208.7	230.6	239.2	38.0	19.0	0.1116	0.8895
2666	546.6 - 211.7	548.2 - 208.0	230.7	239.2	39.2	19.5	0.1085	0.8642
2740	546.9 - 212.1	548.4 - 208.4	230.4	239.5	39.4	19.5	0.1080	0.8604

TABLE XI TEST CELL NO. 17 DATA (W AND SnTe)

Elapsed Time (Hours)	Temperatures (°C)		Ave. Seebeck Coefficients		Resistances		Power Output P ($\Delta T=340^{\circ}\text{C}$)	Relative to 0.1170
	P Leg	N leg	S _p	S _n	R _p	R _n		
3	550.8 - 202.6	548.0 - 198.3	244.8	241.2	40.9	24.9	0.1038	0.8872
20	549.5 - 207.5	546.3 - 202.8	248.1	242.8	40.2	23.5	0.1092	0.9333
26	546.9 - 213.3	544.5 - 208.9	249.8	243.5	39.8	23.7	0.1107	0.9462
67	547.9 - 213.1	545.3 - 208.7	249.7	243.6	38.3	23.4	0.1139	0.9735
140	547.3 - 213.2	545.0 - 208.8	247.8	243.0	36.8	23.1	0.1161	0.9923
163	546.7 - 213.3	544.2 - 208.9	247.2	242.9	36.5	23.0	0.1165	0.9957
188	546.4 - 213.1	544.0 - 208.7	246.6	242.7	36.2	23.0	0.1168	0.9983
213	547.4 - 213.0	544.8 - 208.5	246.0	242.5	36.2	23.0	0.1165	0.9957
236	546.7 - 213.0	544.3 - 208.9	245.7	242.4	36.0	22.9	0.1170	1.0000
269	547.9 - 213.0	545.3 - 208.8	243.2	240.8	36.0	23.5	0.1139	0.9735
290	547.0 - 212.5	544.5 - 208.2	244.7	241.9	36.2	23.7	0.1142	0.9761
318	547.0 - 211.4	544.5 - 207.1	244.3	241.6	36.0	23.6	0.1144	0.9778
344	547.4 - 211.8	545.0 - 207.4	243.9	241.5	36.1	23.7	0.1139	0.9735
365	547.3 - 212.0	544.8 - 207.5	243.7	241.4	35.8	23.5	0.1144	0.9778
438	547.5 - 212.8	545.3 - 208.7	243.2	242.6	35.9	24.0	0.1138	0.9726
463	547.5 - 212.6	545.2 - 208.1	243.1	241.1	36.0	23.8	0.1132	0.9675
487	547.7 - 212.3	545.5 - 208.0	242.9	241.0	36.0	23.8	0.1131	0.9667
510	547.5 - 212.0	545.2 - 207.6	242.5	240.9	36.0	23.8	0.1130	0.9658
535	548.0 - 212.3	545.8 - 208.0	242.4	240.9	36.0	23.8	0.1128	0.9641
608	548.5 - 212.7	546.2 - 208.3	242.0	240.6	36.1	23.8	0.1123	0.9598
632	549.4 - 212.3	547.1 - 207.9	241.6	240.5	36.1	23.9	0.1118	0.9558
656	548.9 - 212.5	546.7 - 208.1	241.5	240.3	36.1	23.9	0.1117	0.9551
680	548.4 - 212.1	546.1 - 207.8	241.3	240.2	36.0	23.8	0.1118	0.9559
703	548.5 - 212.1	546.2 - 207.6	240.9	240.1	36.0	23.9	0.1115	0.9528
775	548.0 - 211.9	545.5 - 207.6	240.1	239.9	36.1	23.8	0.1113	0.9510
824	548.0 - 212.3	545.7 - 207.8	240.3	239.6	36.1	23.9	0.1109	0.9483
871	550.0 - 213.2	547.7 - 208.6	239.9	239.5	35.4	23.4	0.1128	0.9643
943	549.4 - 212.9	547.0 - 208.4	239.5	239.5	36.3	24.0	0.1099	0.9393
1034	549.6 - 213.5	547.4 - 209.1	239.2	239.2	36.4	24.0	0.1092	0.9336
1110	549.8 - 213.0	547.6 - 208.4	238.6	238.9	36.5	24.6	0.1077	0.9203
1153	549.3 - 212.7	546.8 - 208.1	238.4	238.9	36.6	24.7	0.1073	0.9175
1203	549.0 - 212.3	546.8 - 207.8	237.9	238.5	36.6	24.7	0.1068	0.9131

TABLE XI TEST CELL NO. 17 DATA (W and SnTe)

Elapsed Time (Hours)	Temperatures (°C)		Ave. Seebeck Coefficients		Resistances		Power Output P($\Delta T=340^{\circ}\text{C}$)	Relative to 0.1170
	P Leg	N Leg	S _p	S _n	R _p	R _n		
1274	548.9 - 212.5	546.6 - 208.0	237.9	238.7	36.8	24.9	0.1063	0.9082
1328	549.9 - 212.5	547.6 - 207.9	237.4	238.2	36.9	25.0	0.1055	0.9018
1376	550.7 - 212.8	548.3 - 208.2	237.2	238.2	37.0	25.2	0.1049	0.8966
1467	549.8 - 212.7	547.5 - 207.9	236.6	237.9	37.0	25.3	0.1044	0.8919
1544	549.8 - 212.8	547.5 - 208.4	236.2	237.7	37.2	25.6	0.1034	0.8839
1611	550.7 - 213.1	548.3 - 208.6	236.0	237.6	37.1	25.6	0.1033	0.8827
1707	550.4 - 213.1	548.1 - 208.7	235.5	237.4	37.4	25.9	0.1020	0.8722
1781	550.7 - 213.4	548.5 - 209.0	235.2	237.4	37.5	26.2	0.1012	0.8651

TABLE XII TEST CELL NO. 18 DATA (W ELECTRODE-NO SnTe)

Elapsed Time (Hours)	Temperatures (°C)		Ave. Seebeck Coefficients		Resistances		Power Output P ($\Delta T=340^{\circ}\text{C}$)	Relative to 0.1135
	P leg	N leg	S _p	S _n	R _p	R _n		
3	547.5 - 199.9	546.0 - 196.9	247.7	249.3	43.4	25.3	0.1038	0.9145
20	546.8 - 202.9	545.5 - 199.2	249.5	248.9	43.9	23.4	0.1066	0.9392
26	551.6 - 210.5	550.6 - 207.5	251.6	250.3	44.5	23.6	0.1069	0.9419
67	551.7 - 211.5	550.7 - 208.4	252.0	249.6	44.3	22.2	0.1094	0.9639
140	550.8 - 211.8	549.8 - 208.7	251.3	248.7	43.8	21.1	0.1112	0.9797
163	550.6 - 212.2	549.2 - 209.1	251.0	248.5	43.7	20.8	0.1116	0.9833
188	550.1 - 211.8	548.8 - 208.7	250.6	247.9	43.6	20.6	0.1117	0.9841
213	550.5 - 211.7	549.5 - 208.7	250.5	247.6	43.7	20.5	0.1116	0.9833
236	551.9 - 212.3	550.4 - 209.6	250.4	247.6	43.1	20.0	0.1135	1.0000
311	548.7 - 210.3	547.9 - 207.0	249.1	247.0	44.0	20.3	0.1105	0.9736
332	549.8 - 210.6	548.9 - 207.5	249.3	247.1	44.3	20.2	0.1103	0.9718
386	549.8 - 210.0	548.9 - 206.5	248.8	246.6	44.3	20.0	0.1102	0.9709
407	549.7 - 210.2	548.8 - 206.9	249.1	246.4	44.5	20.0	0.1101	0.9700
480	550.5 - 211.0	549.7 - 208.3	248.7	246.8	44.7	20.0	0.1097	0.9665
505	551.2 - 211.0	550.1 - 207.9	248.7	246.2	44.8	19.8	0.1095	0.9648
530	551.1 - 211.0	550.2 - 207.9	248.4	246.2	44.6	19.8	0.1098	0.9674
552	551.0 - 210.7	550.2 - 207.4	248.2	246.0	44.4	19.7	0.1100	0.9692
577	551.1 - 210.7	550.3 - 207.3	248.2	246.0	44.8	19.8	0.1091	0.9612
650	551.4 - 211.0	550.6 - 207.9	248.0	245.8	45.0	19.7	0.1089	0.9595
674	551.4 - 210.2	550.9 - 207.0	247.7	245.7	44.9	19.7	0.1089	0.9599
698	551.9 - 210.9	551.3 - 207.6	247.6	245.7	44.8	19.6	0.1091	0.9616
722	551.9 - 210.7	551.3 - 207.5	247.5	245.7	45.0	19.7	0.1086	0.9566
745	551.6 - 210.6	551.1 - 207.2	247.3	245.6	45.1	19.7	0.1083	0.9544
817	551.0 - 209.9	549.9 - 207.2	246.9	245.3	45.1	19.4	0.1085	0.9556
866	550.5 - 210.6	549.9 - 207.3	246.7	245.2	45.1	19.4	0.1084	0.9551
913	550.5 - 210.8	549.8 - 207.5	246.4	245.1	45.1	19.3	0.1082	0.9537
985	550.8 - 210.8	550.0 - 207.6	246.1	245.1	45.3	19.4	0.1077	0.9490
1076	550.9 - 211.3	550.3 - 208.1	245.8	245.1	45.6	19.4	0.1072	0.9442
1152	550.9 - 210.3	550.5 - 207.3	245.4	244.9	45.7	19.4	0.1067	0.9398
1195	550.9 - 210.5	550.4 - 207.4	245.0	244.8	45.4	19.3	0.1069	0.9420
1245	550.7 - 209.8	550.1 - 206.5	244.7	244.6	45.3	19.3	0.1071	0.9434
1316	550.2 - 210.3	549.6 - 207.1	244.4	244.6	45.4	19.2	0.1069	0.9415
1370	551.2 - 209.9	550.6 - 206.7	244.1	244.7	45.5	19.3	0.1066	0.9390

TABLE XII TEST CELL NO. 18 DATA (W ELECTRODE-NO SnTe)

Elapsed Time (Hours)	Temperatures (°C)		Ave. Seebeck Coefficients		Resistances		Power Output $P(\Delta T=340^{\circ}\text{C})$	Relative to 0.1135
	<u>P leg</u>	<u>N leg</u>	<u>S_p</u>	<u>S_n</u>	<u>R_p</u>	<u>R_n</u>		
1418	551.0 - 210.5	550.4 - 207.1	244.0	244.6	45.7	19.3	0.1061	0.9351
1509	551.1 - 210.4	550.3 - 207.2	243.5	244.6	45.8	19.2	0.1058	0.9320
1586	551.1 - 210.9	550.5 - 207.7	243.0	244.6	45.8	19.2	0.1057	0.9315
1653	551.5 - 210.9	550.6 - 207.7	242.8	244.6	45.9	19.2	0.1054	0.9282
1749	551.2 - 211.2	550.4 - 208.0	242.5	244.6	45.9	19.2	0.1053	0.9279
1823	551.4 - 211.5	550.5 - 208.1	242.1	244.4	45.8	19.1	0.1053	0.9278

TABLE XIII

OCCURRENCE AND DURATION OF POWER OUTAGES FOR THE LIFE TEST CELLS

<u>DATE</u>	<u>CELL NO.</u>	<u>HOURS OF OPERATION AT TIME OF OUTAGE</u>	<u>LENGTH OF OUTAGE</u>
7-13-68	13	1971	42 hrs.
8-17-68	13	2761	46 hrs. 45 min.
7-13-68	14	1900	42 hrs.
8-17-68	14	2690	6 min.
7-13-68	15	1104	42 hrs.
8-17-68	15	1894	6 min.
7-13-68	16	1226	42 hrs.
8-17-68	16	2016	6 min.
7-13-68	17	267	42 hrs.
8-17-68	17	1057	6 min.
7-13-68	18	267	7 min.
8-17-68	18	1099	6 min.

FIGURE 1 Parts of the Extrusion Apparatus used to Fabricate 0.225" Diameter Rods of PbTe.

FIGURE 2

Seebeck Coefficients of an Extruded p-type Rod and Two Hot-Pressed Rods of Similar Composition as a Function of Temperature

- ⊙⊙⊙ E-29 Sample 6 - Extruded from P116A - $\text{PbNa}_{0.006}\text{Te}_{1.004}$
 — P-99 Hot-pressed from P-99 $\text{PbNa}_{0.0058}\text{Te}_{1.0034}$
 --- P-102 Hot-pressed from P-102 $\text{PbNa}_{0.0058}\text{Te}_{1.0039}$

FIGURE 3

Electrical Resistivities of an Extruded p-type Rod and Two
Hot-Pressed Rods of Similar Composition as a Function of Temperature

- ⊙ ⊙ ⊙ E-29 Sample 6 - Extruded from P116A - $\text{PbNa}_{0.006}\text{Te}_{1.004}$
 — P-99 Hot-pressed from P-99 $\text{PbNa}_{0.0058}\text{Te}_{1.0034}$
 - - - P-102 Hot-pressed from P-102 $\text{PbNa}_{0.0058}\text{Te}_{1.0039}$

FIGURE 4 Tin telluride brazed thermoelements prepared for thermocouple performance test with current leads and temperature measuring thermocouples.

FIGURE 5 R&DC thermocouple setup in jig for performance testing.

FIGURE 6 Thermocouple in performance test jig mounted on base of test cell with leads attached.

FIGURE 7 Thermocouple performance test cell mounted on ultrahigh vacuum system for bakeout procedure.

FIGURE 8A View of the Life Test Cells showing the cells with insulation on the top flange, ice junction dewars, connecting cables, and cell power panels, with controllers and over temperature protection circuits.

FIGURE 8B Second View of the Life Test Cells, showing the cells with insulation on the top flange, ice junction dewars, connecting cables, and cell power panels, with controllers and over temperature protection circuits.

FIGURE 9

Normalized Power Output to Matched Load for
 Test Cell No. 13 (○), No. 14 (□), and No. 15 (◇). $T_h = 550^\circ\text{C}$, $\Delta T = 340^\circ\text{C}$

FIGURE 10

Normalized Power Output to Matched Load for Test

Test Cell No. 16 (Δ), No. 17 (∇), and No. 18 ($\square$). $T_h = 550^\circ\text{C}$, $\Delta T = 340^\circ\text{C}$

FIGURE 11

Power Output Degradation for Test Cell No. 13 (○)
and Test Cell No. 14 (□). $T_h = 550^\circ\text{C}$, $\Delta T = 340^\circ\text{C}$

FIGURE 12

Power Output Degradation for Test Cell No. 15 (◈)
and Test Cell No. 16 (Δ). $T_h = 550^\circ\text{C}$, $\Delta T = 340^\circ\text{C}$

FIGURE 13

Power Output Degradation for Test Cell No. 17 (∇)
and Test Cell No. 18 ($\triangle$). $T_h = 550^\circ\text{C}$, $\Delta T = 340^\circ\text{C}$

FIGURE 14

Average Seebeck Coefficients for Test Cell No. 13 (○)
and Test Cell No. 14 (□). $T_h = 550^\circ\text{C}$, $\Delta T = 340^\circ\text{C}$

FIGURE 15

Average Seebeck Coefficients for Test Cell No. 15 (◊)
and Test Cell No. 16 (Δ). $T_h = 550^\circ\text{C}$, $\Delta T = 340^\circ\text{C}$

FIGURE 16

Average Seebeck Coefficients for Test Cell No. 17 (∇)
and Test Cell No. 18 ($\triangleright$). $T_h = 550^\circ\text{C}$, $\Delta T = 340^\circ\text{C}$

FIGURE 17

Element Resistances for Test Cell No. 13 (○)
and Test Cell No. 14 (□). $T_h = 550^\circ\text{C}$, $\Delta T = 340^\circ\text{C}$

FIGURE 18

Element Resistances for Test Cell No. 15 (◇)
and Test Cell No. 16 (△). $T_h = 550^\circ\text{C}$, $\Delta T = 340^\circ\text{C}$

FIGURE 19

Element Resistances for Test Cell No. 17 (▴) and Test Cell No. 18 (▢). $T_h = 550^\circ\text{C}$, $\Delta T = 340^\circ\text{C}$