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THE THERMAL CONDUCTIVITY
OF A MOLTEN Pu-Fe EUTECTIC (9.5 at/o Fe)

by

John E. Deverall

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ABSTRACT

The thermal conductivity of the Pu-Fe eutectic mixture (9.5 a/o Fe) was measured in both the solid and liquid states by the use of a guarded cylinder apparatus which operated in a high vacuum. The apparatus was designed as an absolute measuring device and was checked with a known material.

The thermal conductivity values obtained varied from 0.034 at 220°C to 0.042 at 400°C in the solid state, and from 0.043 at 420°C to 0.046 at 520°C in the molten state, with the units of cal/cm sec °C. The curves appear to be linear with positive temperature coefficients for both phases.

Results are estimated to be accurate to less than $\pm 5\%$.

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1. INTRODUCTION

In the design calculations for nuclear reactors, one of the important properties of the fuel which must be known is the thermal conductivity. As the Pu-Fe alloy appears to have suitable characteristics for a molten fuel, a special apparatus was designed and built to determine the thermal conductivity of the Pu-Fe eutectic (9.5 a/o Fe).

After consideration of the problems involved and a study of the various methods for determining thermal conductivity, it was decided to design an apparatus of the guarded cylinder type.

One of the few materials which is able to contain molten Pu-Fe alloy is tantalum. Because tantalum oxidizes readily at high temperature, a special system had to be designed which would operate in either an inert atmosphere or a high vacuum. Because it is extremely difficult to obtain a good inert atmosphere in a heavily insulated system and convection effects would prevent the use of radiation shielding, it was decided to build a guarded cylinder system which would operate in a high vacuum. The major problem then was to develop a radiation shield system which would be equivalent to an insulation system so that adequate guarding could be obtained.

The apparatus developed was designed as an absolute measuring device for steady-state, one-dimensional flow. The total watts input to the specimen heater was assumed to be heat flow directed down the specimen bar, and temperatures measured along the tantalum container were assumed to be the alloy temperatures. The conductivity values were calculated by a formula derived from the linear differential equation for one-dimensional heat flow which includes a heat generation

factor, as shown in Appendix A. The heat generation is due to alpha heating in the plutonium and is independent of temperature.

2. RESULTS

Sufficient runs were made to establish the thermal conductivity of the Pu-Fe eutectic (9.5 a/o Fe) from 220°C to 400°C in the solid state and from 420°C to 520°C in the molten state. The curves plotted for these ranges appear to be linear with positive temperature coefficients in both regions, as shown in Fig. 1. The curves are estimated to be accurate to within $\pm 5\%$. It was not possible to determine the thermal conductivity at the melting point; however, an extrapolation of the solid and liquid state curves to the melting point shows very little discontinuity through the phase change.

A check of the accuracy of the heat flux measurement and the guarding efficiency was made by determining the thermal conductivity of the tantalum section at the bottom of the bar and comparing the values with those determined in the previous tantalum thermal conductivity test. Points were obtained from 150°C to 300°C and when plotted showed a maximum deviation from the original curve of 5% (see Fig. 1).

There appears to be a slight difference in the temperature coefficients of conductivity between the solid and liquid states. The coefficient for the solid state is 13×10^{-4} per °C, while for the liquid state it is 7.0×10^{-4} per °C.

3. APPARATUS

3.1 General Arrangement

The apparatus was made up of three major sections: the vacuum system, temperature measuring circuits, and the power supply and control panel. Each section was a complete unit mounted on wheels for portability, as shown in Fig. 2. Wires which connected the sections were

Fig. 1. Thermal Conductivity of Pu-Fe Eutectic.

Fig. 2. General Arrangement of Apparatus.

attached to terminal blocks and all water connections were rubber tubes. This simplified separation of the apparatus into individual units. The portability feature was adopted to facilitate the transfer of the apparatus to a hot laboratory in the event this became necessary.

3.2 Bell Jar

The bell jar, as shown in the upper left corner of Fig. 2, was constructed by silver soldering a flange on each end of a 10 inch diameter brass tube 18 inches long. An O-ring seal was used at each end to provide vacuum tight connections to the base and lid. Kovar seals were soft soldered into the lid (see Fig. 3) to provide for feed-through of the power and thermocouple leads. Cooling at the upper section of the bell jar was obtained by a copper water coil which was wound around the bell jar and soldered to the outer surface. The base of the bell jar (see Fig. 3) was a 1 inch brass plate containing an O-ring groove and bolt holes for attaching the jar. Two separately controlled water coolers imbedded in grooves in the base provided cooling for both the base plate and the guard heater.

3.3 Specimen Bar Cooler

The heat sink for the specimen bar was a flanged brass pedestal containing a socket at the top and a water cooled chamber at the bottom. The flange was bolted to the base plate of the bell jar on an O-ring seat, as shown in Fig. 3, with the cooler projecting through a clearance hole in the plate. This arrangement isolated the heat sink from the coolers of the base plate and provided better control for the matching of the guard temperatures. The brass base of the specimen bar was machined and tinned to be a tight fit in the socket for good thermal contact. The bar was inserted by driving it into the socket and removed by the use of an easy-out.

3.4 Guard Heater

The guard heater (see Fig. 4) was machined from a 11-3/8 inch section of 4 inch stainless steel pipe with a 1/2 inch wall. The outside diameter was machined to 4-7/8 inches over the entire length except for

Fig. 3. Vacuum System Disassembled.

Fig. 4. Main Guard Heater.

twelve ribs $1/16$ inch wide. These ribs, in pairs, were used to locate and support the six heater windings. A flanged brass ring section soldered to the base of the heater provided for attachment to the base plate of the bell jar. A silver gasket between the flange and the base plate increased the thermal contact to the heat sink. The heater windings were 26 gauge nichrome wire, wound in a $1/16$ inch diameter helix and threaded through $1/4$ inch diameter porcelain beads. The helix made it possible to have more resistance in a given space, as well as to compensate for differences in thermal expansion which eliminated wire breakage over a large temperature range. Additional isolation of the windings from the tube was obtained by the application of a coat of fused alumina between each pair of supporting ribs. Each heater consisted of four turns of the helix wrapped around the stainless tube and had a resistance of about 25 ohms. After the windings were in place the porcelain beads were covered over by a layer of Alundum cement.

A porcelain insulator, $3/8$ inch in diameter and $5/8$ inch long on each side of a winding, provided a means for anchoring the ends of the winding and attaching the power leads. The insulators were recessed half way into the wall of the tube and bolted from the inside. Thermocouple holes were drilled into the wall at spacings which corresponded to the spacings of the thermocouples of the specimen bar. The holes were $1/4$ inch deep with a diameter to accept $1/8$ inch diameter, two-hole porcelain insulators. These insulators separated the leads of the thermocouple and were anchored to the wall by Sauereisen cement. The junction was formed on each thermocouple by twisting the wires together and welding with an arc process under oil. All the leads were covered with Refrasil sleeving for insulation and brought up the side of the heater to octal plugs for attachment to the Kovar fittings in the lid. The power leads from each heater were connected to the Kovar fittings in a similar manner.

3.5 Top Heater

The top heater as shown in left foreground of Fig. 3 was composed

of two discs of Lavite 6 inches in diameter and $1/4$ inch thick. The lower disc had double thread spiral grooves, $1/16 \times 1/16$, machined in it to contain the winding. After machining, the Lavite was fired at 900°C . A 26 gauge nichrome wire helix was folded double, laid in the grooves, and the ends were brought out to terminal bolts. This formed a noninductive type of winding with a resistance of 26 ohms. After the grooves were filled with Alundum cement for better contact between the wire and the Lavite, the upper disc was bolted to the lower to form a complete unit. Diametrically opposite holes near the periphery provided for bolting the heater to brackets on the top of the main guard heater (see Fig. 4). The heater temperature was measured by a thermocouple mounted in a porcelain insulator and inserted in a hole at the center of the heater.

3.6 Specimen Heater

The specimen heater, as shown in the lower left corner of Fig. 3, was a $1/2$ inch diameter by 2 inch long porcelain cylinder containing 48 longitudinal holes through the length of it, equally spaced over the cross sectional area. A 30 gauge nichrome wire was threaded back and forth through these holes, starting with the hole at the center and working toward the outside to form a resistance of approximately 50 ohms. Each of the two ends of the wire was wound around a copper wire and silver soldered to form leads to the octal plug.

3.7 Specimen Bar

The specimen bar (see Fig. 5) was made in two parts, the main body and cap. Both parts were machined from solid tantalum bars 0.800 inch in diameter. Before machining, the bar was sonically tested and x-rayed to make sure it was free from voids. A hole was bored and reamed to a uniform diameter of 0.637 inch by $4-3/8$ inches deep in the main body to provide a chamber for the alloy. The outside diameter of the chamber was turned to 0.700 inch except for three ribs $1/16$ inch wide spaced 1 inch apart. These ribs were then milled off except for two diametrically opposite sections $1/8$ inch long. This provided

Fig. 5. Specimen Bar for Pu-Fe Thermal Conductivity Test.

sufficient metal to drill thermocouple holes without danger of penetrating the wall. Below the specimen chamber the tantalum was turned to 7/16 inch diameter for a length of approximately 3-1/2 inches. This provided a tantalum test section with a conductivity about the same as that of the Pu-Fe section which produced a relatively linear temperature gradient over the length of the bar. A 1/4-20 thread was tapped up into the bar for a depth of 3/8 inch for the attachment of a brass base. The brass male thread inside the tantalum maintained a good thermal contact at high temperature as the expansion coefficient of brass is about four times that of tantalum.

The cap was a tantalum section 2-47/64 inches long with a 33/64 inch diameter hole bored in it to contain the specimen heater. A small flange was turned on the outside diameter to mate with a similar flange on the body to provide for welding the parts together after loading. A 1/4 inch diameter tip, about 7/16 inch long, protruded from the bottom of the cap into the Pu-Fe specimen to assure more even distribution of heat to the specimen.

Eight thermocouple holes, 0.030 inch in diameter, were drilled into the bar to a depth of 0.050 inch. These holes were accurately spaced along the bar by indexing them in a milling machine.

The Pu-Fe specimen (see Fig. 6) was loaded into the bar chamber as a solid cylinder machined to size and containing a hole at the top to accept the tip protruding from the bottom of the cap. The machined specimen weighed 258 grams. The chamber was flushed with helium to obtain an inert atmosphere before the specimen was inserted. The specimen was slipped in through a wrapping of aluminum foil to prevent contamination of the outside edges of the bar. The cap was then inserted and welded. The welding was done by Heliarc under a bell jar containing an argon atmosphere to prevent oxidation of the tantalum. After welding, the bar was completely monitored for any alpha contamination. The assembled bar is shown in Fig. 7.

After the bar was loaded, thermocouples were put in each of the

Fig. 6. Pu-Fe Specimen.

Fig. 7. Pu-Fe Specimen Bar Assembled.

thermocouple holes and peened in position by a small prick punch. To reduce lead loss, each thermocouple lead was wrapped around the bar once before it was brought up along the bar. All leads were covered by Refrasil sleeving for insulation and brought up to octal plugs for attachment to the Kovar fittings in the lid.

3.8 Radiation Shielding

The most difficult problem in building this apparatus was the development of an adequate shield system. The shielding had to be equivalent to an insulation system for proper guarding, without restricting flow to the vacuum pumps from all parts of the system. Additional precautions had to be taken to eliminate conduction paths along the shielding which could cause unpredictable heat leaks. All radiation shields were fabricated from 0.002 inch polished stainless steel shim stock.

The main body of the specimen bar was shielded by a series of discs spaced $1/2$ inch apart, as shown in Fig. 8. The discs were supported by three $1/16$ inch diameter tubes spaced 120 degrees apart and parallel to the specimen bar. The tubes were crimped above and below each disc to hold it in position. To prevent radial radiations, a strip of shim stock was inserted between each pair of discs to form a vertical cylinder concentric with the bar and about 2 inches in diameter. The top of the bar was shielded by a pair of concentric cylinders, one $1-1/2$ inch in diameter and the other 2 inches in diameter, which rested on top of the lower radiation structure. A disc was then placed on top of these cylinders to reduce end effects. This completely enclosed the specimen bar in a shield system, but the loose fits of all the parts provided the flow paths needed to obtain a good vacuum, as well as producing a minimum of conduction paths. Additional shielding was obtained by slipping a sheet of shim stock in the form of a cylinder inside the full length of the main guard heater and placing discs across the top and bottom of the heater.

Although heat losses from the guard heater to the bell jar did not

Fig. 8. Specimen Bar Radiation Shielding.

affect the accuracy of the data, it was necessary to cover the outside of the guard heater with Fiber-Frax insulation to reduce the current in the heater windings (see Fig. 9). Without this insulation, the currents necessary to maintain a specified heater temperature at high temperature levels caused numerous heater failures. To obtain good vacuums, this insulation was held in position by a heavy mesh screen so that the maximum area would be exposed to paths to the vacuum pumps. The screen was in the shape of a cylinder with a diameter 2 inches larger than that of the guard heater and measuring its full length. This cylinder rested on a screen disc bolted to the bottom of the guard heater.

Surrounding the screen cylinder was another shield system similar to that around the specimen bar but on a larger scale, as shown in Fig. 9. This assembly consisted of seven discs with an outside diameter slightly smaller than that of the bell jar and spaced about 2 inches apart. The discs were held in position by three threaded rods with a nut above and below each disc. The rods rested on the base plate of the bell jar. To reduce radial radiation to the bell jar, vertical shields were inserted between each pair of discs. Loose fits of all the parts, plus scallops cut in outer edges of each disc, provided paths to the vacuum pumps.

The upper radiation shield system was suspended from a copper ring soldered to the lid, as shown in Fig. 3. The main reflector was a heavy gauge stainless steel disc which was electropolished to a mirror finish. This disc was slightly smaller than the diameter of the bell jar and had two holes cut in the outer edge to allow for passage of the thermocouple and power leads to the octal plugs. Below this disc was suspended a smaller diameter disc plus a shield in the form of a bonnet. The bonnet reduced both vertical and radial radiations from the top heater.

To reduce radiation scattering, the inside of the bell jar was painted with carbon black. The radiation absorbed was conducted to the water coolers of the base plate and bell jar.

Fig. 9. Guard Heater Shield System.

The radiation shield system proved very satisfactory in all respects. Accurate guarding was obtained over the whole length of the specimen bar, the desired temperatures were obtained with relatively low watts input to the heaters, and vacuums better than 10^{-6} mm Hg were readily attainable.

3.9 Vacuum System

High vacuums were attained by the use of two pumps: a mechanical fore pump and a water-cooled, oil-diffusion pump. A liquid nitrogen cold trap was placed between the diffusion pump and the bell jar. A 2-1/2 inch diameter tube 10 inches long connected the cold trap to the system and contained two vacuum gauges. One gauge was the thermocouple type which operated the safety control and the other a Phillips gauge which was used for the vacuum measurement.

The cold trap was constructed from brass tubing and had a capacity of about 4 liters of liquid nitrogen. The inner can containing the nitrogen was suspended from the lid by two stainless steel fill tubes which were machined to a thin wall to reduce losses by conduction. Radiation losses were reduced by covering the inner surfaces of the outer container and the outer surfaces of the nitrogen can by aluminum foil. The complete trap was covered by a layer of insulation and wrapped with aluminum foil. The lid of the trap seated on an O-ring and was sealed by bolts through the flange of the outer can to provide easy access for cleaning.

A special system was constructed to fill the cold trap automatically because normal runs took about 2 weeks of continuous operation. This system was composed of an electronic circuit which operated a three-way solenoid valve on a nitrogen gas tank. When the valve opened, gas flowed into a liquid nitrogen dewar and forced the liquid into the cold trap. Two resistors were used as sensing elements in the trap. These resistors were spaced 7 inches apart on a vertical fiber rod, one to call for nitrogen and the other to close the solenoid when the trap was full. In closed position the three-way valve allowed the venting of the dewar so

liquid flow was stopped almost immediately. With this system, the vacuum equipment could be run for 4 or 5 days without attention.

A diffusion pump protect-circuit was installed in the vacuum pump system to shut the diffusion pump off in the event of water failure, over-heating, or loss of vacuum. A pressure operated mercury switch in the cooling water line would interrupt the diffusion pump heater circuit in the event of a water failure. Interruptions of the heater circuit were also made by relays controlled by the thermocouple gauge for loss of vacuum and a thermostat for overheating. The protect-circuit contained an auxiliary power outlet which was also interrupted when the diffusion pump was shut off. This power outlet was used to operate the relay which controlled all the heaters, the temperature controller, the Phillips gauge, and the cold trap filling circuit. In the event of any failure which would shut off the diffusion pump, all equipment was automatically shut off except the fore pump. The protect-circuit had a manual reset so that continuity was not re-established until the operator had located the trouble and established the conditions necessary for circuit operation.

4. INSTRUMENTATION

All temperatures were measured by 28 gauge chromel-alumel thermocouples. Each junction was made by twisting the two wires together and welding them by an arc process under oil. All the thermocouples were attached to a "Rubicon B" potentiometer through three 10-point selector switches. The cold junction was inserted in a thermos bottle filled with ice made from distilled water. The proper temperature was indicated by the use of an electronic null indicator in conjunction with the bridge circuit of the potentiometer. Thermocouple No. 1, at the top of the guard heater, was connected to a double pole, double throw switch so that it could be connected to either the potentiometer or the temperature controller. Thermocouple No. 13 at the top of the specimen bar was

connected to a similar switch so that it could be connected to the potentiometer or the circular chart temperature recorder. Except when data were being taken, these two thermocouples were connected to the controller and the recorder (see Fig. 10).

The temperature controller was used only as an upper limit temperature switch. It was set at a temperature which was considered the safe upper limit for the particular conditions of the experiment. If this temperature was reached while the apparatus was unattended, the controller would shut the main power off and on to maintain this temperature $\pm 5^{\circ}\text{C}$. The controller had a range from 0°C to 1000°C .

The temperature recorder was a Leeds and Northrup, 24 hour circular chart type with a range from 0°C to 1000°C . It was put in the circuit primarily to indicate whether steady-state conditions were being maintained while the apparatus was unattended. It was also useful as it would show if failures occurred in the heaters and the time of the failure.

Accurate measurements of watts input to the specimen heater were made by the use of precision resistors in the heater circuit. One resistor, with a resistance of 0.10 ohm, was placed in series with the heater and was used for the current measurement. A 500 ohm resistor, in series with a 500 k ohm resistor and in parallel with the heater, was used for the voltage measurement. Voltage drops across the resistors were measured by the potentiometer. Two points on one of the thermocouple selector switches were used for the switching system. When the voltages were being read, the cold junction was cut out of the potentiometer circuit by a double pole, double throw switch which had one pair of terminals shunted. The resistance values were chosen so that the digits indicated by the potentiometer were the same as the current and voltage digits, and it was necessary only to locate the decimal points. As D. C. was supplied to the specimen heater, the watts input was taken as the product of the measured current and voltage. Losses in the lead wires were negligible. Current could be measured to 0.00005 ampere and

Fig. 10. Thermocouple Wiring Diagram

voltage to 0.005 volt.

The power to the specimen heater was supplied by a specially constructed D. C. power supply. It had a capacity of 1200 watts and the output was controlled by a Variac at the input.

Each guard heater winding was supplied with A. C. controlled by a Variac. A Masonite panel contained all the controls, meters, switches and fuses. The power to all the controls was fed through a large Variac so that the supply voltage could be lowered to all heater Variacs and the D. C. power supply. This provided more sensitivity in control at low watts input (see Fig. 11).

5. TEST PROCEDURE

To obtain sufficient data for establishing a thermal conductivity curve over a range from 100°C to 500°C for a given specimen, a run of approximately 2 weeks was required. At the beginning of each run the vacuum pumps were operated until a vacuum of 10^{-7} mm of Hg was obtained. This required about 2 days. The water coolers were then turned on and power applied to the heaters. The rate of heating was limited by the amount of outgassing of the system and the inputs to the heaters were adjusted so that the system pressure never exceeded 10^{-5} mm Hg. During the heating up process, adjustments were made continually in an effort to match all the guard temperatures with the corresponding specimen bar temperatures. When predetermined temperature levels were reached, steady-state conditions were established and conductivity values computed from the data taken. It required from 24 to 48 hours to establish steady state at each temperature level. Curves were plotted of temperature versus length for both the guard heater and the specimen bar each time a set of readings were taken. When the curves were coincident, it was assumed that adequate guarding had been established. For each temperature level, several conductivity values could be computed by using different intervals down the bar. The number of values obtainable

Fig. 11. Heater Wiring Diagram.

depended upon the type of specimen bar used.

After the highest temperature level had been established, the power was reduced and several temperature levels were established while cooling. This gave additional conductivity values, as well as a check on the reproducibility of the apparatus.

Each set of readings was recorded on a special data sheet, shown in Fig. 12. Temperatures were recorded in their respective positions on the sketch of the apparatus. This facilitated the adjustment of the controls to obtain adequate guarding as it produced a visual check on all unmatched temperature conditions.

6. CALIBRATION TESTS

Although this apparatus was designed as an absolute measuring device, it was considered advisable to calibrate it by first running a test with a standard material. The material chosen was 304 stainless steel with a chemical analysis as shown in Appendix C. This choice was made because its conductivity values are well established, the conductivity of alloyed metals is not appreciably affected by small amounts of impurities, and it is stable at high temperatures. Before the bars were machined, the stock was radiographed and sonically tested for voids or flaws.

In order to check the instrumentation, controls, and watts measuring system, the first test of the standard was made with the vacuum assembly replaced by another guarded cylinder arrangement in an insulated system as shown in Fig. 13. The specimen bar in this case was 1 inch in diameter and 14-1/4 inches long with the copper tubing for the heat sink wound in a recess at the base of the bar (see Fig. 14). A similar arrangement was made for cooling the guard heater. A curve plotted from the data agreed with the ASM values to within less than 2% (see Fig. 15).

When this calibration test proved that the instrumentation was accurate and the control system adequate, a second test was made using the guarded cylinder arrangement in the vacuum system. The same material

(Text continues on page 35)

DATA SHEET

DATA: _____ SHEET NO.: _____

TIME: _____

HEATERS:

	P.S.	AMPS.
1		
2		
3		
4		
5		
6		
TOP		

SPECIMEN HTR: VOLTS _____ AMPS. _____

SPECIMEN HTR: VOLTS _____

SHUNTS: AMPS. _____

MAIN POWERSTAT: _____

VACUUM: _____ MM. HG

Fig. 12. Sample Data Sheet.

Fig. 13. Insulated Guarded Cylinder Apparatus.

Fig. 15. Thermal Conductivity of 304 Stainless Steel.

was tested; however, in this case the specimen bar was 10 inches long and had a uniform diameter of $3/4$ inch except for the heater section at the top, which was 1 inch in diameter, as shown in Fig. 16. From the data obtained in this test, a curve was plotted between 100°C and 550°C which agreed with the ASM values within 2% (see Fig. 17).

A thermal conductivity test was also run with a 440-C stainless steel bar of the same dimensions and a curve plotted from 100°C to 450°C . A survey of the literature revealed only one value at 100°C and at this temperature the curve agreed within 2%. The general curve was very similar in shape and absolute values to curves established for similar 400 series stainless steels (see Fig. 18).

On the basis of the above tests, it was assumed that adequate guarding was being obtained, the watts measurements were accurate, and the conductivity values which could be obtained on a specimen would be accurate to less than 5%.

7. TANTALUM THERMAL CONDUCTIVITY TEST

In the calculations for the thermal conductivity of the Pu-Fe alloy, it was necessary to know the thermal conductivity of tantalum to correct for the heat flow down the container wall. Because a survey of the literature failed to reveal any values except at 18°C and 100°C , it was necessary to run a test to establish a curve from 100°C to 500°C . The specimen bar was machined from a tantalum rod supplied by Fansteel Corporation with a purity of 99.9% (see Fig. 19). The bar was not machined to $3/4$ inch diameter as were the stainless steel specimens, because the same bar was to be later machined to the dimensions required for the Pu-Fe specimen. Before machining, the stock was radiographed and sonically tested for voids or flaws. After machining, hardness measurements were made over the length of the bar before attaching the thermocouples.

The same test procedure was used as before, and from the data
(Text continues on page 40)

Fig. 16. Stainless Steel Specimen Bar for Vacuum Apparatus.

Fig. 17. Thermal Conductivity of 304 Stainless Steel.

Fig. 18. Thermal Conductivity of 440-C Stainless Steel.

Fig. 19. Tantalum Specimen Bar.

obtained, a curve was plotted from 100°C to 500°C. The conductivity over this range proved to be essentially constant. The point at 100°C agreed with the handbook value within 3% (see Fig. 20).

After completion of the run, a hardness test revealed that there had been no increase in hardness and in some sections the bar was even slightly annealed. This was an indication that an adequate vacuum had been maintained.

8. Pu-Fe ALLOY CONDUCTIVITY TEST

The Pu-Fe alloy tested was the eutectic mixture containing 90.5 a/o Pu and 9.5 a/o Fe with a melting point of 410°C.

After the bar had been loaded with the alloy and the cap welded on, hardness measurements were made over the length of the bar. The thermocouples were then attached and the bar placed in the vacuum apparatus as shown in Fig. 21. The usual 2 day period was used to establish a good vacuum before any heat was turned on. The same test procedure was used as in previous tests except that no data could be taken until the alloy was completely melted to establish contact of the alloy with the tantalum wall.

After sufficient data were taken in the molten state, lower temperature levels were established to procure data for the solid state. No attempt was made to take data below 200°C as it was calculated that at temperatures below this, the alloy could contract away from the wall and wall temperatures would not indicate the specimen temperatures. As no discontinuities occurred in the solid state curve in the range considered, it was assumed that no separation had occurred.

Three separate runs were made with the specimen. The same procedure was used in each case except that after the first run, data could be taken for the solid state during the heating period as well as during cooling period.

Data from the three runs were plotted and conductivity curves

Fig. 20. Thermal Conductivity of Tantalum.

Fig. 21. Vacuum-guarded Cylinder Apparatus.

established for the solid and liquid states. A curve was also plotted for tantalum from data taken at the lower section of the bar.

Upon completion of the tests, the specimen bar was radiographed to ascertain if the specimen was homogeneous and free from any voids. The radiograph showed only one void 0.050 inch in diameter at the top section of the specimen which was considered insignificant and would cause no appreciable effect on the heat flow pattern. A metallographic examination of sections of the specimen bar revealed that there was no significant separation of the plutonium and iron and that the tantalum wall was not attacked.

A recheck of the hardness of the specimen bar showed no change in hardness. This indicated that an adequate vacuum had been maintained during the tests.

Whenever tests were made with radioactive specimens, a filter was attached to the outlet of the forepump so that in case the specimen container ruptured, there would be no possibility of alpha contamination getting out of the system into the room. The filter was removed from its case at least once every day and monitored for alpha count.

Before personnel entered the room containing the apparatus, the vacuum gauge reading was always observed through the glass door to assure that no vacuum failure had occurred. In the event that the vacuum had failed, a mask was put on before entering and worn until the cause of the failure could be ascertained.

Although no containers ever ruptured, it was believed that if the molten alloy had ever leaked out it would have been obvious by a radical change in the temperature gradient down the bar. In this event, the complete vacuum system would have been removed to a dry box for dismantling.

Whenever a specimen bar was removed from the apparatus after a test, masks and rubber gloves were worn for the operation in case the container had cracked upon cooling. As each part of the apparatus was removed, it was checked carefully with an alpha counter.

9. CONCLUSIONS

From the results of the calibration tests it was estimated that the apparatus was capable of determining thermal conductivities of metals in the range from 100°C to 550°C with an error of less than 5%.

The thermocouples were not calibrated as it did not seem feasible to calibrate them before the peening operations and once the thermocouples were mounted on the bar, the alpha heating made any calibration uncertain. During the tests, however, it was found that results were duplicated very closely if a new complete set of thermocouples was mounted on a bar. It was also noted that relatively smooth curves were obtained when temperature versus length was plotted for the various bars, indicating that no erratic temperature readings were being obtained. Further, the conductivities of stainless steel obtained were less than 2% in error. These facts indicated that the thermocouple wire used was uniform and the mounting procedure consistent. On this basis, temperature differences along the specimen bars were estimated to be accurate to less than 2%.

Another possible source of error was radiation leakage through the shielding around the bar, resulting in heat exchange between the bar and the guard system. Here again however, the calibration tests indicated that any such leakage must have been small. The same shield system was used for all tests and because each specimen was tested over approximately the same temperature range, it was assumed that no appreciable error was introduced by leakage. If in future testing the temperature range is extended to higher levels, it would be advisable to run a calibration test at the same levels since radiation is a function of the fourth power of temperature and leakage could cause appreciable error.

It was assumed that any heat transfer by convection was negligible as vacuums of 10^{-6} mm Hg or better were maintained when data were taken.

Deviations from linear heat flow caused by alpha heating and variation in k as a function of temperature were investigated by

B. M. Carmichael and M. E. Battat. The result of this investigation indicated that errors introduced by radial heat flow were negligible, as shown in Appendix B.

Calculations showed that changes in geometry of the stainless steel specimen due to thermal expansion affected the result less than 1%. Because tantalum has a coefficient of expansion which is $1/4$ that of stainless steel, the error introduced in the Pu-Fe alloy tests were considered negligible.

The apparatus was designed so that the specimen bars were heated at the top with a temperature gradient decreasing toward the bottom. This was done to eliminate natural convection in the molten alloy, and since the radial temperature gradient due to alpha heating was shown to be very small, it was assumed that convective effects in the molten alloy were negligible.

It is believed that this type of apparatus provides a suitable method for accurately determining the thermal conductivities of metals which oxidize readily in normal atmospheres without the complications of plating or cladding. The temperature ranges used were determined by the current needs; however, there appears to be no reason why data at higher temperatures could not be obtained except that more time would be needed to outgas the system while maintaining the required vacuum. It could also be used as a standard guarded cylinder apparatus for other metals by packing the bell jar and guard heater full of insulation and not using the vacuum system.

BIBLIOGRAPHY

Metals Handbook, 1954 Supplement, The American Society for Metals, Cleveland.

Smithells, C. J., Metals Reference Book, Interscience Publishers, Inc., New York, 1949.

Handbook of Chemistry and Physics, 37th edition, Chemical Rubber Publishing Company, Cleveland, 1955-56.

Jakob, Max and Hawkins, G. A., Elements of Heat Transfer and Insulation, 2nd edition, John Wiley and Sons, Inc., New York, 1954.

Jakob, Max, Heat Transfer, Vol. 1, John Wiley and Sons, Inc., New York, 1949.

McAdams, W. H., Heat Transmission, McGraw-Hill Book Company, Inc., New York, 1954, p. 21.

Appendix A

DERIVATION OF THE EQUATION FOR LINEAR HEAT FLOW WITH HEAT GENERATION

The general equation for temperature distribution in a body with heat generation is

$$\frac{\partial t}{\partial \theta} = \alpha \nabla^2 t + \frac{w}{\rho c} \quad (\text{A.1})$$

where

t = temperature

θ = time

α = thermal diffusivity, $\frac{k}{\rho c}$

w = heat generation per unit volume

k = thermal conductivity

ρ = density

c = specific heat

For one-dimensional, steady-state heat flow

$$\frac{\partial t}{\partial \theta} = 0$$

$$\nabla^2 t = \frac{d^2 t}{dx^2}$$

Substituting in Eq. A.1 gives

$$0 = \alpha \frac{d^2 t}{dx^2} + \frac{w}{\rho c}$$

or

$$\alpha \frac{d^2 t}{dx^2} = - \frac{w}{\rho c} \quad (\text{A.2})$$

however

$$\alpha = \frac{k}{\rho c}$$

therefore

$$\frac{k}{\rho c} \frac{d^2 t}{dx^2} = - \frac{w}{\rho c}$$

or

$$\frac{d^2 t}{dx^2} = - \frac{w}{k} \quad (\text{A.3})$$

Double integration gives

$$\frac{dt}{dx} = - \frac{wx}{k} + c_1 \quad (\text{A.4})$$

$$t = - \frac{wx^2}{2k} + c_1 x + c_2 \quad (\text{A.5})$$

From the boundary conditions

$$\begin{aligned} x = 0, & \quad t = t_o \\ x = L, & \quad t = t_L \end{aligned}$$

then

$$c_1 = \frac{t_L - t_o}{L} + \frac{wL}{2k} \quad (\text{A.6})$$

$$c_2 = t_o \quad (\text{A.7})$$

Inserting these constants in Eq. A.5 and substituting $\Delta t = t_o - t_L$ gives

$$t_x = -\frac{wx^2}{2k} + \left(\frac{-\Delta t}{L}\right)x + \frac{wL}{2k}x + t_o$$

or

$$t_x = -\frac{w}{2k} \left[x^2 - Lx \right] - \left(\frac{\Delta t}{L}\right)x + t_o \quad (\text{A.8})$$

Heat flow in the Pu-Fe system at any point x is represented by

$$q_x = -kA \frac{dt}{dx} \quad (\text{A.9})$$

Substituting Eq. A.4 for $\frac{dt}{dx}$ and Eq. A.6 for c_1 gives

$$q_x = -kA \left(-\frac{wx}{k} - \frac{\Delta t}{L} + \frac{wL}{2k} \right) \quad (\text{A.10})$$

or

$$k = \frac{qL}{A} + W \left(\frac{L^2}{2} - Lx \right) \frac{1}{\Delta t} \quad (\text{A.11})$$

If q is the heat entering section at $x = 0$, then

$$k = \left(\frac{qL}{A} + \frac{wL^2}{2} \right) \frac{1}{\Delta t} \quad (\text{A.12})$$

In this experiment where the alloy is contained in a tantalum specimen bar, the numerical value of q is equal to the watts input to the specimen heater plus the alpha heating in the Pu-Fe above the section under consideration, minus the heat flow down the tantalum wall as expressed by

$$q_t = k_t A_t \frac{\Delta t}{\Delta x} \quad (A.13)$$

where

k_t = thermal conductivity of tantalum

A_t = cross sectional area of the tantalum wall

$\frac{\Delta t}{\Delta x}$ = temperature gradient across the section under consideration

Equation A.12 was used to calculate the thermal conductivity values for both the solid and liquid states.

Appendix B
DEVIATIONS FROM LINEAR HEAT FLOW IN THE PLUTONIUM CONDUCTIVITY
EXPERIMENTS

by
B. M. Carmichael and M. E. Battat
June 3, 1958

Introduction

In the plutonium conductivity experiments the plutonium alloy was contained in a thin cylindrical tantalum shell. The thermal conductivity of the plutonium alloy was inferred from the measurement of two temperatures at points 2.54 cm apart along the outer vertical tantalum surface, and from a measurement of the total vertical heat flow rate through the tantalum - plutonium alloy configuration. Guard heaters were used to prevent heat from radiating radially out from the outer tantalum surface.

In a two-region cylindrical system of this type, assuming constant conductivities, no internal heat sources, and no end effects, the heat flow is one-dimensional in the vertical direction. In this case the conductivity of the unknown alloy is determined uniquely from the three measured quantities using the two-region solutions to the diffusion equation

$$- K \frac{d^2 T}{dz^2} = 0 \quad (B.1)$$

where K is the conductivity, T is the temperature, and z is the vertical

coordinate. In this report we discuss the effects which might give rise to radial heat flow and thus cause deviations from the above linear heat flow assumption.

Approximate Solution with Radial Heat Flow

In the general case the heat diffusion equation in cylindrical geometry is

$$\frac{1}{r} \frac{\partial}{\partial r} \left(r K \frac{\partial T}{\partial r} \right) + \frac{\partial}{\partial z} \left(K \frac{\partial T}{\partial z} \right) = - w(r, z) \quad (\text{B.2})$$

where r is the radial coordinate, and $w(r, z)$ is the internal heat source density. The geometry and boundary conditions for applying Eq. B.2 are illustrated in Fig. B.1. We assume the conductivities are given by

$$K_p = K_o [1 + \alpha (T - T_o)] \text{ w-cm/cm}^2\text{-}^\circ\text{C} \quad (\text{B.3})$$

$$K_t = 0.562 \text{ w-cm/cm}^2\text{-}^\circ\text{C} \quad (\text{B.4})$$

for the plutonium alloy and tantalum respectively, where K_o is the alloy conductivity at the temperature T_o , α is the fractional change in conductivity per $^\circ\text{C}$, and the subscripts p and t are used to denote quantities in the alloy and tantalum regions, respectively. Experimental results support the validity of the above assumptions (Eqs. B.3 and B.4).

Denoting the two regions as R_p and R_t , we make the change of variables in R_p

$$v_p = \frac{K_o}{2\alpha} [1 + \alpha (T_p - T_o)]^2 \quad (\text{B.5})$$

and obtain from Eq. B.2 the regional equations

$$\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial v_p}{\partial r} \right) + \frac{\partial^2 v_p}{\partial z^2} = - w_p \quad (\text{B.6})$$

$$a = 0.815 \text{ CM}$$

$$b = 0.885 \text{ CM}$$

$$L = 2.54 \text{ CM}$$

Fig. B.1. The Geometry and Boundary Conditions of the Pu-Fe System.

$$\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial T_t}{\partial r} \right) + \frac{\partial^2 T_t}{\partial z^2} = 0 \quad (\text{B.7})$$

since the only internal heat source is the natural alpha heat source in the plutonium which is denoted as w_p in Eq. B.6. To separate the radial and vertical parts of Eqs. B.6 and B.7, we assume

$$T_p(r, z) = T_{pr}(r) + T_{pz}(z) \quad (\text{B.8})$$

$$T_t(r, z) = T_{tr}(r) + T_{tz}(z)$$

From Eq. B.5 we have

$$v_p = \frac{K_o}{2\alpha} [1 + \alpha (T_{pr} + T_{pz} - T_o)]^2$$

We expect T_{pr} to be small and it will be seen in the Numerical Solution section that T_{pr} varies only by about 0.1°C over the thickness of the bar. Thus

$$\begin{aligned} v_p &\approx \frac{K_o}{2\alpha} [1 + \alpha (T_{pz} - T_o)]^2 + K_o [1 + \alpha (T_{pz} - T_o)] T_{pr} \\ &\approx v_{pz} + K_p T_{pr} \end{aligned}$$

where v_{pz} and K_p follow from Eqs. B.3 and B.5. We assume further that the radial heat flow is generated by the fictitious heat sources w'_p and w'_t in R_p and R_t , respectively, such that the Eqs. B.6 and B.7 are separated into

$$\frac{1}{r} \frac{d}{dr} \left(r \frac{dT_{pr}}{dr} \right) = - w'_p / K_p$$

$$\frac{1}{r} \frac{d}{dr} \left(r \frac{dT_{tr}}{dr} \right) = - w'_t / K_t$$

(B.9)

$$\frac{d^2 v_{pz}}{dz^2} = -(w_p - w'_p)$$

$$\frac{d^2 T_{tz}}{dz^2} = w'_t / K_t$$

Applying the boundary conditions

$$T_{tr}(b) = 0, \quad \left(\frac{dT_{tr}}{dr} \right)_b = 0 \quad (B.10)$$

$$T_{tr}(a) = T_{pr}(a), \quad K_t \left(\frac{dT_{tr}}{dr} \right)_a = K_p \left(\frac{dT_{pr}}{dr} \right)_a$$

to the radial equations in Eq. B.9, we obtain solutions

$$T_{pr} = T_c - \frac{w'_p r^2}{4K_p} \quad (B.11)$$

$$T_{tr} = \frac{w'_t}{2K_t} \left[\frac{b^2 - r^2}{2} - b^2 \ln(b/r) \right]$$

where

$$w'_t = - \frac{a^2}{b^2 - a^2} w'_p \quad (B.12)$$

and T_c , the central temperature in R_p , is given by

$$T_c = \frac{w'_p a^2}{4} \left\{ \frac{1}{K_p} - \frac{1}{K_t} \left[1 - \frac{2b^2 \ln(b/a)}{(b^2 - a^2)} \right] \right\} \quad (B.13)$$

Thus w'_p is the single radial flow parameter left undetermined. From Eq. B.11 it is seen that the radial heat flux originates from the

source w'_p in R_p and terminates in the sink w'_t in R_t . However, it is seen in the vertical equations of Eq. B.9 that w'_p is a sink and w'_t is a source from the point of view of the vertical fluxes.

Applying the boundary conditions that the temperature and flux at $z = 0$ are T_{po} and q_{po} , respectively, to the vertical equation in R_p , we obtain the solution

$$\frac{K_o}{2\alpha} \left\{ 1 - [1 + \alpha (T_z - T_{po})]^2 \right\} = q_{poz} + (w_p - w'_p)z^2/2 \quad (B.14)$$

Letting $T_z = T_L$ at $z = L$ and defining

$$\Delta T_z = T_{po} - T_L \quad (B.15)$$

we find

$$\begin{aligned} \Delta T_z K_o \left(1 - \alpha \frac{\Delta T_z}{2} \right) &= \Delta T_z \bar{K}_p \\ &= q_{po}L + (w_p - w'_p) \frac{L^2}{2} \end{aligned} \quad (B.16)$$

where $\bar{K}_p$ is the conductivity at the average vertical temperature in R_p . Similarly

$$\Delta T_z K_t = q_{to}L - (w'_t) \frac{L^2}{2} \quad (B.17)$$

In addition, the total vertical heat flow is

$$Q = A_p q_{po} + A_t q_{to} \quad (B.18)$$

where A_p and A_t are the cross sectional areas in R_p and R_t . We combine Eqs. B.16, B.17, and B.18 to eliminate q_{po} and q_{to} and use Eq. B.11 to eliminate w'_t . This gives for the R_p conductivity

$$\overline{K}_p = \frac{LQ}{A_p \Delta T_z} - K_t \frac{A_t}{A_p} + \frac{w_p L^2}{2 \Delta T_z} \quad (B.19)$$

Since the radial heat sources cancel out of Eq. B.19, the equation becomes identical with the expression obtained for $\overline{K}_p$ from the linear heat flow assumption.

Numerical Solution

A code for an IBM 704 computer was devised for solving Eq. B.2 by the relaxation method. Two problems were solved by this method. In both problems the data used were those of an experimental run made February 3, 1958, on Pu-Fe alloy in the solid state. This case was chosen since α is a maximum in the solid state region and the source effects ($w_p = 0.03432$ w/cc) would be larger compared to the heat source of this run ($Q = 8.73$ w) than they would be for higher temperatures, e.g., in the molten state.

In the first problem the effects of the geometric perturbations below the alloy in the experimental arrangement were investigated. The geometry and results for this case are illustrated in Fig. B.2. It is seen that the perturbations in the isothermal lines damp out quite rapidly in the region of the alloy and should have no significant effect in the region of measurement.

In the second problem, a two-region case similar to that shown in Fig. B.1 was examined except the overall length of the system was increased to 10.5 cm, which was the height of the alloy in the experiment. A flat radial temperature was assumed at the top and the problem was set up to determine the amount of radial flow developing over the length of the system. The radial temperature plots obtained are shown in Fig. B.3. Three radial plots are given in the figure at three different vertical levels; namely, at levels 2.54, 5.08, and 7.62 cm from the top of the configuration.

The crosses shown on the plots are points calculated from the equations of the preceding section. The source w'_p was calculated using Eq. B.13 and normalizing T_c to the central temperature of the traverse

Fig. B.2. The Temperature Distribution at Bottom of Pu-Fe Specimen.

Fig. B.3. Radial Temperature Distributions in Pu-Fe Specimen.

at the 5.08 cm level. Equations B.11 are then used to calculate the temperatures at the other points. The differences in the three traverses are due to the fact that K_p has three different values from the top to the bottom traverse. The agreement obtained serves as a check on the theory of the preceding section.

Appendix C
CHEMICAL ANALYSES

Analysis of the 304 Stainless Steel Specimen Bar

Cr	18.51%
Ni	9.09
C	.053
Mn	.67
P	.025
S	.028
Si	.53

Analysis of the Tantalum

O	.0140%
N	.0060
H	.0025
C	.0025
Ti	.0001
Be	.0002
B	.0003
Na	.0003
Mg	.0001
Al	.0010
Si	.0003
Ca	.0001
Ti	.0005
V	.0020
Cr	.0010

Mn	.0003%
Fe	.0010
Co	.0020
Ni	.0010
Cu	.0001
Zn	.0100
Sr	.0020
Zr	.0030
Cb	.0300
Mb	.0050
Ag	.0005
Sn	.0030
Ba	.0010
W	.0100
Pb	.0030
Bi	<u>.0010</u>
Total Impurities	.1038%