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# THE VISCOSITY OF A LIQUID PLUTONIUM-IRON EUTECTIC ALLOY

by
L. J. Wittenberg, L. V. Jones, D. Ofte

Mound Laboratory
Operated by
The Monsanto Chemical Company
for the
United States Atomic Energy Commission

## SUMMARY

The viscosity of a liquid plutonium-iron eutectic alloy, which contains 9.5 atom per cent iron and melts at 411°C., was determined up to 808°C. at Mound Laboratory by an oscillating cup viscosimeter. This type of apparatus employed a right-circular cylindrical cup containing the liquid under investigation attached to a torsion fiber. The dampening effect of the liquid upon the normal oscillations of the pendulum was a function of the viscosity of the liquid. The amplitudes of the oscillations of the pendulum were measured by a photographic technique. The periods of the oscillations were determined by an automatic timing mechanism. The reliability of the viscosimeter was demonstrated by following the expected function of the viscosity of liquid lead and bismuth over a larger temperature range than was previously reported.

It was necessary to employ two different periods for the pendulum before the viscosity of the plutonium alloy could be calculated over the reported temperature range. The viscosity of the alloy increased as expected with decreasing temperatures, from 6.14 centipoise at 808°C. to 22.4 centipoise at 433°C., and the activation energy for viscous flow was determined to be 5237 calories per mole. Both the viscosity values and the activation energy are large values for liquid metal systems.

## INTRODUCTION

One concept of a fast-breeder nuclear power plant has proposed the use of plutonium or an alloy of plutonium as a liquid fuel [1]. The plutonium-iron eutectic alloy containing 9.5 atom per cent iron has a melting point of 411°C. (Figure 1) [2]. Engineering design of a reactor using this liquid fuel system requires knowledge of the viscosity of this alloy. The method of measuring the viscosity of liquid metals and the data obtained at Mound Laboratory for the plutonium-iron eutectic alloy up to 800°C. are presented in this paper.

## APPARATUS

The oscillating cup viscosimeter was selected because of the following advantages:

1° It is a simple system to operate;

2° The molten metal under investigation is contained in a sealed cup;

3° It is an absolute method which does not need to be calibrated at high temperatures.

The oscillating cup viscosimeter is a torsion pendulum that consists of a right-circular cylindrical cup which is filled with the liquid and attached to a torsion fiber. The normal oscillations of the pendulum are dampened by the drag of the liquid on the side-walls of the cup and the time per cycle is increased. The amplitudes of the oscillations and the times of the cycles of the empty and filled cups are measured.

The oscillating pendulum is the main part of the viscosimeter (Figure 2). The pendulum consists of a mono-filament torsion fiber and the inertial system. Molybdenum wires of either 0.005 or 0.010 inch diameter and eight inches long are used as the torsion fibers.

The inertial system (Figure 2) consists of the cup containing the fluid, a stainless steel supporting rod, a disc with lines accurately scribed every 0.5 millimeter along its periphery, a steel inertial bar, and a mirror formed by

polishing a 1/4-inch wide section of the central aluminum shaft. A pin-vise centered in the top of the shaft engages the torsion fiber. The upper end of the fiber is attached to a microchuck, which is anchored to a large brass disk.

Each cup is fabricated from tantalum tubing, 5/8 inch outside diameter and 0.020 inch wall thickness, with the inner surface of the wall honed to a microfinish. The top cap, 0.040 inch thick, and the adapter for the shaft are fabricated from a single piece of tantalum rod. The top cap is attached by inert gas-arc welding before the hole is bored to accommodate the shaft so that the axis of the shaft coincides with the axis of the cup. The bottom closure plug, fabricated from tantalum rod, is a recessed cap, 0.040 inch thick on the bottom with 0.020 inch side-walls machined to be a tight fit in the tubing. The bottom closure plug is welded in place after the crucible is filled with the alloy to be measured.

The assembled suspension system is placed in the viscosimeter apparatus so that the upper end of the torsion fiber is firmly held while the inertial system oscillates freely (Figure 3). The cup containing the fluid is in the isothermal region of the furnace while the remainder of the suspension system is above the furnace area. A thermal radiation shield, containing a hole only slightly larger than the shaft diameter, is placed on a water-cooled brass

plate at the top of the furnace area. The top assembly is covered with a glass envelope, 4-1/2 inches outside diameter with 1/4-inch walls. A brass cap covers the upper end of this tube. Within the furnace area, the cup is centered in a 1-3/4 inch stainless steel tube, which is connected by copper tubing to a high vacuum system.

All of the demountable joints of the apparatus are assembled with the use of neoprene rubber "O"-ring seals. Water-cooled plates protect the rubber seals adjacent to the furnace area. With these seals a pressure of $5 \times 10^{-6}$ mm. of mercury is maintained within the apparatus. While it is not essential for the oscillating system to operate at a reduced pressure, the reduced pressure operation is more convenient for the following reasons:

1° The effect of air-drag on the suspension system is eliminated;

2° Convection currents, which tend to disturb the suspension system, are not formed;

3° The oxidation of the tantalum is reduced.

The temperature of the sample is determined by means of a nickel shielded chromel-alumel thermocouple placed at the height of the liquid but displaced radially to a position adjacent to, but not touching the cup. Since the cup and the thermocouple are heated only by radiation, and their position, size, and emissivity characteristics are different,

the temperature indicated by the thermocouple is different from that of the liquid metal. It was necessary, therefore, to calibrate this thermocouple against a second thermocouple which was placed in liquid metal inside of the cup under conditions identical to those encountered during a determination.

The temperature control of the furnace is achieved by means of a Foxboro potentiometer controller whose sensing thermocouple is placed in the furnace windings. A variable transformer with a differential controller is used to regulate the voltage available to the 220-volt, 1000-watt resistance wound electric furnace. By positioning the contact arm of the variable transformer so that the heating and cooling cycles are of equal length of time, the temperature variations in the apparatus are controlled to within plus or minus one degree Centigrade.

Since a possibility of the spread of radioactive material exists if a sealed cup leaks during an experiment, the entire viscosimeter is placed inside of an alpha handling, gloved, stainless steel box two feet deep, six feet long, and six feet high, containing two full-height clear plastic windows. The apparatus in the box is mounted on a heavy framework resting on foundation rock to eliminate external vibrations which disturb the oscillating motion of the pendulum.

The oscillation of the torsion pendulum is initiated by an electromagnet whose magnetic field exerts a tangential force on the ferromagnetic inertial bar located on the suspension system. The electromagnet is energized periodically to gradually increase the amplitude of each succeeding oscillation up to an amplitude of 50 degrees when the 0.005 inch diameter fiber is used and 13 degrees when the 0.010 inch diameter fiber is used. This method of initiation avoids the occurrence of nonlinear effects which are encountered when the oscillations are initiated by orienting the pendulum to its largest amplitude and releasing it.

The amplitudes of the oscillations of the suspension system, observed by the motion of the graduated disk, are recorded photographically with a motion picture camera sighted through a telescope equipped with a vertical cross-hair. By the use of a measuring system on the optical projection the smallest division of the aluminum disk is divided into 180 subdivisions and can be read to within $\pm 2.5$ subdivisions. Six successive oscillations are recorded per series. The mathematical solutions of this problem require the calculation of the logarithmic decrement, which is the Naperian logarithm of the ratio of successive amplitudes in the same direction from the midpoint of the oscillations. The logarithmic decrements reported are the average of at least two independent series of amplitude readings.

An automatic timing system is used to determine the periods of the oscillations. A beam of light, reflected from the mirror of the suspension system, actuates a photoelectric cell at exactly the midpoint of the oscillation. The signal from the photoelectric cell starts a timing mechanism which continues until automatically stopped after the desired number of oscillations. The timing mechanism is a 60-cycles-per-second oscillator which is counted on a scalar unit. The standard deviation in measuring a period of 11 seconds amounted to 0.0002 second for ten measurements. As a result of fatigue, or possibly creep in the wire, the period is reproducible to only 0.005 second over a period of several weeks.

The moment of inertia of the suspension system with an empty cup is determined by measuring the periods of the oscillations with and without two identical inertial weights which are accurately positioned, one on each arm of the inertial bar of the suspension system. The moment of inertia of the suspension system is calculated as follows:

$$I_E = \frac{I_w t_E^2}{t_w^2 - t_E^2} \qquad (1)$$

where $I_E$ = moment of inertia of the suspension system with an empty cup.

$I_w$ = effective moment of inertia of the inertial weights on the inertial bar.

$t_E$ = period of oscillation of the suspension system with an empty cup.

$t_w$ = period of oscillation of the suspension system with the inertial weights on the inertial bar.

## EXPERIMENTAL PROCEDURE

The plutonium alloys were prepared by weighing together the required amounts of each element, heating in a vacuum induction furnace to a sufficient temperature above their liquidus to achieve good mixing, and casting into cylindrical rods. The compositions reported for these alloys were determined by chemical analyses from samples of these castings. These rods were machined into right-circular cylinders about 0.550 inch in diameter and two inches long, equivalent to about nine cubic centimeters of liquid when the sample melted.

The plutonium alloy was loaded into a crucible in an open uncontaminated fume hood by slipping the rod into the crucible, using a funnel made of metal foil to prevent contamination of the open end of the crucible. The crucible was flushed with helium gas during the loading operation, and the end cap was immediately inserted to its full depth. The crucible was sealed by inert gas-arc welding the end cap in place. By this handling technique, the outside of the crucible showed no direct alpha contamination after sealing.

The sample in the viscosimeter was subjected to an initial extended heat treatment at elevated temperatures to completely wet the tantalum surface with the liquid before a determination was made. These heat treatments were as follows: 24 hours at 600°C. for Sample No. 1, Table 1, 24 hours at 700°C. for Sample No. 2, and 12 hours at 600°C. for Sample No. 3. The temperature was decreased then to the

next lower value and the system equilibrated for six hours before a new determination was made. The temperature was decreased in this manner until the solidus was approached except in the case of Sample No. 2, where the measurements were made at 750 and 800°C. after the lower temperature determinations were made but before the solidus had been reached. The schedule was continued without interruption to prevent solidification of the alloy during a determination.

The crucible was usually distorted by the alloy upon cooling to room temperature. Examination of the alloy after an experiment showed it to be clean metal, and the tantalum crucible showed no detrimental corrosion. The alloy rod was removed from Sample No. 2 and inserted in a new crucible to form Sample No. 3.

## RESULTS AND DISCUSSION

It is necessary to know the relationship between the kinematic viscosity, the logarithmic decrement, and the physical dimensions of the system to use this apparatus as an absolute method for measuring viscosity. This relationship is made difficult by the fact that the physical effects of the liquid on the pendulum change with the viscosity. In the case of a liquid of low viscosity with the pendulum moving rapidly, the vast majority of the liquid does not move with the cup. Kestin and Newell [3] have called this the "large-cup" situation for which the logarithmic decrement increases with an increase in viscosity, and the period is only slightly increased over that of the empty cup. As its viscosity increases, the liquid becomes more like a solid, so that more of the liquid moves with the oscillating cup. Since a solid does not have a logarithmic decrement, the logarithmic decrement for a viscous liquid decreases as the viscosity increases. The moment of inertia of the system in this case is increased by the amount of fluid moving with the cup causing a large increase in the period. This latter situation is called the "small-cup" behavior.

A graphical presentation of the physical parameters involved, Figure 4, was suggested by Mr. K. C. Jordan of Mound Laboratory, although it had been considered by Priss [4] and Beckwith and Newell [5]. The symbols used are identified as follows:

$\delta_L$ = logarithmic decrement of the suspension with a full cup.

$\delta_E$ = logarithmic decrement of the suspension with an empty cup.

$I_E$ = moment of inertia of the suspension system with an empty cup.

$I_F$ = moment of inertia of fluid.

$\eta$ = viscosity.

$T_L$ = period of oscillation of the suspension system with a full cup.

$\rho$ = density.

R = radius of the cup.

Actually, there is not one curve but a family of curves for each value of the ratio $I_E/I_F$. In an attempt to consolidate this family of curves into one line, the quantity $f(I_F)$, where "f" represents the fraction of fluid which is responsive to the movement of the cup, has been used. Its value varies from zero on the left to one on the right. Since it is a difficult quantity to evaluate, it is much more convenient to use one of the mathematical solutions of the system to determine the viscosity rather than the graphical presentation. It is necessary to note, however, that none of the treatments give satisfactory viscosity values near the maximum of the curve.

Several mathematical treatments have been given for the oscillating cup viscosimeter of the "large-cup" behavior since the apparatus was first proposed by Meyer in 1891 [6]. The first complete treatment which yielded a simplified solution for the problem was given by Andrade and Chiong [7] for a sphere filled with the fluid. Their treatment was modified to include a right-circular cylinder enclosing the fluid by Hopkins and Toye [8], Toye and Jones [9], Roscoe [10] and Shvidkovskii [11]. All the viscosities reported in this paper were calculated by the equations derived by Roscoe.

In contrast to the several investigators who have considered the "large-cup" viscosimeter, only Priss has given a satisfactory solution for the "small-cup" viscosimeter, which has proven to be reliable for viscous oils.

For nonassociated liquids the variation of viscosity with temperature [12, 13] has been shown to follow the relationship,

$$\eta = Ae^{E/RT} \qquad (2)$$

where $\eta$ = viscosity coefficient.

A = a constant.

E = activation energy for viscous flow.

R = the gas constant.

T = temperature in degrees absolute.

It can be seen that a straight line should be generated when the logarithm of $\eta$ is plotted against 1/T.

The reliability of the apparatus was determined by measuring the viscosity of liquid lead and bismuth over a range of temperatures (Figures 5 and 6) in stainless steel crucibles of identical size to the tantalum crucibles. It can be seen that the viscosity values determined by the present method follow the expected dependency of temperature over a larger temperature range than the literature values [14].

Sample No. 1 of the plutonium-iron alloy was inserted in the viscosimeter. The physical dimensions of the system are shown in Table 1. At the highest temperature for this sample, 707°C., the logarithmic decrement is almost three times larger than for liquid lead at this temperature, which corresponds to nearly a six-times increase in the viscosity of the alloy as compared to lead, Table 2. The liquid density values reported in Table 2 were determined by Knight [15]. The logarithm of the viscosity of the alloy increased as a first-order dependency of the reciprocal temperature as the temperature decreased to about 550°C. Below this temperature it was impossible to calculate meaningful viscosity values for the alloy by using either the mathematical solution of Roscoe or the graphical interpretation (Figure 4).

A second sample of alloy was prepared with a slightly different composition than the first sample (Table 1). Although this sample was taken to 808°C, it reacted in a

similar manner to the first sample in that a high viscosity was found and the logarithm of the viscosity increased as a first-order function of the reciprocal temperature as the temperature decreased to 604°C. (Table 2). Below 600°C. it was impossible to interpret the experimental data.

It is necessary to return to Figure 4 to understand why it was not possible to interpret the data for Samples No. 1 and 2 at the lower temperatures. For liquid lead and bismuth in a system of the present dimensions the abscissa values are in the range of 0.2. For the plutonium-iron alloy with its much larger viscosity, the abscissa values are 0.45 and larger, which are near the maximum of the curve. Since it is known that calculated viscosity values are not reliable near the maximum of the curve, it is necessary to change the dimensions of the system so that the Pu-Fe alloy abscissa value would be in the range of 0.2. This change was accomplished by using a torsion wire with a period one-fourth as long as the previous fibers for Sample No. 3 (Table 1).

The viscosity values determined by Sample No. 3 agreed with the previous determinations within one per cent at 597°C. and four per cent at 555°C., Table 2. In addition, the logarithm of the viscosity increased as a first-order dependency with the reciprocal temperature as the temperature decreased to 433°C.

The viscosity values from the three series of measurements are presented graphically in Figure 7 where the straight line has been drawn as a least squares function of all 17 values. It should be noted that this alloy is much more viscous than most liquid metal systems previously reported. The activation energy for viscous flow of this alloy is 5237 calories per mole which is also greater than that reported for most liquid metal systems.

This plutonium-iron alloy has proven to have unique characteristics among liquid metal systems. Consequently, this research is being extended at Mound Laboratory to include the viscosity of plutonium metal and other plutonium alloys. In addition, investigation is continuing to extend the range of applicability for the oscillating cup viscosimeter.

TABLE 1

PHYSICAL DIMENSIONS FOR EACH SYSTEM

| Sample No. | Moment of Inertia of Empty Cup (gm. $cm.^2$) | Radius of Cup at 25°C. (cm.) | Weight of Pu-Fe Alloy (gm.) | Atom Per Cent Iron | Molybdenum Fiber Diameter (in.) | Period with Empty Cup (sec.) |
|---|---|---|---|---|---|---|
| 1 | 412.70 | 0.7413 | 150.603 | 10.43 | 0.005 | 11.049 |
| 2 | 408.80 | 0.7132 | 139.55 | 9.81 | 0.005 | 11.023 |
| 3 | 409.27 | 0.7424 | 139.26 | 9.81 | 0.010 | 2.715 |

TABLE 2

DEPENDENCE OF VISCOSITY UPON TEMPERATURE
FOR Pu-Fe ALLOY

| Run Number | Temperature °C. | Logarithmic Decrement ($\delta_L - \delta_E$) | Period Seconds | Density gm./cc. | Viscosity Centipoise |
|---|---|---|---|---|---|
| 1 | 707 | 0.084301 | 11.244 | 15.63 | 7.36 |
| 1 | 654 | 0.087085 | 11.254 | 15.72 | 8.84 |
| 1 | 603 | 0.090385 | 11.273 | 15.80 | 10.2 |
| 1 | 553 | 0.093535 | 11.298 | 15.88 | 11.4 |
| 2 | 808 | 0.069602 | 11.304 | 15.37 | 6.14 |
| 2 | 759 | 0.071501 | 11.308 | 15.53 | 6.57 |
| 2 | 706 | 0.074228 | 11.310 | 15.63 | 7.51 |
| 2 | 656 | 0.076533 | 11.320 | 15.72 | 8.71 |
| 2 | 604 | 0.078925 | 11.332 | 15.80 | 9.64 |
| 3 | 597 | 0.052195 | 2.720 | 15.80 | 10.3 |
| 3 | 555 | 0.055281 | 2.724 | 15.88 | 11.9 |
| 3 | 532 | 0.058358 | 2.725 | 15.92 | 13.4 |
| 3 | 511 | 0.059846 | 2.731 | 15.96 | 14.7 |
| 3 | 494 | 0.062538 | 2.739 | 16.00 | 16.5 |
| 3 | 474 | 0.064760 | 2.747 | 16.02 | 18.5 |
| 3 | 451 | 0.067860 | 2.755 | 16.06 | 20.8 |
| 3 | 433 | 0.069834 | 2.757 | 16.10 | 22.4 |

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Pu-Fe EQUILIBRIUM DIAGRAM

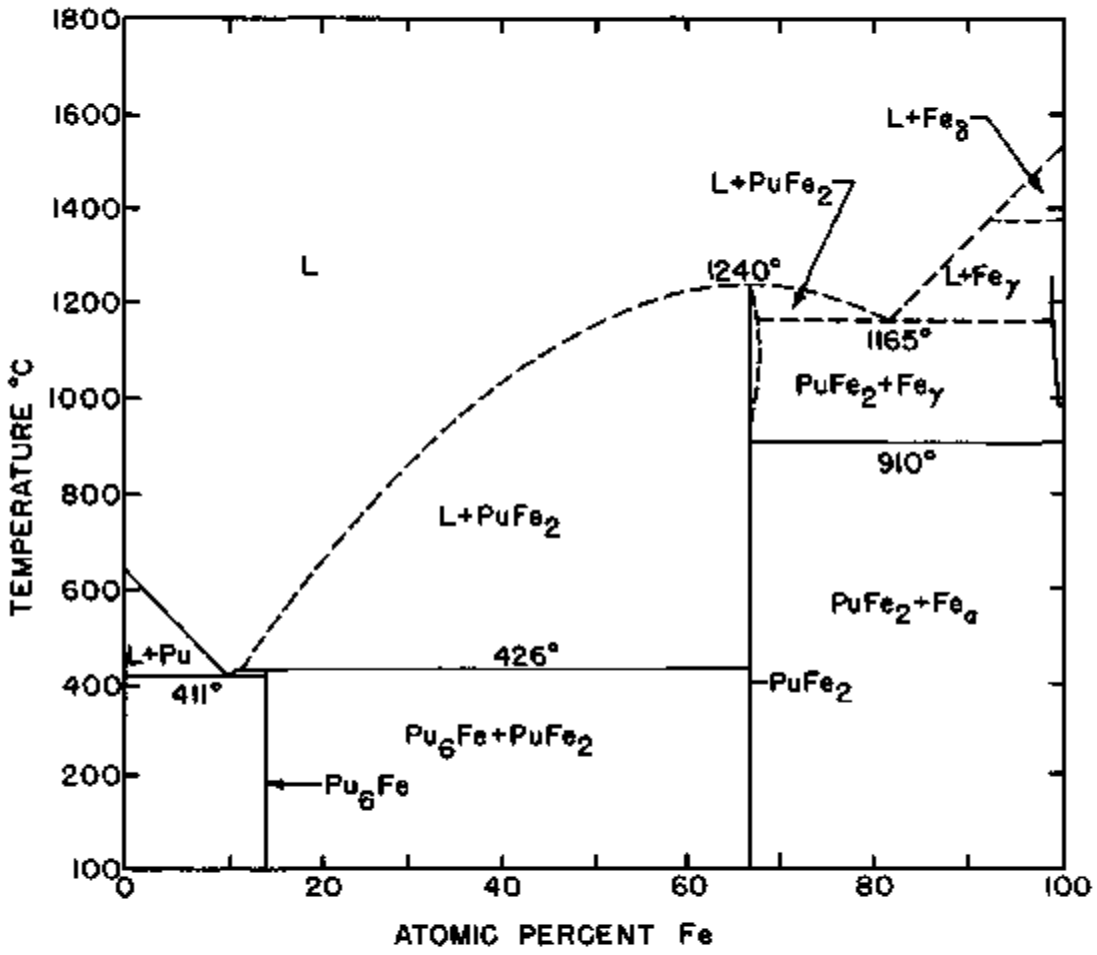


FIGURE 1

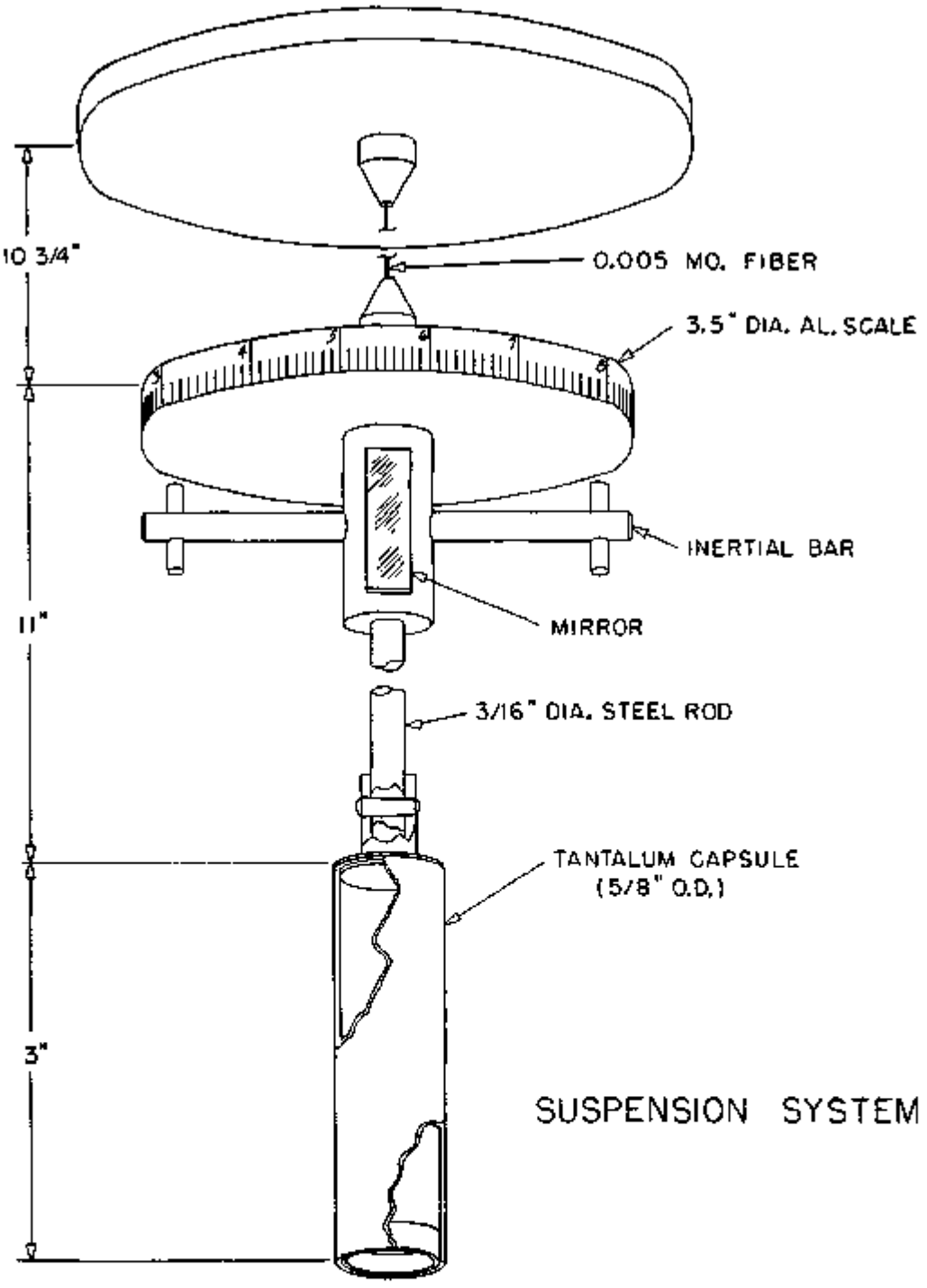
10 3/4"
0.005 MO. FIBER
3.5" DIA. AL. SCALE
INERTIAL BAR
11"
MIRROR
3/16" DIA. STEEL ROD
TANTALUM CAPSULE
(5/8" O.D.)
3"
SUSPENSION SYSTEM

FIGURE 2

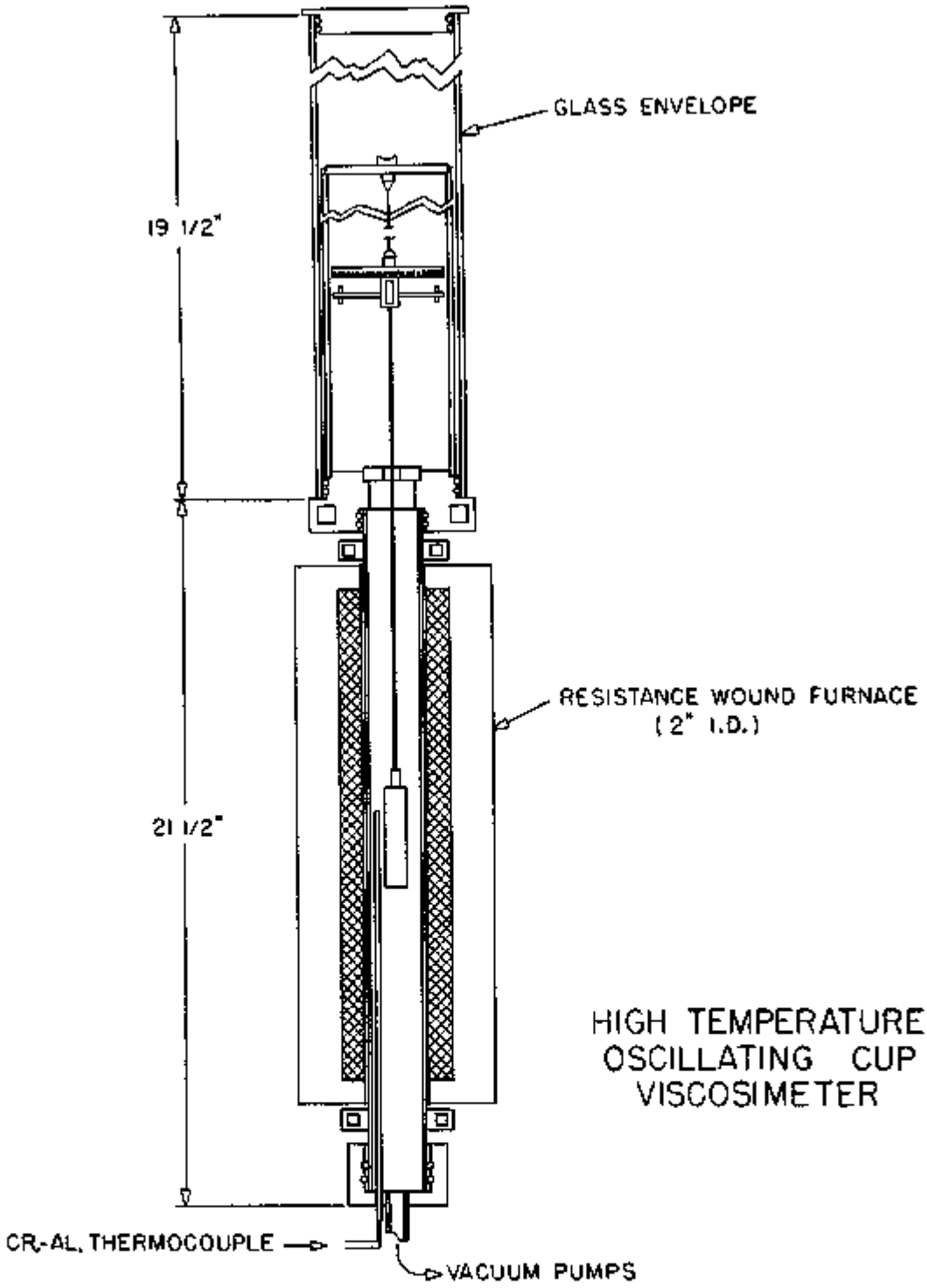
GLASS ENVELOPE
19 1/2"
RESISTANCE WOUND FURNACE
(2" I.D.)
21 1/2"
HIGH TEMPERATURE
OSCILLATING CUP
VISCOSIMETER
CR-AL. THERMOCOUPLE
VACUUM PUMPS

FIGURE 3

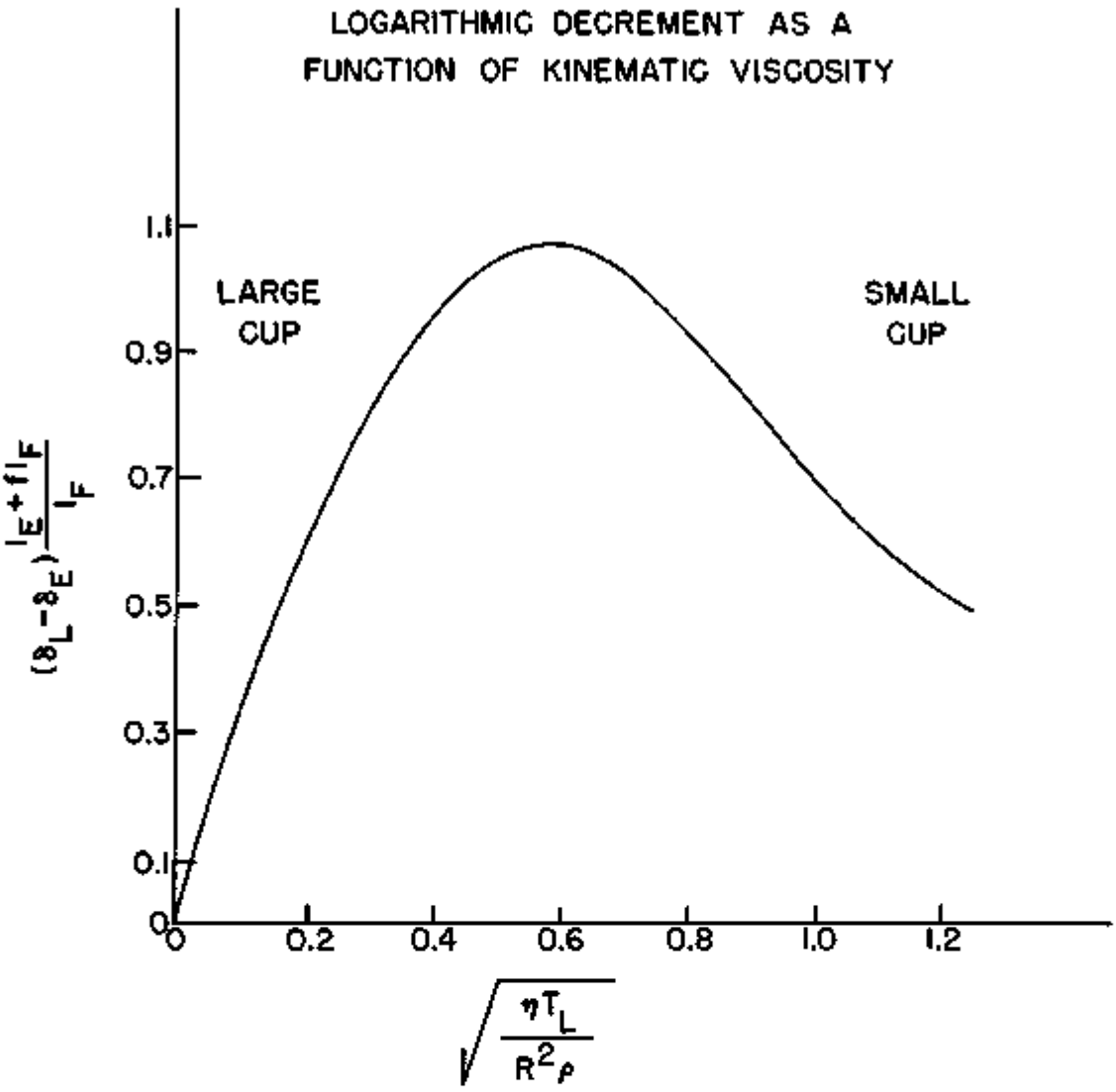
LOGARITHMIC DECREMENT AS A
FUNCTION OF KINEMATIC VISCOSITY
LARGE
CUP
SMALL
CUP
1.1
0.9
0.7
0.5
0.3
0.1
0
0.2
0.4
0.6
0.8
1.0
1.2
$(\delta_L-\delta_E)\frac{I_E+fI_F}{I_F}$
$\sqrt{\frac{\eta T_L}{R^2\rho}}$

FIGURE 4

VISCOSITY OF BISMUTH AS A
FUNCTION OF ABSOLUTE TEMPERATURE

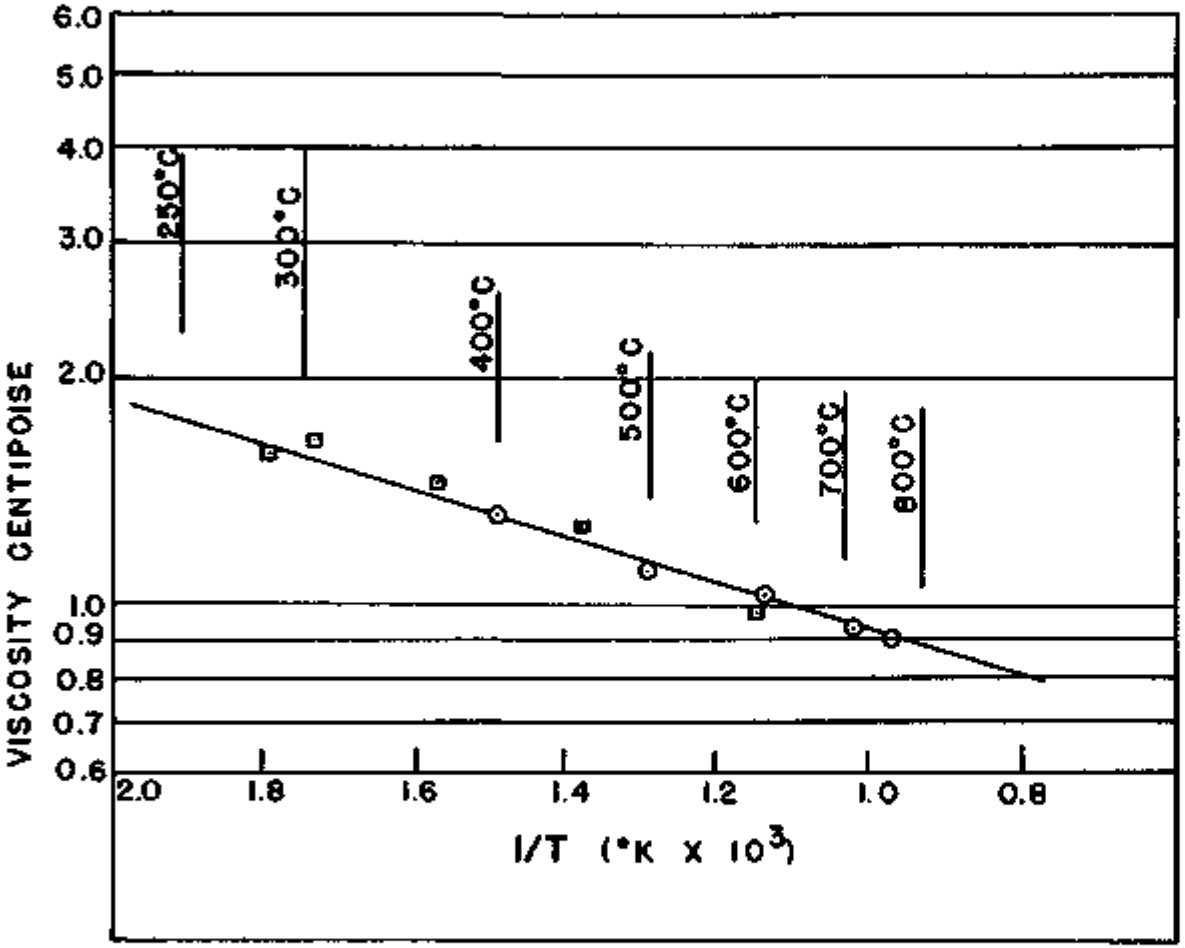


FIGURE 5

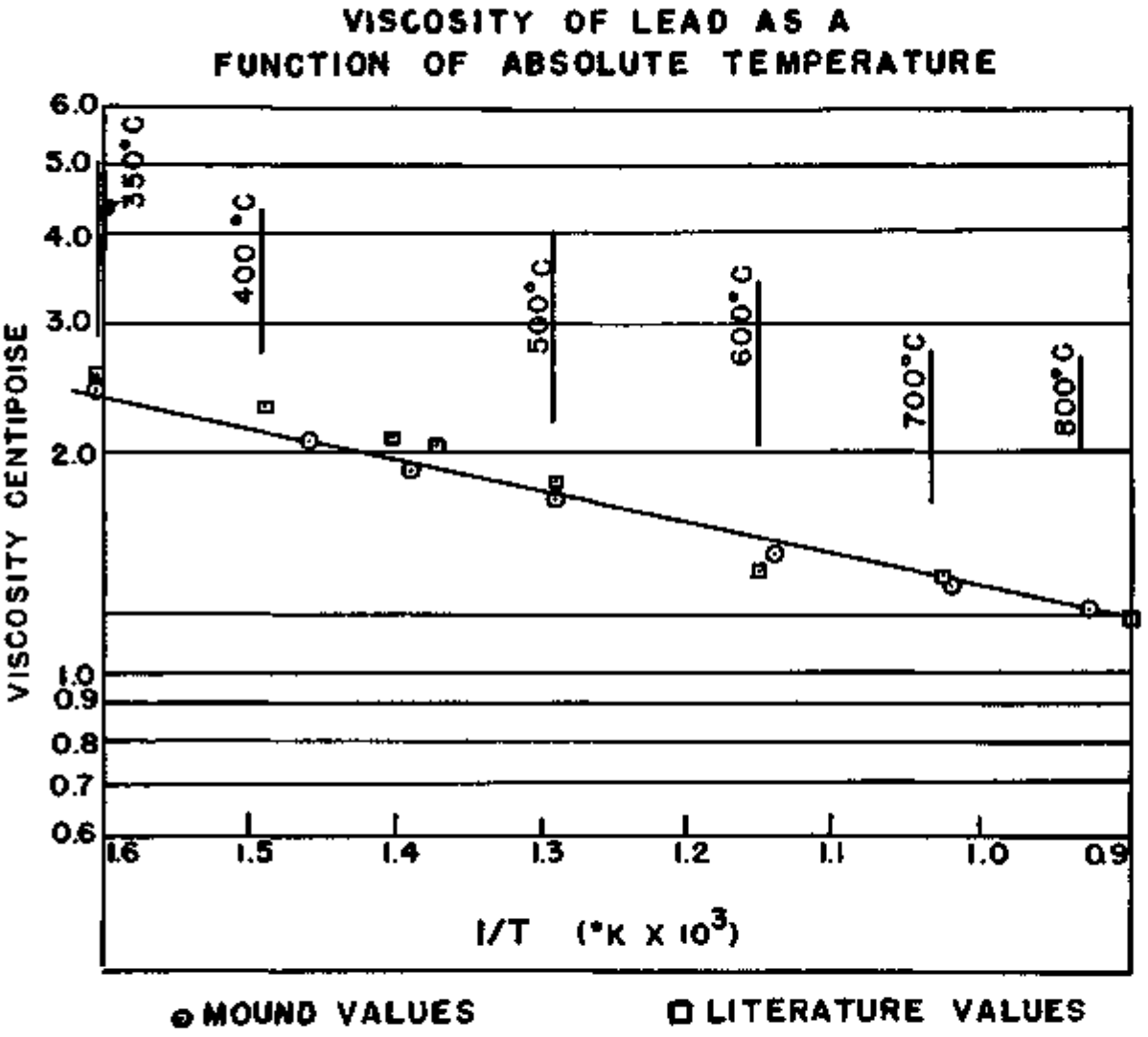
VISCOSITY OF LEAD AS A
FUNCTION OF ABSOLUTE TEMPERATURE
VISCOSITY CENTIPOISE
6.0
5.0
4.0
3.0
2.0
1.0
0.9
0.8
0.7
0.6
350°C
400°C
500°C
600°C
700°C
800°C
1.6
1.5
1.4
1.3
1.2
1.1
1.0
0.9
1/T (°K X 10³)
MOUND VALUES
LITERATURE VALUES

FIGURE 6

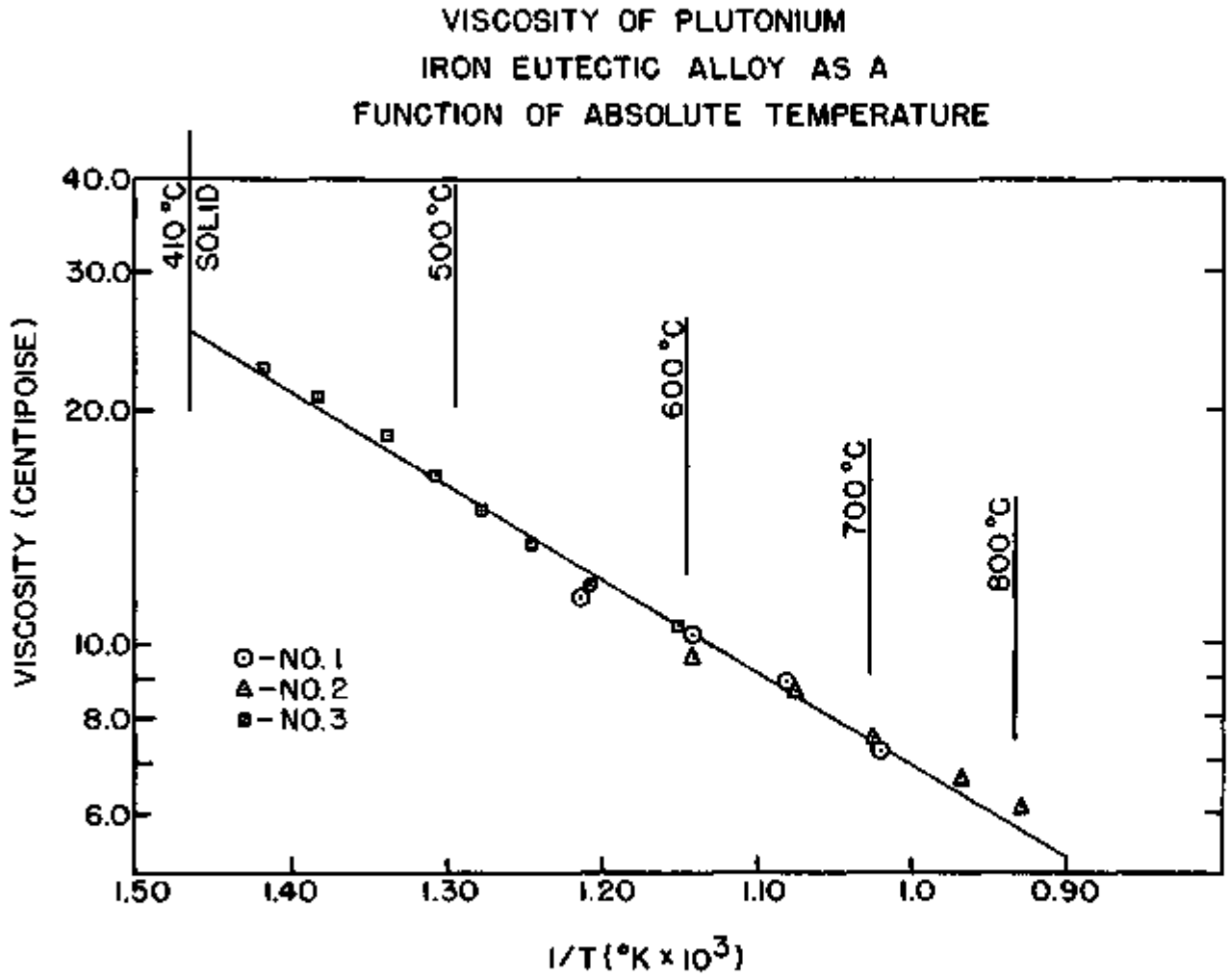
VISCOSITY OF PLUTONIUM
IRON EUTECTIC ALLOY AS A
FUNCTION OF ABSOLUTE TEMPERATURE
VISCOSITY (CENTIPOISE)
40.0
30.0
20.0
10.0
8.0
6.0
410 °C
SOLID
500 °C
600 °C
700 °C
800 °C
⊙-NO. 1
▲-NO. 2
■-NO. 3
1.50
1.40
1.30
1.20
1.10
1.0
0.90
1/T (°K x 10³)

FIGURE 7