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MEASUREMENT OF PLUTONIUM LIQUID DENSITY

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For The Atomic Energy Commission
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Chief, Declassification Branch

TECHNOLOGY--PLUTONIUM

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TECHNOLOGY--PLUTONIUM

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ABSTRACT

The density of molten plutonium has been determined by measuring the volume of a known weight of liquid plutonium in a quartz tube. The method involved the use of an X-ray shadowgraph technique. The density was found to be 16.5 ± 0.08 gm/cm^3 at about 665° C.

From the shapes of two different plutonium menisci, as observed in the shadowgraphs, a rough value of 100 dyne/cm was calculated for the surface tension. Also, a rough value of 5×10^{-5} was calculated for the coefficient of thermal expansion. Both of these were obtained in the region around 700° C.

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ACKNOWLEDGMENTS

It is a pleasure to acknowledge the discussions, criticisms, and assistance of T. A. Sandenaw. Also, this experiment would have been completely unsatisfactory had it not been for the application of the X-ray technique to this problem by N. C. Miller of Group GMX-1.

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The density of molten plutonium has been determined by a method in which a sample of the metal of known weight was melted in a quartz funnel under vacuum and allowed to run down into a quartz tube. The tube had been previously calibrated with mercury. The quartz funnel and the tube were enclosed in a nickel tubular radiation shield which was in turn contained in a quartz vacuum jacket. The nickel tube was heated by an induction coil placed outside the quartz jacket. The calibrated quartz tube had a small thermocouple well in the bottom.

A sketch of the apparatus is shown in Fig. A. Two vertical slots (not shown) were on opposite sides of the nickel tube in the region of the plutonium meniscus. These were 1 in. long and 3/16 in. wide. An induction coil of many turns was pulled apart as shown to permit observation of the plutonium meniscus through the slots in the nickel tube. The quartz washer provided insulation of the thermocouple leads from the nickel plug.

The quartz tube was calibrated by measuring with a cathetometer the distance between the top of a known weight of mercury and a machined tantalum ring which sat on top of the tube (as shown in Fig. A). The height of the mercury meniscus was also measured. Later, using the same tube and mercury from the same bottle, X-ray shadowgraphs were

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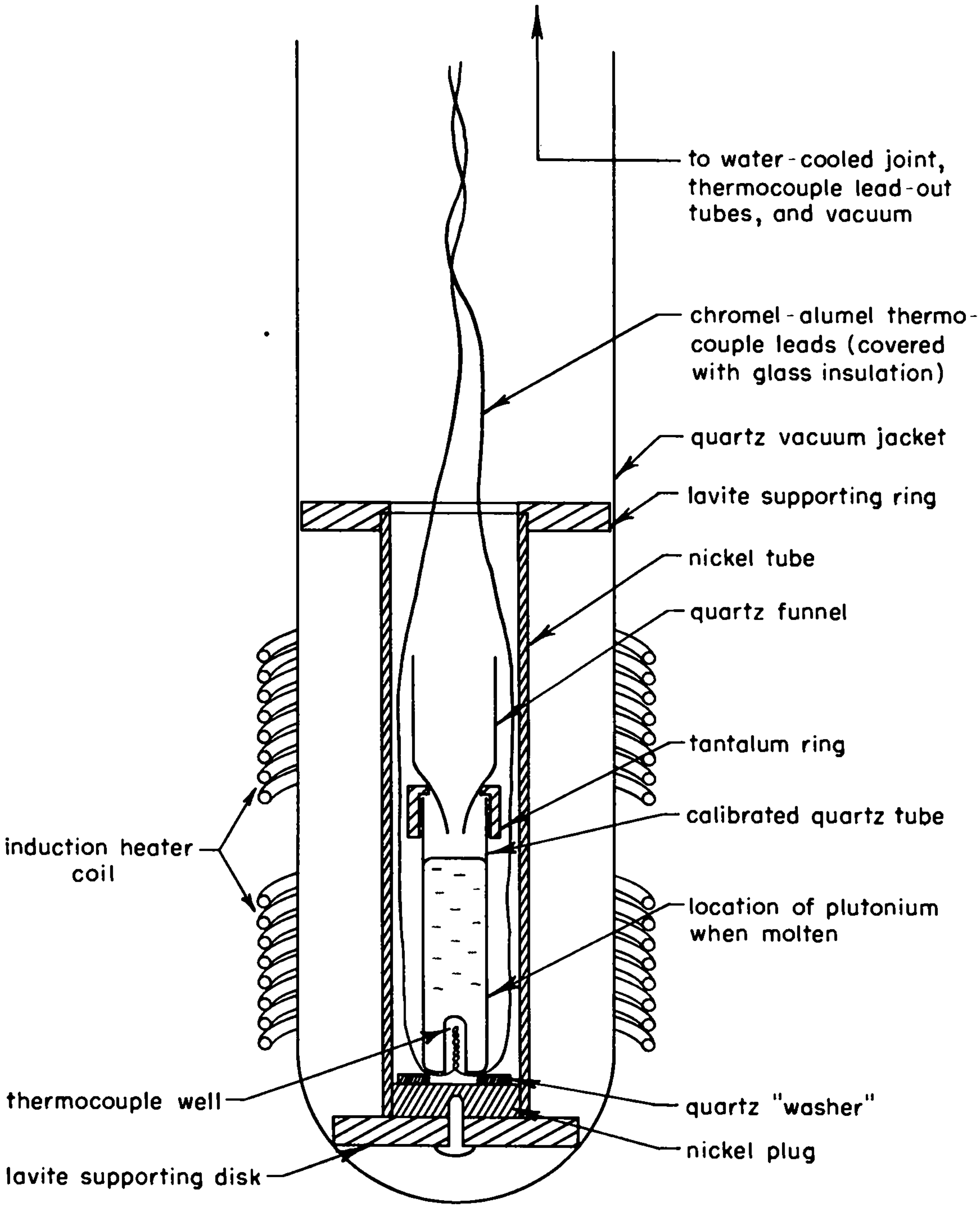

Fig. A

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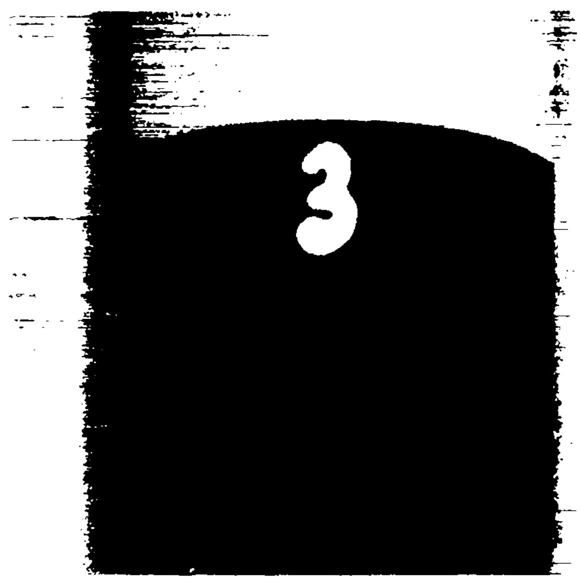

Figure B

taken of mercury menisci of various heights using an X-ray machine from GMX-1. Figure B is one of the photographs thus obtained. The meridian curve of each of these was traced onto graph paper. Measurements with a ruler were made on these curves. A table of the height y of the curve above the horizontal base line of the meniscus as a function of the distance x from the center of the meniscus was made up for each meniscus. $2\pi x y$ was calculated for each value of x. The units mm' and cm' used in this report are the direct measurements of the graph of the meridian curves in mm and cm; the unprimed coordinates are reserved for the dimensions of the actual meniscus. For each meniscus $2\pi x y$ has been plotted against x, and

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the area under the curve obtained; assuming that the meniscus is a surface of revolution, this area, after it has been scaled down from the size of the photograph to the size of the actual meniscus, gives the volume of the meniscus.

Using a bore diameter of .978 cm for the quartz tube, the scaling factor is calculated as follows. The bore diameter as it appears in the photograph is 10.4 cm'.

$$.978 \text{ cm} = 10.4 \text{ cm}'$$

$$1 \text{ cm}' = .978/10.4 = .940 \text{ mm}$$

$$1 (\text{cm}')^3 = .83 \text{ mm}^3$$

Applying this scaling factor, the volumes of the mercury menisci and their heights are:

	#3	#4	#5	#6
Volume	.040 cm^3	.053 cm^3	.061 cm^3	.069 cm^3
Height	.940 mm	1.22 mm	1.32 mm	1.55 mm

These are plotted and a solid line drawn through the points in Fig. C. The plot was used for the meniscus volume correction. The mercury calibration and the mercury calibration minus the meniscus volume (calibration of the cylindrical section of the tube) is shown in Table I. The corrected calibration is plotted against the distance between the edge of the meniscus and the tantalum ring in Fig. D. Only the calibration in the region actually occupied by the plutonium meniscus in the plutonium run is plotted.

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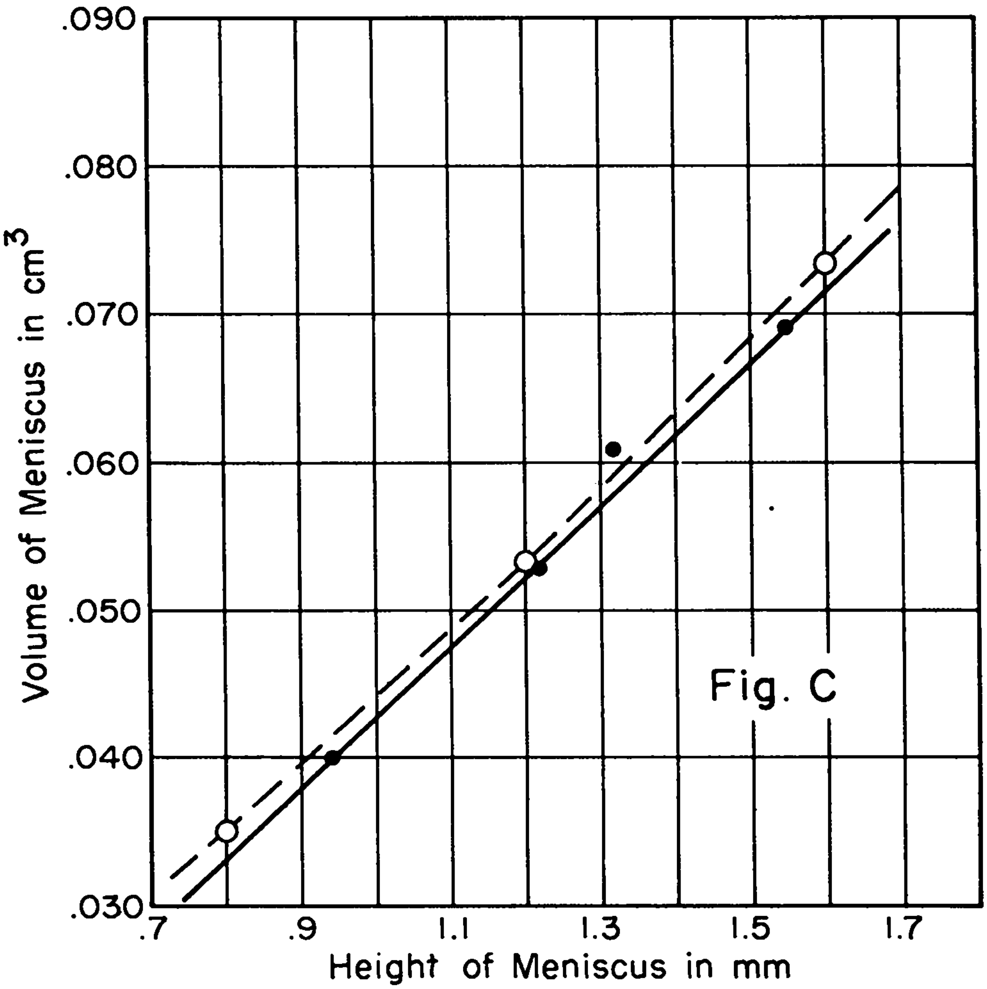

Fig. C

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Table I

Volume of Hg	Edge of meniscus to tantalum ring	Height of meniscus	Volume of meniscus from Fig. C	Volume of Hg minus volume of meniscus
3.914 cm^3	1.69 cm	1.3 mm	.057 cm^3	3.857 cm^3
4.114	1.42	1.4	.062	4.052
4.303	1.18	1.5	.066	4.237
4.494	.95	1.8	.080	4.414
4.661	.74	1.6	.071	4.590
4.796	.52	1.3	.057	4.739
4.948	.33	1.4	.062	4.886
5.105	.11	1.0	.043	5.062
3.995	1.56	1.1	.048	3.947
4.147	1.34	1.0	.043	4.104
4.441	.97	.8	.034	4.407
4.649	.74	1.1	.048	4.601
4.834	.51	1.2	.052	4.782
3.478	2.19	.75	.032	3.446

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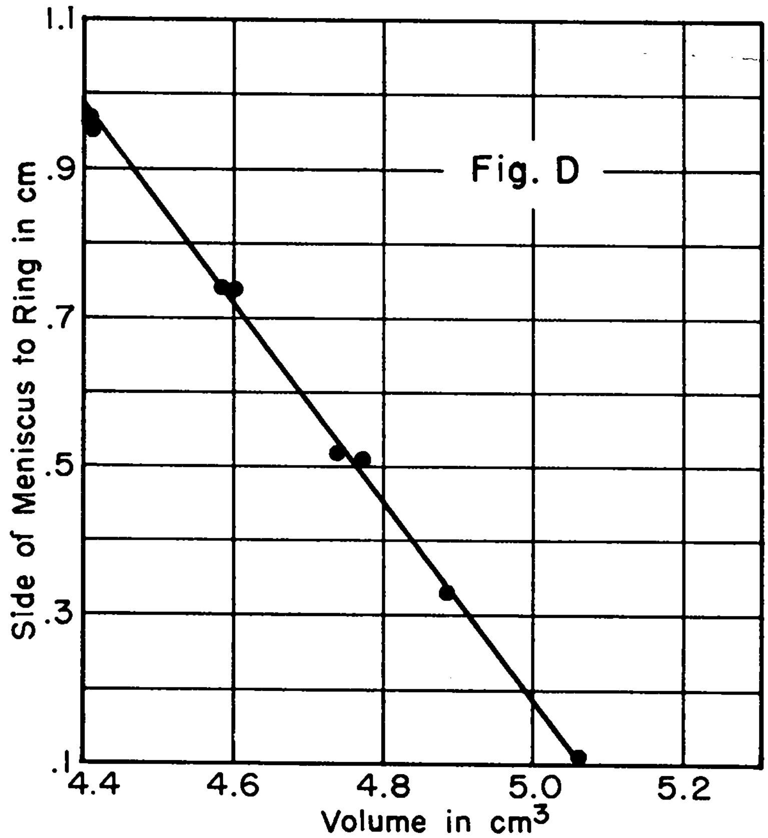

Fig. D

A straight line was drawn through the points of the calibration curve, because the quartz tube was unusually clear and uniform. No point is more than .4% of the total volume represented by that point from the straight line. The reciprocal of the slope of the line is the cross-sectional area of the tube. It corresponds to a diameter of .978 cm. The diameter of .978 cm, used in the above calibration, was obtained in this way by first calculating a calibration curve

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assuming a diameter of 1 cm. A check on this value will be mentioned later.

The measurement was carried out in the following manner. A sample of plutonium weighing 82.4423 gm was placed in the funnel, the apparatus assembled, and the system pumped to a good vacuum. The induction heater was turned on and the sample heated. The sample melted and flowed dropwise into the quartz tube. Thermocouple readings were taken regularly. The temperature was controlled as carefully as possible by turning the induction heater on and off. When the temperature was at a desired value, a film was placed in a holder several inches behind the sample. An X-ray machine several feet in front of the sample was turned on to give an exposure lasting 1-1/2 minutes. Four exposures in all were taken. A cathetometer reading was also taken, but the lighting of the meniscus was poor.

The next day when the apparatus was cool, the sample was removed. It was a very clean-looking sample. The quartz tube had been shattered, but the thermocouple well was still contained in the sample. This was almost completely removed by smashing it out with a punch. The weight of the sample containing the small amount of quartz which was left was 79.28 gm. Also, the metal and oxide held up in the funnel were removed and weighed. The weight was 3.18 gm. The weight of the original sample, 82.44 gm minus 3.18 gm, is

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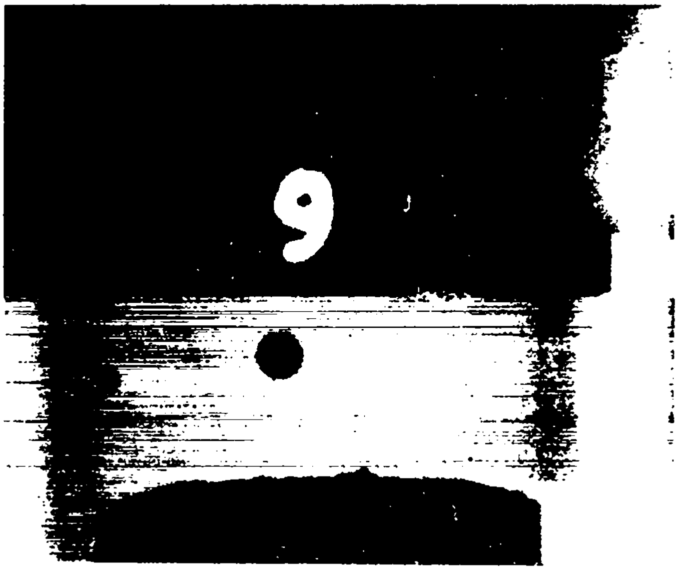

Figure E

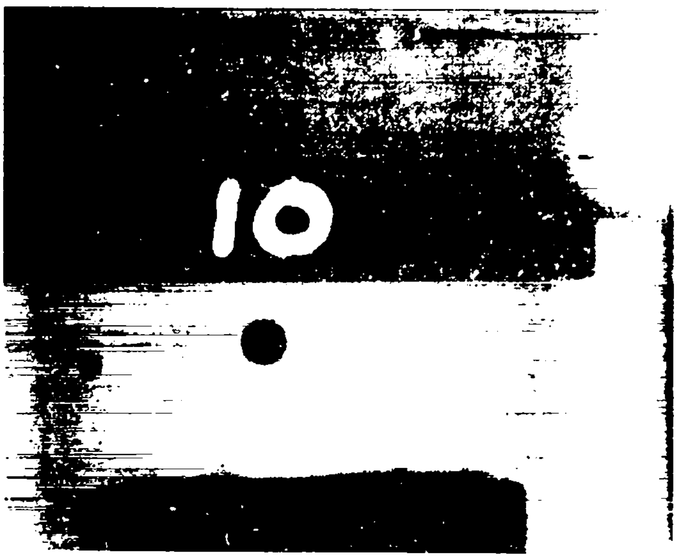

Figure F

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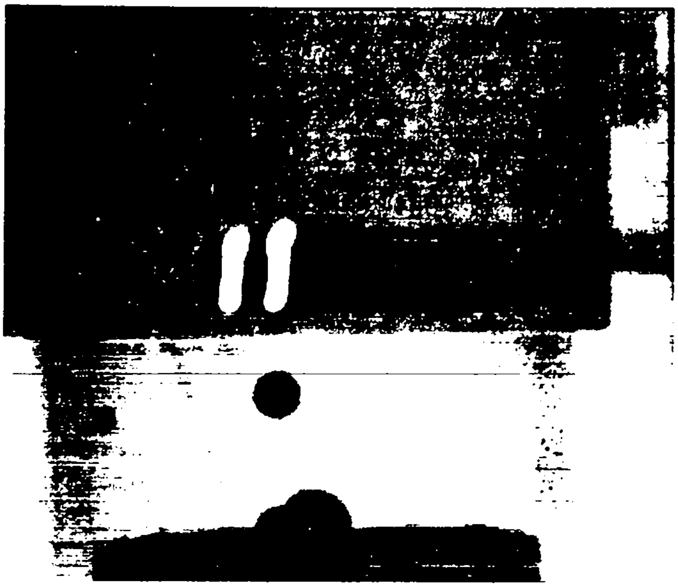

Figure G

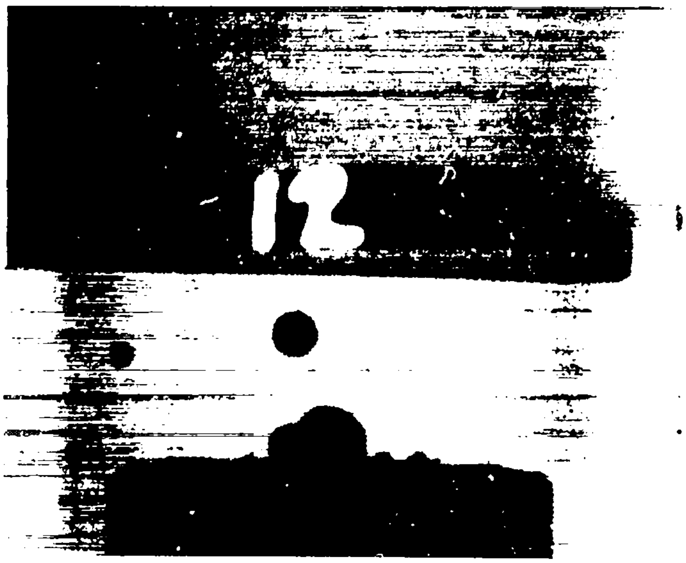

Figure H

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79.26 gm, and this is the weight used in making the density calculation.

The four shadowgraphs of the plutonium are shown in Figs. E, F, G, and H. None of the menisci is very smooth. Apparently, partial solidification had occurred in Figs. G and H. The temperature data for these four shadowgraphs are given in Table II.

Table II

	Highest temp. in millivolts	Highest temp. in °C	Lowest temp. in millivolts	Lowest temp. in °C	Difference between highest and lowest temp.
#9	31.63	760	30.32	728	32
#10	28.00	673	27.50	661	12
#11	27.13	652	26.50	638	14
#12	23.36	564	22.54	544	20

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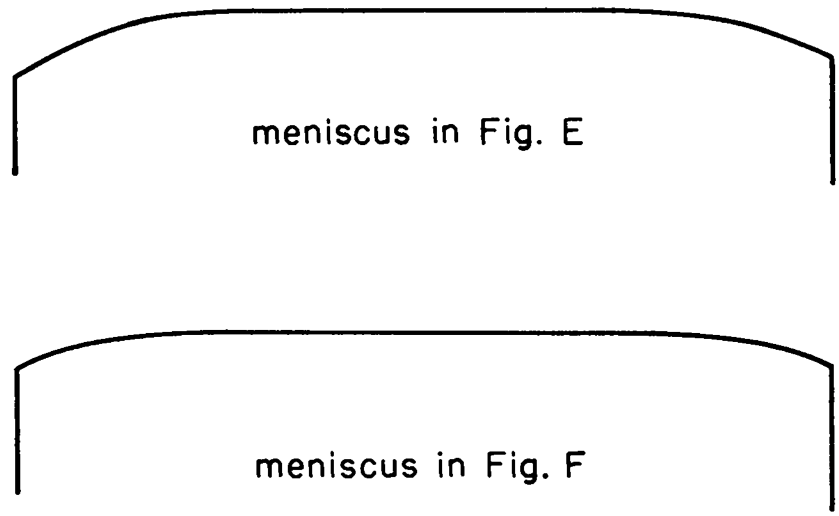

Fig. J

A correction was calculated for the menisci in Figs. E and F, as was done for the mercury. The meridian curves are seen on graph paper in Fig. J. The menisci are so flat that the volume of the central cylinder was calculated,and only the volume of an outer ring of each meniscus was obtained by graphical integration. The data are given in Tables III and IV.

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#9 Table III

x	0 to 2.5 cm'	2.5	3.0	3.5	4.0	4.5	5.0	5.5
y	.85 cm'	.85	.85	.80	.70	.50	.30	0
$2\pi xy$		13.3	16.1	17.6	17.6	14.1	9.4	0

Volume of central cylinder = $\pi r^2 h$ = 16.69 (cm')^3

#10 Table IV

x	0 to 2.5	2.5	3.0	3.5	4.0	4.5	5.0	5.45
y	.5	.5	.5	.45	4.0	3.0	.20	0
$2\pi xy$		7.8	9.4	9.9	10.0	8.5	6.3	0

Volume of central cylinder = 9.82 (cm')^3

The plots of $2\pi xy$ against x are given in Figs. K and L.

The photograph in Fig. F was used to calculate the density of plutonium. A step cut in the tantalum ring (on the right in the photograph) of depth .462 cm was used to calculate the magnification factor. In the photograph its depth is 5.15 cm'. So 1 cm' = .0897 cm. The bore of the tube in the photograph is 10.9 cm'. By use of the magnification factor, the bore of the tube is .975 cm, agreeing well with the .978 cm obtained above. In a similar manner the edge

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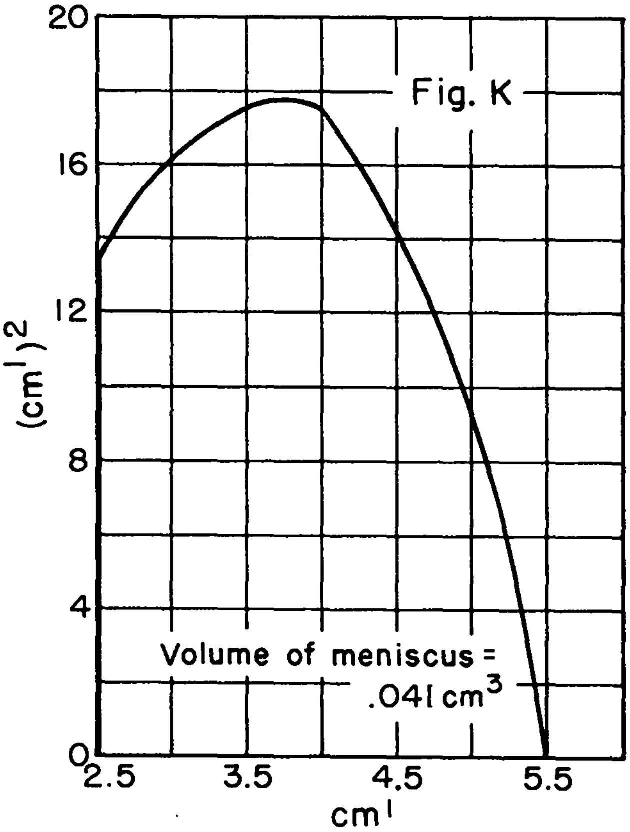

of the meniscus is found to be 4.75 cm below the tantalum ring. The tantalum ring is 2.03 cm long. The linear coefficient of expansion of tantalum is 6.5×10^{-6}. It will expand downward and, assuming the temperature increase to be 700°, the corrected distance between the edge of the plutonium and the tantalum ring is .475 + .009 = .484 cm. This neglects the small expansion of quartz upward. Using the calibration curve and adding the volume of the meniscus, the volume of

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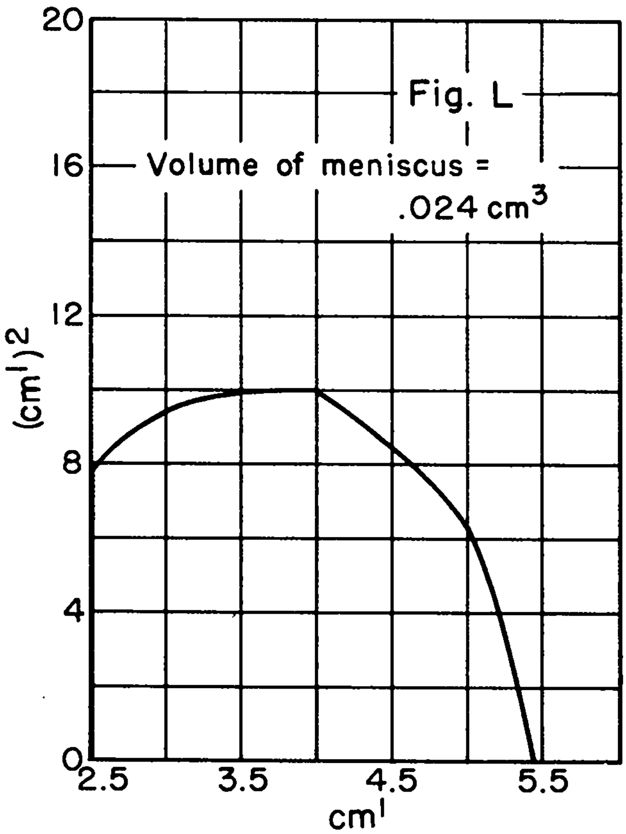

Fig. L

the plutonium in Fig. F is 4.780 + .024 = 4.804. The density is 79.26/4.804 = 16.50 gm/cm^3, at about 665° C.

Careful examination shows that the cooling in going from Fig. E to Fig. F causes a drop in the meniscus without any slipping at the edges. The volume change is therefore .041 - .024 = .017 cm^3. This, divided by the total volume, is .017/4.780 = 3.6 x 10^{-3}. Taking the two extremes of temperature change, 760 - 661 = 99° and 728 - 673 =

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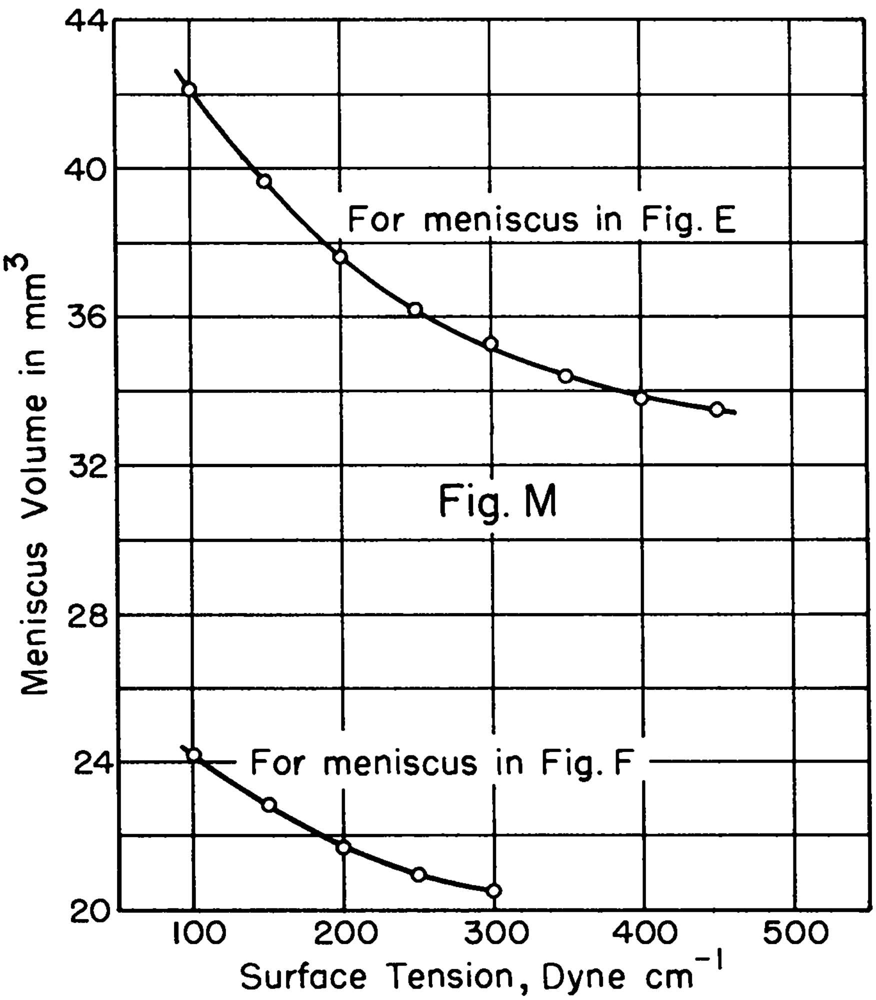

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55°, the volume coefficient of thermal expansion appears to lie between 3.6×10^{-5} and 7.2×10^{-5} at a mean temperature of about 700° C. Therefore, in Fig. F, where the uncertainty in the temperature is perhaps 12° to 20°, the uncertainty in the density is of the order of $7.2 \times 20 \times 10^{-5} = 144 \times 10^{-5}$, which is less than 0.15%.

All sources of error appear to be well within 0.5% of the value given for the density. An earlier rough measurement of the density in which no shadowgraphs were taken or meniscus corrections applied yielded a value of 16.54 gm/cm^3. The agreement with 16.50 is probably partly accidental.

An attempt has been made to find the value of the surface tension of the liquid plutonium observed in this experiment. Using the tables of B. E. Blaisdell,[1] the volume of the plutonium menisci for the tube bore used in this experiment and for a fixed height of meniscus was calculated as a function of surface tension. This calculation was carried out twice, once using as a value for the fixed height the height observed in Fig. E, and a second time using the height in Fig. F. These two functions are plotted in Fig. M. The value of 16.5 gm/cm^3 for the density was used in the calculation.

Using the volume found empirically for each of these plutonium menisci, the surface tension which would give this volume was read from the graph in Fig. M. The two values obtained are 105 dynes/

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cm and 120 dynes/cm, in the region around 700° C.

It can be noticed from the graphs that the observed meniscus volumes occur in a region in which the volume is reasonably sensitive to the surface tension.

It is difficult to evaluate the significance which should be attached to the value of the surface tension obtained for the plutonium in this experiment. It would appear that the method of obtaining the surface tension is satisfactory. However, there is no way of knowing what impurities may have been picked up from the quartz at the temperature of the experiment, what their concentration may have been, or how large their concentration would have to be to lower the surface tension drastically. As Burdon [2] points out, the effect of a small concentration of an impurity depends entirely on the extent to which it lowers the surface free energy and is thereby concentrated in the surface layer.

Very little, if any, visible reaction with the quartz occurred. After the experiment the sample was clean. From the photographs of the plutonium it appears that some solid particles, such as an oxide, were floating on the meniscus. However, it is felt that the surface tension which determined the shape of the observed meniscus was of the order of 100 dynes/cm.

Additional use was made of Blaisdell's tables to calculate the volume of mercury menisci as a function of height, using the bore of the tube used in this experiment. The results are plotted as a

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dashed curve in Fig. C. There is good agreement with the empirical curve. A surface tension of 395 dynes/cm was assumed for the mercury in order to avoid being outside the limits of the table.

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1. Blaisdell, B. E., Journal of Mathematics and Physics, XIX, 217-27 (1940).

2. Burdon, R. S., "Surface Tension and the Spreading of Liquids," Cambridge University Press (1940).

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