APPROVED FOR PUBLIC RELEASE

SIFIED

LA-1656

Copy [illegible] of 109

Series A

[redacted]

UNCLASSIFIED

LOS ALAMOS SCIENTIFIC LABORATORY
of the
UNIVERSITY OF CALIFORNIA

PUBLICLY RELEASABLE
LANL Classification Group
Mark M Jones
10/27/95

Report written:
April 7, 1954

Classification changed to UNCLASSIFIED
by authority of the U. S. Atomic Energy Commission,
Per ALDR(TID-1400-S2)Sept-Oct 1974
By REPORT LIBRARY Pete Martinez, 3/19/75

LA-1656 This documents consists of 20 pages,

[redacted]

HANFORD
55851

THE DENSITY OF ALPHA-PHASE PLUTONIUM

Classification changed to ~~CONFIDENTIAL~~
by authority of the U. S. Atomic Energy ~~Commission.~~
Per ~~ACRC (TID-1100) 9-~~55
By REPORT LIBRARY P. Martinez 12-2-57

Work done by:
Henry L. Laquer

Report written by:
Henry L. Laquer

SPECIAL REREVIEW FINAL DETERMINATION	Reviewers	Class.	Date
Class: U	WHL	U	4/6/82
	SAU	U	..

CHEMISTRY-TRANSURANIC ELEMENTS

VERIFIED UNCLASSIFIED
Mark M Jones
10/27/95

UNCLASSIFIED

[redacted]

[redacted]

[redacted]

UNCLASSIFIED

[redacted] [redacted]

UNCLASSIFIED

CHEMISTRY-TRANSURANIC ELEMENTS

Distributed: JUL 28 1954

Distributed: JUL 28 1954	LA-1656
Los Alamos Report Library	1-20
American Machine and Foundry Company	21
Argonne National Laboratory	22-29
Armed Forces Special Weapons Project, Washington	30
Army Chemical Center	31
Atomic Energy Commission, Washington	32-33
Babcock and Wilcox Company	34
Brookhaven National Laboratory	35-36
Carbide and Carbon Chemicals Company (K-25 Plant)	37-38
Carbide and Carbon Chemicals Company (ORNL)	39-42
Carbide and Carbon Chemicals Company (Y-12 Plant)	43
Chicago Patent Group	44
Commonwealth Edison Company	45
Detroit Edison Company	46
Dow Chemical Company (Rocky Flats)	47
duPont Company, Augusta	48-51
Duquesne Light Company	52
Foster Wheeler Corporation	53
General Electric Company (ANPD)	54
General Electric Company (APS)	55
General Electric Company, Richland	56-59
Goodyear Atomic Corporation	60-61
Hanford Operations Office	62
Iowa State College	63
Knolls Atomic Power Laboratory	64-65
Massachusetts Institute of Technology (Benedict)	66
Monsanto Chemical Company	67
Mound Laboratory	68-70
National Lead Company of Ohio	71
Naval Research Laboratory	72
Newport News Shipbuilding and Dry Dock Company	73
North American Aviation, Inc.	74-75
Patent Branch, Washington	76
Phillips Petroleum Company (NRTS)	77-78
RAND Corporation	79
U. S. Naval Radiological Defense Laboratory	80
UCLA Medical Research Laboratory	81
University of California Radiation Laboratory, Berkeley	82-85
University of California Radiation Laboratory, Livermore	86-87
University of Rochester	88-89
Vitro Corporation of America	90-91
Westinghouse Electric Corporation	92-93
Pacific Northwest Power Group	94
Technical Information Service, Oak Ridge	95-109

UNCLASSIFIED

[redacted] [redacted]

[redacted]

[redacted]

ABSTRACT

The density of a sample of pressed high-purity 600 MWD α-phase plutonium has been determined as 19.737 ± 0.005 g/cm^3.

An attempt has been made to evaluate and, if possible, to eliminate all conceivable errors which might affect density determinations.

[redacted]

[redacted]

[redacted]

CONTENTS

ILLUSTRATIONS

[redacted]

[redacted]

1. Introduction

The density of polycrystalline metals is rarely reported with an accuracy of better than 0.1% (100 parts in 10^5 *). The reason for this limitation is a practical one, because differences in mechanical and metallurgical treatment as well as small amounts of impurities can vary the density of most metals by more than 0.1%. In a severely cold-worked metal, there may be as many as 10^{12} dislocation lines per square centimeter. Assuming a lattice distance of 2.5 Å and that a dislocation produces half a vacancy, one calculates a density decrease of about 30 parts in 100,000. On the other hand, in a well annealed metal, dislocation densities may be as small as 10^8 lines/cm^2, a factor of 10,000 less. When an ideal or "theoretical" density value is needed, recourse may be had to the values obtained on single crystal specimens or from precision X-ray measurements.

The density of α-phase plutonium is usually reported with a probable error of ± 0.05 g/cm^3 (250 parts in 10^5), although measurements obtained on any one experimental set-up within one to six months usually exhibit a much better internal consistency or precision of as good as ± 0.01 g/cm^3. We also know that the density of α-phase plutonium is greatly affected by the presence of impurities which probably stabilize small amounts of the β-phase, so that the density of apparently sound castings has been found to vary from 19.3 to 19.6 g/cm^3. It is most probable that α-phase plutonium is in a highly cold-worked condition, i.e., contains a great number of dislocations, first because it has undergone the $\beta \rightarrow \alpha$ transition and is not capable of self-annealing at the low temperatures at which the α-phase is stable, and secondly from the continuous radiation damage of the 5 Mev alpha particles. In pure Pu^{239} with a half life of 24,400 years, the alpha radiation amounts to 1.15×10^{17} disintegrations/month/cm^3. In 600 MWD material which contains about 4.5% of Pu^{240} (with a half life of 6150 years) and 8×10^{-3}% of Pu^{238} (with a half life of 89 years), the alpha activity is increased by about 15%. Whether or not these radiation damage effects are really worth considering depends on the number and length of dislocations produced by each disintegration, a question which we cannot answer at present. The simple mass effect of a 4.5% Pu^{240} concentration amounts to a density increase of 19 parts in 100,000 and might well compensate for the decrease in density due to the additional radiation damage.

At any rate, since we do not know the crystal structure of α-phase plutonium, and since there is little hope of obtaining single crystals, a precision determination on a sample of the highest known density would seem to be the only way of establishing a lower limit to the "theoretical" density of the metal.

---

*To simplify comparisons, all errors and uncertainties will be expressed as parts in 100,000.

UNCLASSIFIED

[redacted]

2. Materials

Two cylinders approximately 1" long and of 3/4" diameter were vacuum-cast in lime crucibles on September 28 and 29, 1953, by CMR-5 from very high purity buttons prepared by CMR-11. They were then pressed around October 14, 1953, also by CMR-5, at 325°C and 50,000 psi, and cooled to 50°C in about four hours with the load maintained. Subsequently, the pressed cylinders 2020 and 2021 were machined slightly by CMR-11 and some observations were made on them. However, for our final study they were re-machined more extensively by CMR-11 and labeled 2020-A and 2021-A.

Table I gives the chemical analyses of the original buttons, M V and M VI, and of the specimens. Two analysis samples were submitted for each specimen. The first, 2020-X and 2021-X, were taken from the top of the castings before pressing. Their oxygen and calcium values are probably too high. The second, 2020-Y and 2021-Y, were fine turnings taken from the end and cylindrical surfaces when the specimens were re-machined into 2020-A and 2021-A. Unfortunately, these fine turnings had corroded badly by the time they were analyzed so that their oxygen values are also too high. The high calcium results on 2021-Y are completely inexplicable. A sampling of the core of the specimen would be desirable. However, we are not ready to do this at the present time. Nevertheless, it is probable that the specimens, or at least specimen 2020, contain less than 300 ppm of impurities by weight, which corresponds to an impurity concentration of 0.2 to 0.3 atomic percent.

Our reason for choosing these specific samples for a precision density study was that preliminary "regular" density determination by both CMR-5 and CMR-11 indicated the highest densities which we had so far observed on any sample of α-phase plutonium.

3. Method and Limitations

The density, or mass per unit volume, for any material requires a knowledge of the weight, or more accurately, the mass in vacuo, and of the volume of the sample. The weight can be determined, of course, only by direct weighing, but the volume can be found in two different ways:

a. From the dimensions, if the sample is of simple shape.

b. Through Archimedes' principle, i.e., through the weight loss in a fluid of known density.

3.1 Weighing

Any good analytical balance together with a set of calibrated weights should allow mass determinations to ±0.1 mg. Even under the most adverse conditions which might prevail in a

[redacted]

TABLE I

CHEMICAL AND SPECTROCHEMICAL ANALYSES (ppm)

	M V	2020-X	2020-Y	M VI	2021-X	2021-Y
C	30-60	25-25	45-50	15-30	55-55	75-85
O	---	720-1040*	400-460**	---	270-410*	520-620**
Fe	80	30	150	70-80	60	160
F	---	4	---	---	6	---
S	<10	7	---	<10	<5	---
Li	< 0.2	< 0.2	---	< 0.2	< 0.2	---
Be	< 0.2	< 0.2	---	< 0.2	< 0.2	---
B	< 0.5	0.5	---	< 0.5	0.5	---
Na	< 5	< 5	---	5	<5	---
Mg	< 3	< 3	---	3	<3	---
Al	5	< 5	5	<5	5	10
Si	20	8	5	12	5	5
Ca	20	350*	30	15	50*	1000-1600***
V	<20	<20	---	<20	<20	---
Cr	< 5	20	<5	<5	<5	<5
Mn	30	30	35	30	25	30
Co	< 10	<5	---	<5	<5	---
Ni	2	10	<1	<1	1	<1
Cu	8	10	10	6	3	5
Zn	< 10	< 20	---	<20	<20	---
As	---	< 20	---	<20	<20	---
Mo	<20	< 20	---	<50	<20	---
Ag	< 1	< 1	---	<1	<1	---
Cd	< 1	< 1	---	5	<1	---
In	< 1	< 1	---	<1	<1	---
Sn	< 1	< 1	---	<1	<1	---
Sb	---	<10	---	<1	<10	---
Ba	---	<10	---	---	<10	---
La	<10	<10	---	<10	<10	---
Hg	< 3	< 3	---	---	< 3	---
Pb	1	3	---	<1	<1	---
Bi	< 1	< 1	---	<1	< 1	---

*Possibly due to poor sampling.

**Fine turnings badly oxidized.

***Inexplicable, re-checked four times.

[redacted]

[redacted]

dry box, accuracies of 1 part in 100,000 should be attainable for samples weighing 10 g or more.

The correction due to the difference in buoyancy of air on the brass or bronze weights (ρ = 8.4) and on a sample of density 19.5 to 19.9 amounts to -5.7×10^{-5} (at Los Alamos).

Another correction which is sometimes overlooked is that caused by the inequality of the balance arms. It is generally negligible, but amounted to 0.82 parts in 100,000 on the dry box balance which was used in this work.

3.2 Determination of Dimensions

With a calibrated micrometer, dimensions can be determined readily to ±0.000 2". Thus, even if our plutonium cylinders are well machined, we can expect to know their volumes, and hence densities, only with a probable error of ±57 parts in 100,000.

On the other hand, for non-radioactive materials which can be finished to very close tolerances, it is not at all impossible to have meaningful dimensions reported with an accuracy of ±0.000 02" (20 microinches) which then reduces the probable error of the volume determination to ±6 parts in 100,000 (for 1" long, 3/4" diameter cylinders).

3.3 Choice of the Immersion Fluid

The fluid chosen for immersion weighing should be of known and stable density. It should also have low vapor pressure, low viscosity, and high density. It should be non-corrosive to the metal under investigation and it should wet the metal readily. Distilled water would be a very suitable fluid except for its corrosive action on some metals.

The material which has been used in this laboratory as far back as 1944 is bromobenzene. It is stable chemically, does not corrode most metals, is non-hygroscopic, is readily available and purifiable, and does not evaporate too quickly but does have a sufficient vapor pressure so that it is easily possible to clean and dry specimens after immersion.

[There is a variation of the immersion method in which the flotation temperature of the solid in a liquid is found [cf. D. A. Hutchison, Phys. Rev. 62, 32 (1942); 66, 144 (1944)] . This method is amenable to extreme accuracy (1 to 2 parts in 100,000) but unfortunately cannot be used with heavy metals, since it requires an exact matching of fluid and solid densities.]

3.4 Density of Bromobenzene

As already mentioned, bromobenzene of reasonable purity is readily available. The density of this liquid when kept in a screwcap container decreases 3 to 5 parts in 100,000 per month, which is only slightly larger than the reproducibility or precision of our density calibrations (cf. subsection 3.4.4).

[redacted]

UNCLASSIFIED

The actual value of the density of bromobenzene can be obtained from the literature, by Westphal balance, by pyknometer, or by calibration with a solid specimen of known density.

3.4.1 Literature Values. The best values obtained on carefully distilled material are those reported by J. Timmermans and F. Martin [J. Chim. Phys. 23, 747-87 (1926)] . Their reported values are:

$$d_4^0 = 1.522\ 04 \text{ g/ml, from which } \rho^0 = 1.522\ 00 \text{ g/cm}^3$$

$$d_4^{15} = 1.501\ 70 \text{ g/ml, from which } \rho^{15} = 1.501\ 66 \text{ g/cm}^3$$

$$d_4^{30} = 1.481\ 50 \text{ g/ml, from which } \rho^{30} = 1.481\ 46 \text{ g/cm}^3$$

They also report these other physical properties:

$$n_D^{15} = 1.562\ 52$$

$$\text{BP} = 156.15^\circ\text{C}$$

$$\text{MP} = -30.6^\circ\text{C}$$

$$\eta^{15} = 1196 \times 10^{-5} \text{ cgs}$$

$$\eta^{30} = 985 \times 10^{-5} \text{ cgs}$$

The International Critical Tables (III-29) give an equation for the range of 0-80°C:

$$d_t = 1.522\ 31 - 1.345 \times 10^{-3} t - 0.24 \times 10^{-6} t^2 + 0.76 \times 10^{-9} t^3$$
$$\pm\ 0.000\ 5\ (\pm\ 0.000\ 2 \text{ for } 0\text{-}40^\circ\text{C})$$

Values from this equation are higher than Timmermans' by about 21 parts in 100,000 (at 23°C).

The International Critical Tables (III-33) also list a 0°C value

$$d_4^0 = 1.521\ 93 \pm 0.000\ 10 \text{ g/ml}$$

which is 26 parts in 100,000 lower than the ICT equation value and 7 parts in 100,000 lower than Timmermans' value.

The Chemical Rubber Company Handbook value of

$$s_{15}^{15} = 1.499\ 1 \text{ leads to a value of}$$

$$\rho^{15} = 1.497\ 7_5 \text{ g/cm}^3$$

[redacted]

which is lower than Timmermans' value by 262 parts in 100,000, but may be correct for rather crude commercial material of MP 155 to 156°C. (Cf. subsection 3.4.3.)

The temperature coefficient of the density near room temperature is, using Timmermans' data:

$$\Delta\rho/\Delta t = -0.001\ 34_7\ \mathrm{g/cm^3/^\circ C}$$

$$(1/\rho)\ (d\rho/dt) = -90._4 \times 10^{-5}/^\circ\mathrm{C}$$

The ICT equation yields a temperature coefficient of greater absolute value (more negative) by 0.6 ± 0.4%.

3.4.2 Westphal Balance. The Westphal balance gives quick results but is not readily calibrated. Density values can be read to 0.000 1 g/cm^3. However, with an uncalibrated balance, density values were obtained (1950) which were too high by 300 parts in 100,000, although they were self-consistent to within 20 parts in 100,000.

3.4.3 Pyknometer. The pyknometer involves the accurate weighing of a known volume of liquid. The instrument is usually calibrated with water or mercury. Its main limitations lie in the accurate filling and level and meniscus corrections which with a 20 cm^3 volume may well amount to 0.001 cm^3 or 5 parts in 100,000.

Our own pyknometer experience (1950) with bromobenzene redistilled above zinc gave a density 7.4 parts in 100,000 below Timmermans' data. At the same time measurements on "as received" bromobenzene showed that it had a density 156 parts in 100,000 below Timmermans' value.

3.4.4 Calibration with a Solid Specimen of Known Density. Calibration with a solid of known density, determined from dimensions, is probably the best method, particularly since this can eliminate or at least reduce many of the immersion weighing corrections discussed below.

In our recent work we have calibrated a batch of Eastman "Practical" bromobenzene as taken from the bottle. Using four accurately machined and measured wolfram specimens whose calculated densities covered a range of 140 parts in 100,000 with a mean deviation of ±45 parts in 100,000, density values were obtained which, on different days, were consistent to within ±2 parts in 100,000 and at the same time were only 13 parts in 100,000 below Timmermans' data. This seems to indicate that the presently available Eastman "Practical" bromobenzene is of very high quality and that there is little justification for any attempt to further purify the material.

UNCLASSIFIED

[redacted]

[redacted]

A similar check on bromobenzene from a different bottle from the same manufacturer using only two of the wolfram standards gave a value 30 ± 3 parts below Timmermans' data. Material from this bottle had been used by CMR-5 for several years and was also used in our dry box work.

3.5 Immersion Weighing

In spite of the simplicity of Archimedes' principle, which states that the loss in weight is equal to the density of the liquid times the displaced volume, there are a number of factors which are often overlooked. Thus

a. The level of liquid is raised by the sample, lowering the weight of the immersed suspension. With bromobenzene, a 7 cm^3 specimen, a 2" diameter jar, and a B&S 22 ga (0.0253" diameter) suspension wire this correction amounts to 1.5 mg (being more with a narrower jar or thicker wire), or 15 parts in 100, 000, which fraction is independent of the specimen volume for any given set-up.

b. The loss of bromobenzene through spillage, adherence, or evaporation is not entirely negligible either, amounting to a buoyancy difference on the suspension wire of 0.5 mg/mm change in level (for B&S 22 ga wire). However, it can be eliminated by weighing the empty suspension sufficiently frequently.

c. The air buoyancy correction to be applied to the immersed weight before calculating the loss in weight is $-10.3 \pm 0.3 \times 10^{-5}$ (at Los Alamos) and is governed only by the relative density of the air and of the brass weights.

d. The surface condition of the sample may retard wetting by the fluid. This can lead to an apparent slow increase in the immersed weight of 3 to 5 mg. With our wolfram density standards, we found that degreasing with trichloroethylene in a Soxleth extractor reduced the drift time from 30 minutes to less than 5 minutes and the magnitude of the change to less than 1 mg, which then may well have been due to temperature equalization effects alone. In conjunction with this, it appears obvious that enough time should be allowed for the immersed weight to become constant.

e. The temperature coefficient of the density of most liquids is large. For bromobenzene it is $-90._4 \times 10^{-5}/^{\circ}C$. Thus a temperature uncertainty of only ±0.05°C will cause an error of ±5 parts in 100, 000. Temperature gradients in the liquid caused by heat generation in the sample are also undesirable. The correct fluid temperature to be used is the one measured at the level of the sample. The additional weighing error caused by convection currents is discussed separately in section 3.6.

f. The thermal expansion of the metal under investigation should also be taken into account and densities should be reported as at a given temperature. This effect is equal to

[redacted]

[redacted]

$-3\alpha/^{\circ}C$ where α is the coefficient of linear expansion. Thus for wolfram, $\alpha = 4.3 \times 10^{-6}/^{\circ}C$ and $(1/\rho)\ (d\rho/dt) = -1.29 \times 10^{-5}/^{\circ}C$, whereas for plutonium at 25°C, $\alpha = 50.8 \times 10^{-6}/^{\circ}C$, and $(1/\rho)\ (d\rho/dt) = -15.2_4 \times 10^{-5}/^{\circ}C$.

The above figures also indicate the probable errors caused by uncertainties in the sample temperature. In the case of plutonium with its large radioactive heat production, the uncertainty may well be ±0.3°C, which would lead to an error of ±5 parts in 100,000.

3.6 Convection Currents

One hundred grams of plutonium generate about 0.19 watt or more, depending on the isotopic composition of the material. Most of this heat is given off to the immersion fluid causing convection currents, the remainder being radiated. Eventually a steady state is reached and the fluid will assume a temperature above that of its surroundings. It appears almost impossible to calculate from first principles the magnitude of the effect of these convection currents upon the immersed weight of the sample. However, after some experimentation we succeeded in observing the effect with a dummy sample of the same size and shape as our plutonium specimens and containing a 10 ohm resistor. Contacts were made through a Kovar type fused glass terminal, and with mercury on the inside of the copper capsule. Leads of very fine coiled Litz-wire were taken over two knife edges and to a storage battery. The wires did not prevent the balance from swinging freely. They had a resistance of about 6 ohms. The current was measured with a Simpson meter. Reversal of the polarity and a measurement in air indicated that there were no effects due to currents induced in the balance itself. The temperature of the immersion fluid at the height of the specimen was measured to ±1/80°C with a copper-constantan thermocouple and a self-balancing indicating potentiometer which could be read to ±0.5 μV. Figure 1 gives the apparent immersed weight and the microvoltage vs time curves for two different power levels. Table II lists the power used in the various experiments and the extrapolated offsets observed when the power was turned on and off. In general, the offsets appear larger for the "on" case. However, this may be due to the fact that a straight line extrapolation is not correct for the "off" case. The same data are plotted in Fig. 2.

The heat generated by specimen 2020-A was determined as $0.2972_5 \pm 0.0005$ watt by CMR-3, using a twin calorimeter. This value is 14.4% higher than what one would obtain with the same mass of pure Pu^{239}, but it is quite reasonable for 600 MWD plutonium. From Fig. 2 we then obtain a correction of $+5._5 \pm 1$ mg for the immersed weight, or of $-5._5 \pm 1$ mg for the weight loss. It is apparent that this correction is by no means negligible. It should also be noted that the curves of Fig. 2 may not be of general validity since we did not study the effects of different sample shape, size of jar, and depth of immersion.

UNCLASSIFIED

[redacted]

[redacted]

TABLE II

THE EFFECT OF CONVECTION CURRENTS (mg)

Power (Watts)	"On"	"Off"
0.137	-2.7 ± 0.3	+2.9 ± 0.3
0.137	-3.2 ± 0.3	+2.6 ± 0.2
0.043_2	-1.2 ± 0.1	+1.1 ± 0.2
0.067_2	-1.7 ± 0.1	+1.5 ± 0.2
0.567	-9.5 ± 1.0	+7.0 ± 0.5

3.7 Summation of Errors

An attempt has been made in the present work to eliminate and evaluate all possible errors. An alternative approach, which may work almost as well, is to calibrate the complete experimental set-up with a density standard of approximately the same size, shape, and density as the unknown. This will lead to a fictitious density for the fluid, but most of the above-mentioned errors and corrections except those caused by the convection currents will cancel. Whatever method is used, there may be some residual random, as well as systematic, errors. Table III summarizes the probable errors under ideal conditions as well as under the actual dry box conditions under which our work was done. The individual fractional errors which accrue at every experimental step are compounded as the square root of the sum of the squares. It is interesting to note that our actual errors are about twice what might be expected under ideal conditions but using the same equipment.

TABLE III

SUMMATION OF ERRORS (parts in 100,000)

	Ideal Conditions	Actual Conditions
Weight of Specimen in Vacuo	1	1.5 to 4.5
Temperature of Specimen	5	5
Density of Fluid	7	12.4
Temperature of Fluid	5	9 to 18 (45)
Weight Loss on Immersion	3	5 to 10
Convection Currents	5	10
Total Probable Error	11.5	20 to 27 (50)

[redacted]

[redacted]

It is evident that the prime sources of error are the temperature uncertainties in the calibration of the fluid and in the immersion weighing of the unknown radioactive material as well as our still limited knowledge of the convection currents. Any improvement in this work will require more accurate measurement and control of the fluid temperature, preferably to an accuracy or at least a precision of $\pm 0.01^{\circ}C$.

4. Results and Conclusions

Table IV gives the results of the weighings obtained with a dry-box set-up on the wolfram calibration pieces as well as on the two plutonium specimens. This work was done on March 2, 1954, four and a half months after the samples had last been at elevated temperatures. Corrections have been applied for the weight calibration, the inequality of the balance arms, the weight of the spot-welded Nichrome V suspension ($1.299_3 \pm 0.000_1$ g in air, $1.145_0 \pm 0.000_2$ g in bromobenzene at $24.8^{\circ}C$ and at the raised liquid level), and the effect of the convection currents. As already mentioned, the uncertainty in the temperature measurement is the primary limitation of accuracy. The best weighted average value for the density of specimen 2020-A at $25^{\circ}C$ would seem to be 19.737 ± 0.005 g/cm^3. On the other hand, specimen 2021-A, which was prepared in an identical manner, has a density of only 19.717 ± 0.005 g/cm^3.

Table V and the bar graph, Fig. 3, compare our results on the two specimens with the work done by CMR-5 and CMR-11, as well as with densities calculated from dimensions. The $(\Delta\rho/\rho)$ column states the fraction by which the densities of the bromobenzene used in the various determinations deviate from Timmermans' value. There is general agreement within the experimental errors, except that densities determined from dimensions (squares) appear to be systematically and consistently below the immersion weighing results (circles) and by an amount which for the most precise determinations is just outside the sum of the probable errors. This is very unfortunate since it makes us suspect an unevaluated systematic error, or we would have to assume that the temperature of the samples when their dimensions were taken was at least 5 degrees higher than our concededly rough estimate in order to have the probable errors overlap. At any rate, the accuracy of the density determinations on individual samples exceeds the reproducibility presently attainable in their preparation.

Appendix

SUGGESTIONS FOR FURTHER WORK

In order to remove all ambiguities the following additional work would seem desirable:

a. Calibrate the bromobenzene with an accurately machined gold or platinum piece in addition to the previously used wolfram standards, which had been made by powder metallurgical

[redacted]

TABLE IV

RESULTS

Specimen	Weight in Air (corr.) W_1' (g)	Weight in Vacuo W_1'' (g)	Temperature t (°C)	Weight in Bromobenzene (corr.) W_2' (g)	Weight in Bromobenzene (corr. to vac.) W_2'' (g)	$W_1'' - W_2''$	ρ^t_{Std} (g/cm^3)	ρ^t_{Liq} (g/cm^3)	$\rho^{25.0}_{Liq}$ (g/cm^3)	ρ^t_{Pu} (g/cm^3)	ρ^{25}_{Pu} (g/cm^3)
W-2 Standard	$135.125_3 \pm .000_2$	135.117_9	$24.9 \pm .1$	$124.369_7 \pm .000_6$	124.356_9	10.761_0	$18.680_7 \pm .001$	$1.487\ 76_0 \pm .000\ 12$	$1.487\ 62 \pm .000\ 18$		
			$25.2_5 \pm .1$	$124.372_7 \pm .000_6$	124.359_9	10.758_0	$18.680_6 \pm .001$	$1.487\ 33_7 \pm .000\ 12$	$1.487\ 67 \pm .000\ 18$		
			$26.6_5 \pm .1$	$124.386_1 \pm .000_6$	124.373_3	10.744_6	$18.680_3 \pm .001$	$1.485\ 46_1 \pm .000\ 12$	$1.487\ 68 \pm .000\ 18$		
W-3 Standard	$135.519_8 \pm .000_2$	135.512_4	$25.1 \pm .1$	$124.730_6 \pm .000_5$	124.718_8	10.794_6	$18.675_1 \pm .001$	$1.487\ 61_5 \pm .000\ 11$	$1.487\ 75 \pm .000\ 17$		
									Avg $1.487\ 68 \pm .000\ 18$		
2020-A	$132.583_4 \pm .000_2$	132.575_8	$26.4 \pm .2$	$122.607_9 \pm .001_1$	122.595_4	9.980_4	---	$1.485\ 79 \pm .000\ 33$		$19.736_7 \pm .004_9$	$19.740_9 \pm .005_0$
			$26.8 \pm .5$	$122.612_5 \pm .001_1$	122.600_0	9.975_8	---	$1.485\ 25 \pm .000\ 69$		$19.738_6 \pm .009_4$	$19.744_0 \pm .009_4$
			$27.0 \pm .1$	$122.609_4 \pm .001_1$	122.597_0	9.978_9	---	$1.484\ 99 \pm .000\ 23$		$19.729_0 \pm .003_8$	$19.735_0 \pm .004_0$
											W'ted Avg $19.737 \pm .005$
2021-A	$132.639_7 \pm .000_6$	132.632_1	$26.9 \pm .2$	$122.651_7 \pm .001_2$	122.639_2	9.992_9	---	$1.485\ 12 \pm .000\ 33$		$19.711_5 \pm .005_2$	$19.717_2 \pm .005_3$

TABLE V

COMPARISON OF DENSITY DETERMINATIONS

Description of Sample	Method	Group	Weight in Air (g)	Temp. (^oC)	$(\Delta\rho/\rho)_{Fluid}$	ρ^t_{Pu}	ρ^{25}_{Pu}
2020 As Cast	Im. Weigh.	CMR-5	144.8063	23.3	-125×10^{-5}	$19.54_3 \pm 0.02$	$19.53_8 \pm 0.03$
Pressed	Dim.	CMR-5	144.5568	25 ± 5	-----	$19.68_0 \pm .04$	$19.68_0 \pm .04$
	Im. Weigh.	CMR-5	144.5568	19.8	-120×10^{-5}	$19.71_2 \pm .02$	$19.69_6 \pm .03$
Machined	Dim.	CMR-13	142.420	28 ± 2	-----	$19.70_9 \pm .00_9$	$19.71_8 \pm .01_1$
	Dim.	CMR-11	142.420	25 ± 5	-----	$19.69_4 \pm .00_9$	$19.69_4 \pm .01_8$
	Im. Weigh.	CMR-11	142.420	19.0	$+100 \times 10^{-5}$	$19.73_9 \pm .04$	$19.72_1 \pm .04$
2020-A Remachined	Dim.	CMR-11	132.587	25 ± 5	-----	$19.69_2 \pm .00_8$	$19.69_2 \pm .01_7$
	Im. Weigh.	CMR-11	132.587	18.4	$+100 \times 10^{-5}$	$19.72_6 \pm .04$	$19.70_6 \pm .04$
	Dim.	CMR-13	132.5834	25 ± 2	-----	$19.69_4 \pm .01_1$	$19.69_4 \pm .01_3$
	Im. Weigh.	CMR-13	132.5834	-----	$-34 \pm 12 \times 10^{-5}$	see Table IV	$19.737 \pm .005$
2021 As Cast	Im. Weigh.	CMR-5	150.4807	23.5	-130×10^{-5}	$19.52_3 \pm 0.02$	$19.51_8 \pm 0.03$
Pressed	Dim.	CMR-5	140.3392	25 ± 5	-----	$19.66_3 \pm .04$	$19.66_3 \pm .04$
	Im. Weigh.	CMR-5	140.3392	22.0	-125×10^{-5}	$19.68_5 \pm .02$	$19.67_6 \pm .03$
Machined	Dim.	CMR-11	138.842	25 ± 5	-----	$19.66_9 \pm .01_1$	$19.66_9 \pm .01_9$
	Im. Weigh.	CMR-11	138.842	19.0	$+100 \times 10^{-5}$	$19.69_5 \pm .04$	$19.67_7 \pm .04$
2021-A Remachined	Dim.	CMR-11	132.644	25 ± 5	-----	$19.64_2 \pm .01_9$	$19.64_2 \pm .02_4$
	Im. Weigh.	CMR-11	132.644	18.4	$+100 \times 10^{-5}$	$19.69_2 \pm .04$	$19.67_2 \pm .04$
	Dim.	CMR-13	136.6397	25 ± 2	-----	$19.65_2 \pm .02$	$19.65_2 \pm .02_1$
	Im. Weigh.	CMR-13	136.6397	-----	$-34 \pm 12 \times 10^{-5}$	see Table IV	$19.717 \pm .005$

[redacted]

UNCLASSIFIED

techniques and whose densities are appreciably below the "theoretical" value for the metal.

b. Obtain better measurement and perhaps even control of the temperature of the immersion fluid to at least ±0.01°C. Eliminate reading of thermometer by use of recorder.

c. Have balance outside and above dry box.

d. Refine study of convection effects.

e. A concerted effort to produce material of highest density.

f. Redetermine density of the same specimens in six months to a year, as a check on the possibility of cumulative radiation damage.

UNCLASSIFIED

[redacted]

[redacted]

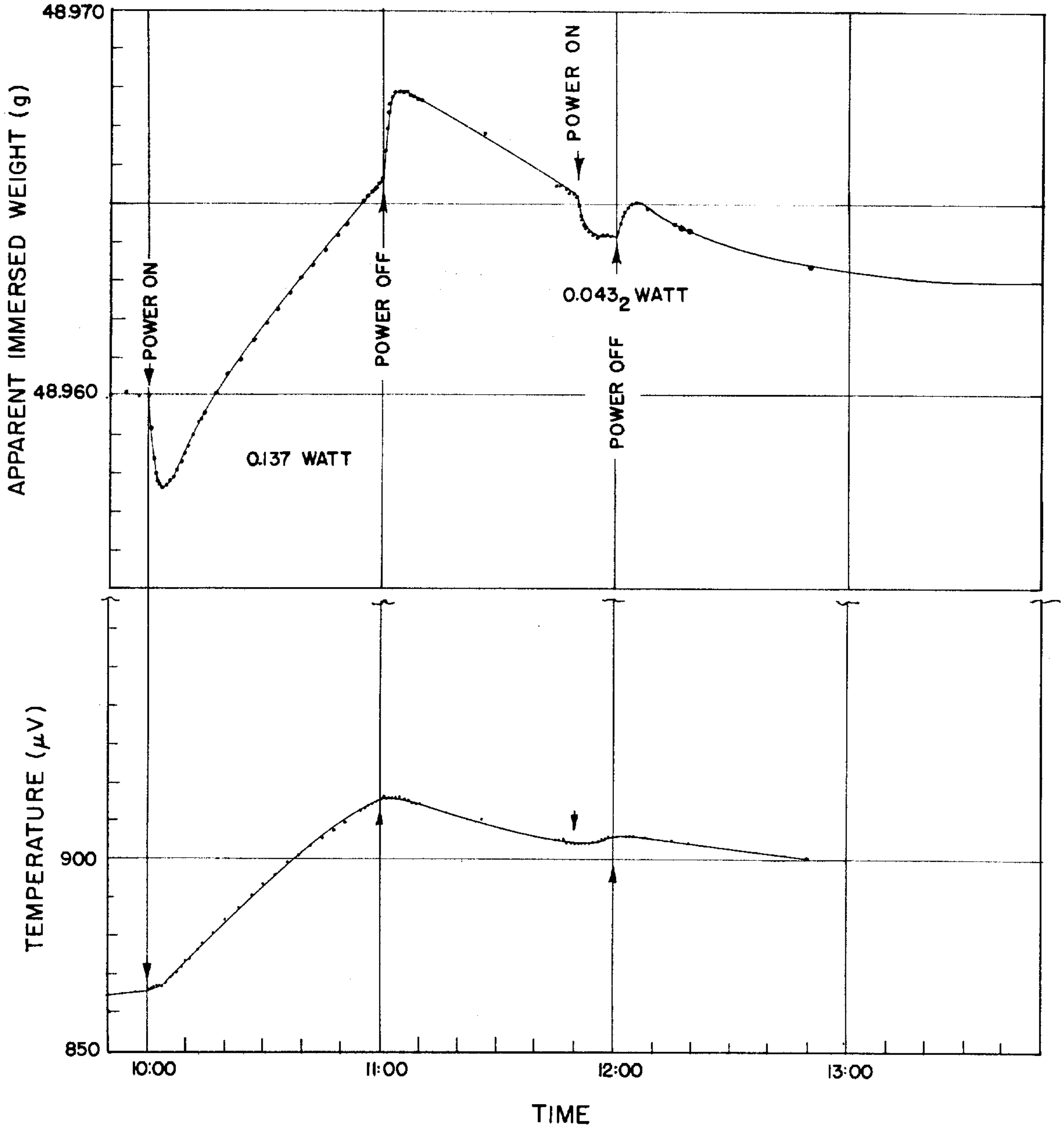

Fig. 1 Measurement of Heating Effects

[redacted]

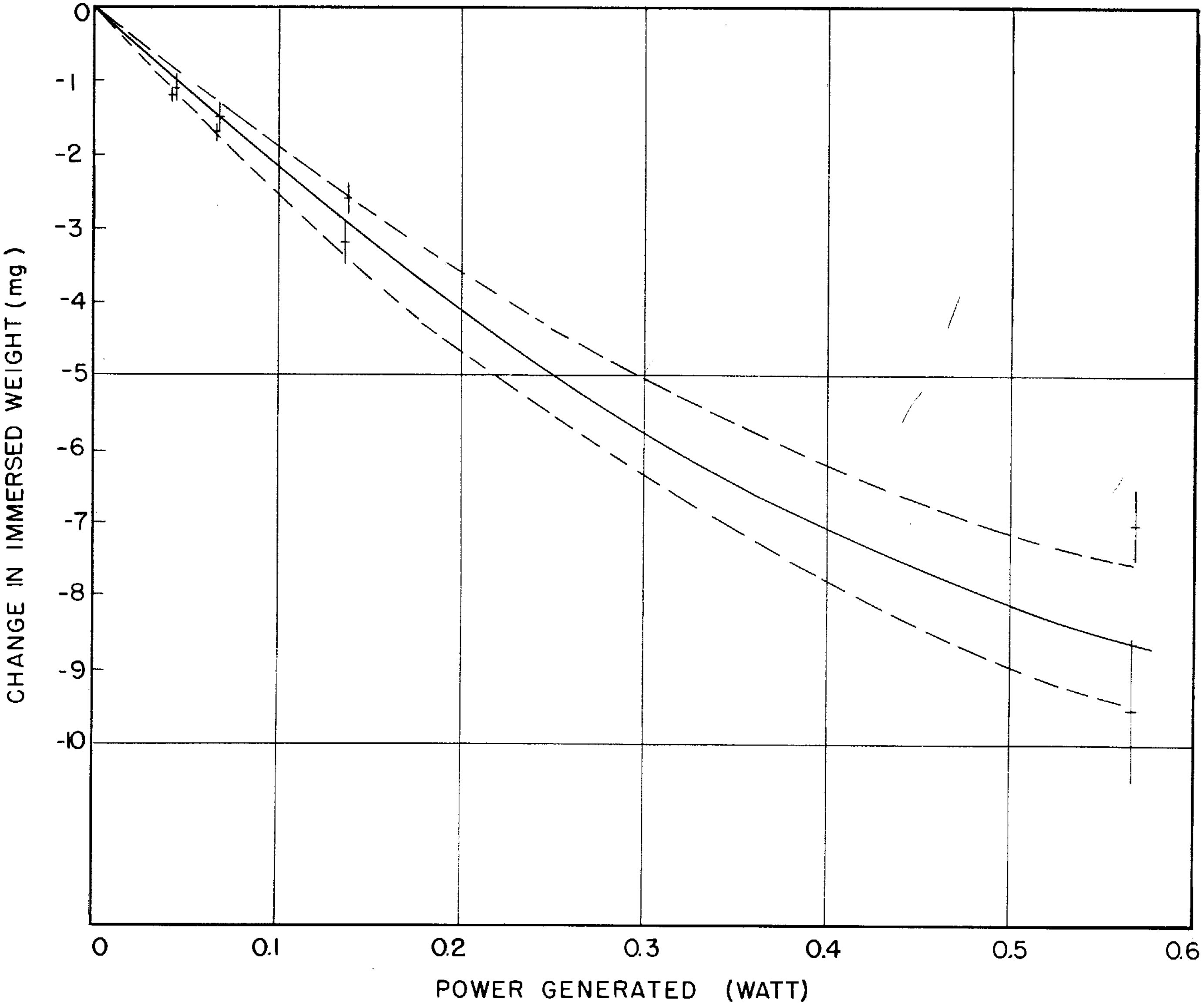

Fig. 2 Effect of Convection Currents

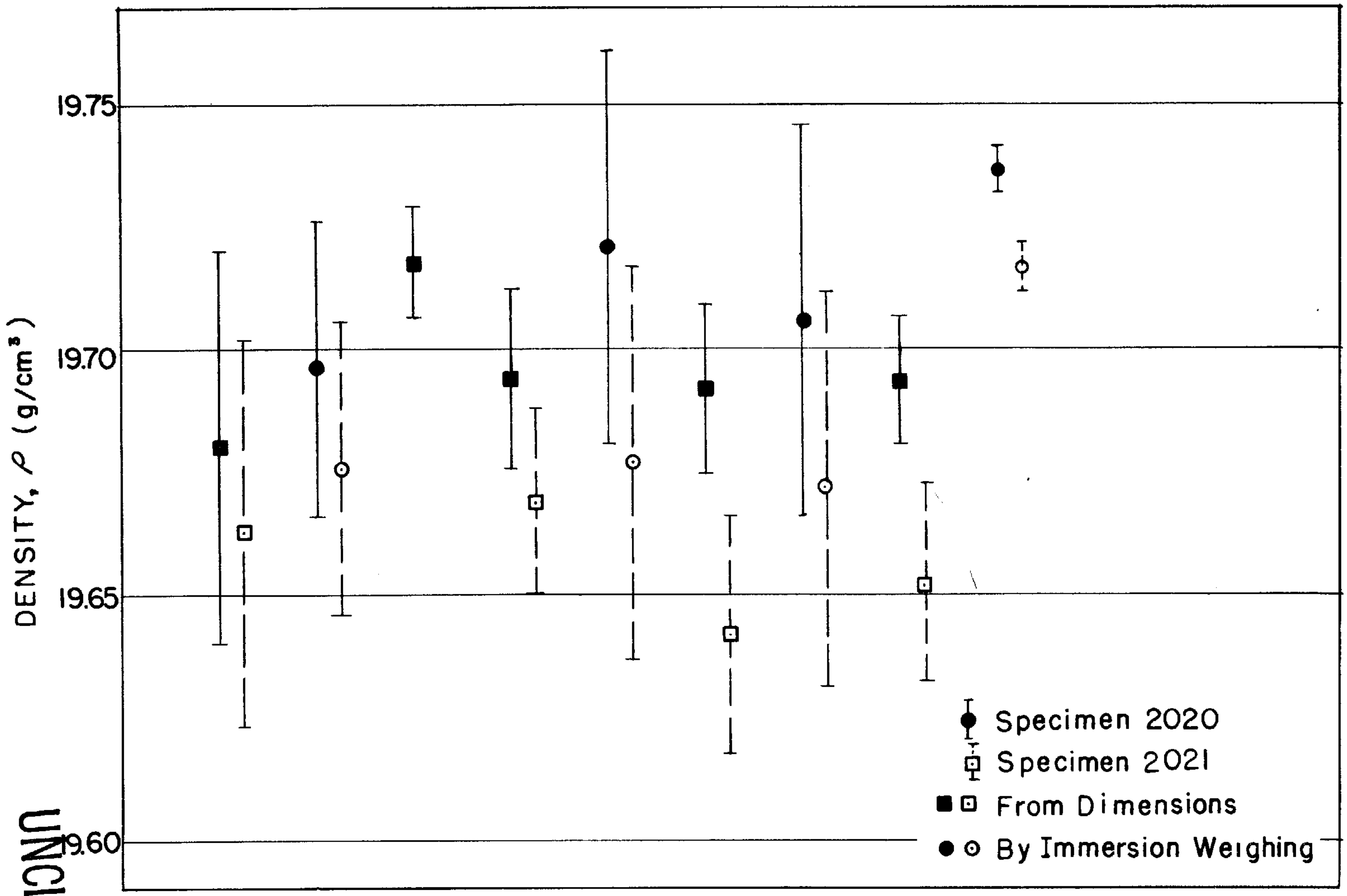

Fig. 3 Summary of Density Determinations (cf. Table V)

SIFIED