ATOMIC ENERGY OF CANADA LIMITED

CHALK RIVER PROJECT

RESEARCH AND DEVELOPMENT

DETERMINATION OF THE DENSITY OF ACTIVE URANIUM

CRMET-752

BY

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TABLE OF CONTENTS

ABSTRACT

A procedure was found to measure the density of irradiated uranium to an accuracy of 0.06% by measuring the weight of the sample in air and in n-octyl alcohol. The measurements were made using a gramatic balance that was readily adapted for remote control in the "cave". Since the n-octyl alcohol was inside the balance for all measurements, the complete apparatus was mobile.

INTRODUCTION

A measurement of the change in density of uranium caused by irradiation will indicate the amount of lattice defects, porosity and cracking produced., The density measurement is more difficult to perform on irradiated uranium because the surface of the sample is usually quite rough and because all the operations must be performed by remote control. A technique developed by G.R. Mallett[1] determines the density of irradiated uranium from the weight of the samples in n-octyl alcohol and air inside a complicated lead-shielded dry box, Since the measurements to be done would be carried out in a 'hot cell" or "cave", a much simpler apnaratus could be used.

The most convenient method to measure the density is to weigh the sample in air and in liquid., If the liquid can be kept inside the balance, the whole apparatus can easily be moved into the cave. This method of density measurement was chosen so that large samples with a hih ratio of volume to surface could be used thus reducing the effect of the rough surface.

DESCRIPTION OF APPARATUS

A gramatic balance with 200 gram maximum load and 0.1 milligram sensitivity was readily adapted for remote control. The weight of the sample in air and in liquid was performed using the special balance pan shown in Figure 1, The upper pan was aluminum, the lower pan copper mesh supported by four 28 gauge chrome1 wires.

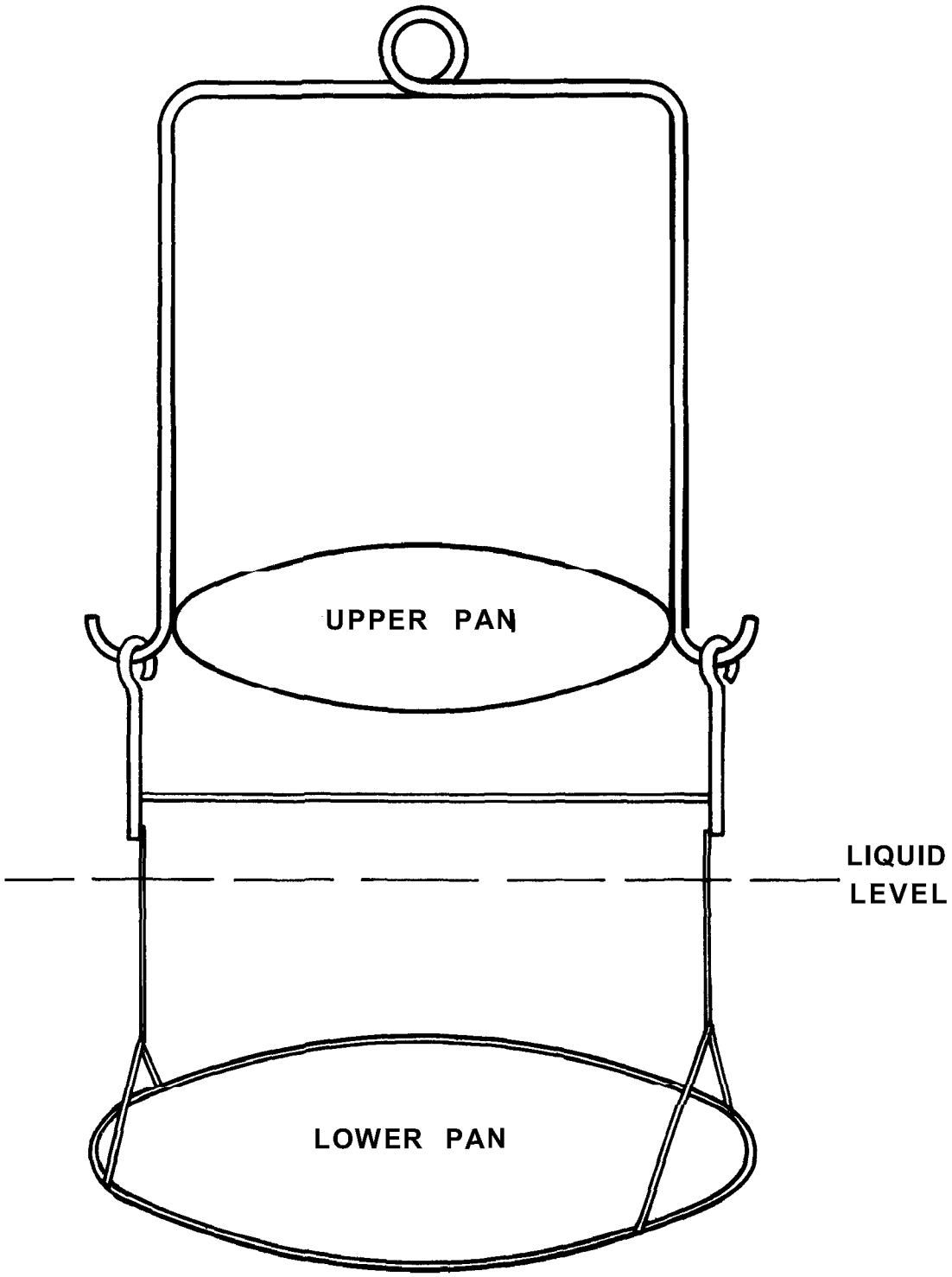

Figure 1: Balance pan assembly for density measurements.

The samplewas put in the upper pan for the weight in air and the lower pan for the weight in liquid. Because the lower pan was always in the liquid, the calculated density was independent of the surface tension caused by the wires entering the liquid. Normally, water is the most convenient liquid to use for density measurements since its density is well tabulated for all temperatures. However, when water was tried, the water vapour in the balance influenced the reading so that just opening and closing the balance door produced a change. To overcome this, aniline and n-octyl alcohol, which have lower vapour pressures, were tried. These were more satisfactory and n-octyl alcohol was finally used for all measurements.

DENSITY OF N-OCTYL ALCOHOL

For preliminary experiments, n-octyl alcohol that had been stored for several years and had turned slightly yellow was used. This was found to have a density three per cent higher than that measured by Mallett[1]. Therefore, fresh liquid was obtained and its density was measured in the temperature range 21 to 26°C by the pycnometer method. The results of these measurements are shown in Figure 2, along with those of Mallett[1] and Dreisbach[2]. The difference between the results reported here and those of Mallett and Dreisbach at 25°C is 0.03 per cent which is within the expected error of the uranium density measurements,,

PROCEDURE

The procedure for measuring the density of uranium that was found most satisfactory is given in detail in Appendix 1. Using this procedure, each density measurement in the cave took about

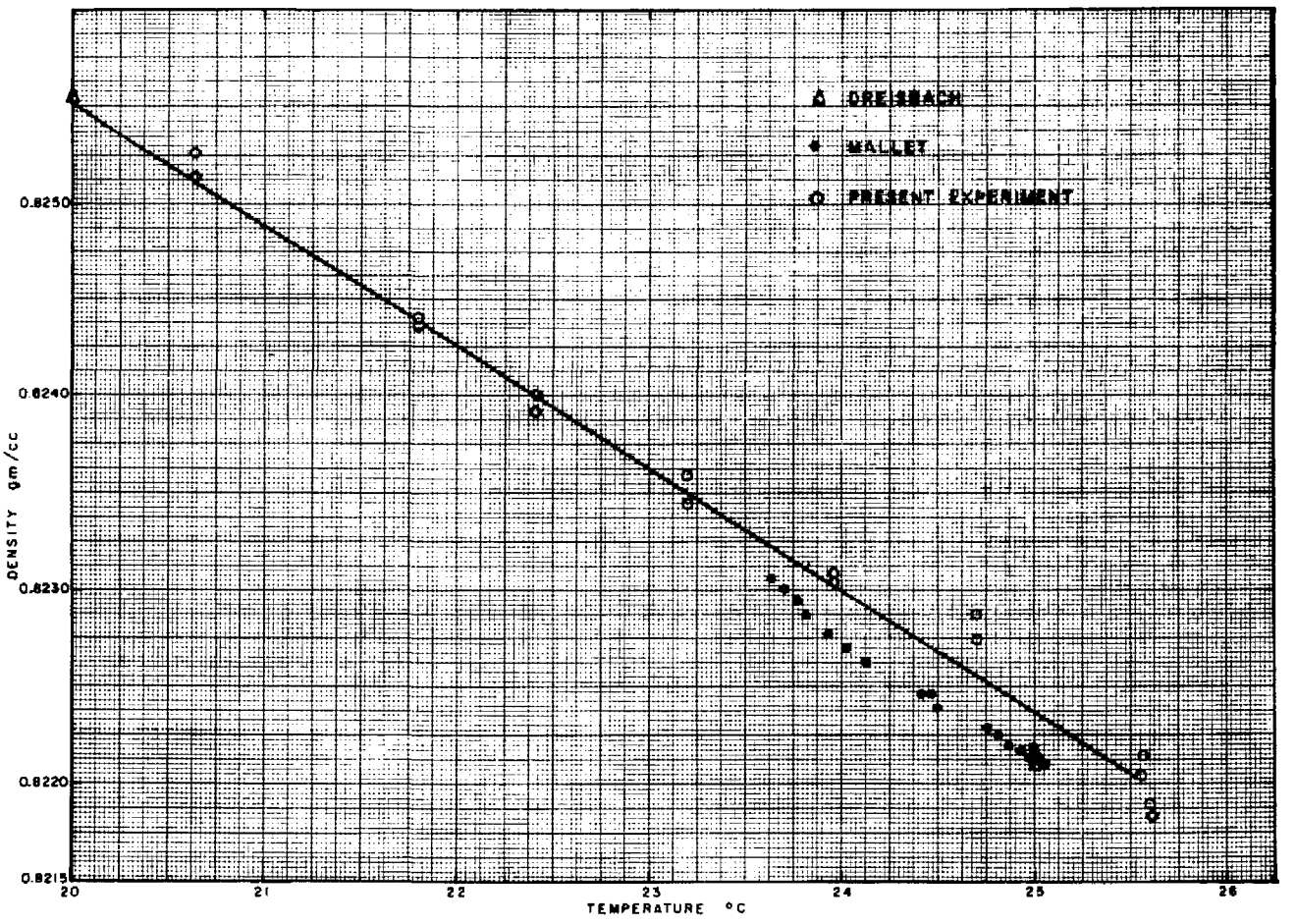

Figure 2: Density of n-octyl alcohol measured with a pycnometer.

80 minutes.

PRELIMINARY EXPERIMENTS

Because the surface of the irradiated samples is very rough, preliminary experiments were done on one piece of unirradiated uranium that was etched in nitric acid for increasing periods of time in order to investigate the effect of this rough surface on the measured density., All the calculated values of the density from these experfments were the same to within 0.02%. The results were reproduceable only if the sample was kept under the liquid a sufficient time to allow the weight to reach equilibrium, With a mildly etched sample, equilibrium was reached in ten minutes but for a heavily etched sample, it tonk thirty minutes. With irradiated samples, desheathed and etched five minutes in concentrated nitric acid, equilibrium was reached in thirty minutes. The weight of the sample in liquid always increased to the equilibrium value, indicating that the pores of the sample were finally filled with liquid when equilibrium was reached, No further change in weight occurred even when the sample remained in liquid overnight.

In order to determine whether the equilibrium reached by waiting thirty minutes corresponded to all the sample pores being filled with liquid, a separate experfment was performed in which the sample was first placed in a flask wfth n-octyl alcohol, that was then evacuated. Since the evacuation allows most of the gas to escape from the pores, they will then fill with liquid when the vacuum is released, Any air remafning will be absorbed by the degassed lfquid, After releasing the vacuum the sample was trans-

ferred directly onto the lower pan of the balance with a film of liquid remaining on it during the transfer, The weight of the sample in liquid was the same as that obtained in the previous case by waiting thirty minutes for the sample weight to reach equilibrium. This indicates that all the air in the sample pores is displaced by liquid before the weight reaches equilibrium.

The accuracy of the density determinations was checked by measuring the density of an aluminum magnesium alloy that had previously been measured using water as the reference liquid [3]. The comparative values are shown in Table 1. The values using n-octyl alcohol are 0.04% higher than those determined in water, but this accuracy is sufficient for the proposed measurements on uranium.

Table 1

Density of Al-2.87% Mg Measured in Water [3] and in n-Octyl Alcohol

Density at 20°C	
in water [3]	in n-octyl alcohol
2.6617	2.6627
	2.6626
	2.6629

In order to find the change in density due to irradiation, the original density of the irradiated piece must be known. If all uranium has the same density, then the original density need not be measured since an average value can be obtained from several

other pieces. To check the variation In density of uranium, several pieces were measured that had been selected at random from a large scrap pile. The results tabulated in Table 2 show that the density of uranium depends markedly upon its history and perhaps impurity content.

It is concluded, therefore, that the density of the uranium sample should be measured before and after Irradiation, or if this is not possible, an exact duplicate from the adjacent section of the original metal; should be measured.

The values for the density of irradiated uranium measured by remote control in the cave were reproduceable to within 0.06% for a 100 gram sample.

After the balance was in the cave for several weeks, the prfsms became "cloudy" due to the radiation. They were, therefore, replaced periodically.

Table 2

Density of a Random Selection of Uranium Pieces

No.	Density gm/cc 23°C	Sample Source
1	18.955	rolled rod
2	18,823	" "
3	18.815	" "
4	18.963	" "
5*	18,927	rolled flat
6*	18.927	" "
7	18.904	" "
8	18.912	" "
9	18,926	" "

* - Samples 5 and 6 were identical as though they were cut from the same flat,

CONCLUSION

The density of irradiated uranium can be measured to ± 0.06% by weighing the sample in air and in n-octyl alcohol that is inside the balance. This is a simple operation that can be done readily by remote control in a "hot cell" or "cave".

BIBLIOGRAPHY

1. G.R. Mallet, Report HW-34079 Rev,

2. R.R. Dreisbach and R.A. Martin, Ind, Eng. Chem. 41, 2875(1949).

3. E.C.W. Perryman, Trans. AIME 206, 1247(1956).

APPENDIX 1

PROCEDURE FOR DENSITY MEASUREMENTS IN CAVE

1. Remove any burrs left on sample by the cut-off wheel.
2. Etch the sample heavily with conc. HNO_3 at room temperature.
3. Wash the sample in water,
4. Wash the sample in alcohol,
5. Dry the sample in a stream of unheated air from a fan for at least 15 minutes, This length of time is necessary to ensure that the alcohol has evaporated from the sample and that the sample has reached room temperature again.
6. Record the weight of the empty pans.
7. Record the temperature of the n-octyl alcohol to at least $0.2^{\circ}C$ and preferably to $0.1^{\circ}C$.
8. Put the sample in the upper pan and record the weight.
9. Remove the sample from the balance and record the weight of the empty pans,
10. Repeat No. 8, No. 9, No. 7.
11. Put the sample in the lower pan under the liquid. Record the weight after 1, 2, 5, 10, 15, 20 and 30 minutes with the weight remaining on the knife edges, This should reach equilibrium in about 10 - 20 mfnutes if the temperature is constant. Take readings until the weight has reached equilibrium, or until a constant correlation with temperature change is found.
12. Measure the temperature as in No, 7.
13. Remove Sample from balance - wash in methyl alcohol and dry in air, This should be a different beaker of alcohol than the previous one if other samples are to be measured.
14. Record the weight of the scale pan assembly,

APPENDIX 2

CALCULATION OF THE DENSITY

The density of the sample was calculated from the following formula:

$$d_s = \frac{w_a \ d_l - w_l \ d_a}{w_a - w_l}$$

where w_a, w_l are the weight of the sample in air and liquid, d_s, d_l, d_a are the density of the sample, liquid and air at the temperature measured, The density of n-octyl alcohol was taken from the results shown in Figure 2.

The sample density was corrected to a mean temperature 23.0°C taking the coefficient of linear expansion 15 x 10^{-6}/°C. This gave a correction of 0.0009 gr/cm^3/°C.