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THE TOTAL HEMISPHERICAL EMITTANCE OF
POLISHED AND OF OXIDIZED
ALPHA PLUTONIUM

- PRELIMINARY REPORT -

by

R. H. Karlsson

THE DOW CHEMICAL COMPANY
ROCKY FLATS DIVISION
U.S. ATOMIC ENERGY COMMISSION
CONTRACT AT (29-1) - 1106

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ABSTRACT

The total hemispherical emittance of polished 99.66 w/o plutonium was determined to be 0.37 at 88.7° and 89.0°C.

The emittance of a polished sample was measured after oxidation in a humid air atmosphere for 24, 66, and 168 hours. Emittances of 0.47, 0.54, and 0.70 were obtained. The apparatus was used to determine a total hemispherical emittance for candle soot of 0.96. Total errors were estimated to be less than $\pm 3\%$.

INTRODUCTION

Total hemispherical emittance is the ratio of the total thermal energy radiated from a unit area of surface at a given temperature to that radiated by a blackbody, or complete radiator, at the same temperature. Plutonium is being considered for applications in which a knowledge of its total hemispherical emittance is needed. This information has not been found in the available literature.

EQUIPMENT

In this study the total hemispherical emittance of polished and oxidized alpha phase plutonium was measured by a method similar to that described by Drummeter and Goldstein.¹ In this method, a heated spherical sample is suspended within an evacuated larger hollow sphere of known emissivity.

A sketch of the experimental apparatus is shown in Figure 1. A plutonium ball, 1 inch in diameter, is suspended concentrically within a hollow aluminum sphere, approximately 12 inches in diameter, by a 0.010-inch tungsten wire. The inside of the aluminum sphere is coated with Cat-A-Lac* bakeable flat black paint having a known emittance of 0.9. The ball and hollow sphere are supported inside a vacuum bell-jar evacuated to 10^{-6} - 10^{-7} Torr on a tripod insulated with Teflon. Two 0.010-inch calibrated iron-constantan thermocouples, one pinned into a small hole in the top of the ball and the other imbedded at the inner surface of the sphere, are used to measure the ball and sphere temperatures at thermal equilibrium. The thermocouple efms are measured

*Trademark of the Finch Paint and Chemical Company, Torrance, California

Figure 1. APPARATUS FOR THE DETERMINATION OF THE TOTAL HEMISPHERICAL EMITTANCE OF PLUTONIUM

by a Certified Leeds and Northrup K-3 Potentiometer. This measuring system allows temperature determinations to be made to within 0.1°C .

PROCEDURE

The plutonium ball was machined from a casting which assayed 99.66 w/o plutonium. Major impurities were Al, Cr, Fe, and Ni. Table I shows the results of an isotopic analysis and the heat contribution of the various isotopes² from which

TABLE I
ISOTOPIC ANALYSIS OF THE PLUTONIUM BALL AND THE HEAT
CONTRIBUTION OF THE VARIOUS ISOTOPES

Isotope	w/o	Heat Contribution (mw/g)
Pu ²³⁸	0.01	0.057
Pu ²³⁹	93.54	1.799
Pu ²⁴⁰	5.94	0.418
Pu ²⁴¹	0.49	0.022
Pu ²⁴²	0.02	0.002
Am ²⁴¹	0.014	0.016
		<hr/> 2.314

the rate of self-heat generation was calculated. The plutonium sample used produced heat at the rate of 2.314 mw/g. The surface of the ball was finished by first lapping and then polishing with a soft cloth and 6- μ diamond paste. The oxidized plutonium samples were prepared by suspending a polished ball in a closed container which held a small amount of water.

The unusual self-heating property of plutonium, a result of alpha decay, provides a ready-made constant heat source for emittance measurements. This eliminates the need for internal heating coil leads which cause unwanted conduction losses. The emittances of many materials can be measured

utilizing this self-heating phenomenon by coating or plating plutonium with the material whose emittance is to be determined. An emittance for candle soot was determined by this technique.

The emittance of the sample is calculated from the following relationship derived from the Stefan-Boltzman equation³:

$$\epsilon_p = \frac{1}{\frac{\sigma A_p (T_p^4 - T_s^4)}{Q_p M - K(T_p - T_s)} - \frac{A_p (\frac{1}{\epsilon_s} - 1)}{A_s}} \quad (1)$$

where:

- ϵ_p = Total hemispherical emittance of the sample ball
- σ = Stefan-Boltzmann constant ($5.669 \times 10^{-12} \text{ w/cm}^2 \text{ } ^\circ\text{K}^4$)
- A_p = Surface area of the sample ball at the test temperature (cm^2)
- A_s = Inside surface area of the coated aluminum sphere (cm^2)
- T_p = Temperature of sample ball at equilibrium ($^\circ\text{K}$)
- T_s = Inside surface temperatures of the coated aluminum sphere at equilibrium ($^\circ\text{K}$)
- Q_p = Rate of self-heat generation of the sample ball (w/g)
- M = Plutonium mass of the sample ball (g)
- ϵ_s = Emittance of the inside surface of the hollow aluminum sphere

K = Thermal conductivity loss factor for thermocouple and suspension wires ($w/^{\circ}\text{K}$)

The value of the second term in the denominator of equation (1) is on the order of 0.0008 and can be eliminated without significant error. Equation (1) then reduces to:

$$\epsilon_p = \frac{Q_p M - K(T_p - T_s)}{\sigma A_p (T_p^4 - T_s^4)} \quad (2)$$

RESULTS

Substitution of the measured data into equation (2) yielded an emittance of 0.37 for the polished plutonium ball at a ball temperature of 88.7°C . A repetition of this experiment using the same ball repolished yielded an emittance of 0.37 but at an equilibrium temperature of 89.0°C . The emittance of the ball oxidized for 24 hours was 0.47. After oxidation for 66 hours the emittance was 0.54 and after 168 hours it was 0.70. Data for these calculations are listed in Table II. Table III is a summary of the emittances obtained from the calculations.

The total hemispherical emittance of candle soot applied to the plutonium ball was calculated from equation (2) to be 0.96 at 56.3°C . The American Institute of Physics Handbook⁴

TABLE II

EXPERIMENTAL DATA USED IN THE CALCULATION OF THE TOTAL
HEMISPHERICAL EMITTANCE OF POLISHED PLUTONIUM, OXIDIZED
PLUTONIUM, AND CANDLE SOOT

Material and Condition	Surface Area of Pu Sample (cm ²)	Mass of Pu Sample (g)	Temp. of Pu Sample (°C)	Temp. of Sphere (°C)
Polished alpha plutonium Run #1	20.17	165.004	88.7	28.8
Run #2	20.17	164.981	89.0	28.2
Polished alpha plutonium oxidized for:				
24 hours	20.16	164.947	79.0	28.7
66 hours	20.14	164.928	72.4	27.3
168 hours	20.13	164.886	63.9	27.3
Candle Soot	20.40	164.791	56.3	29.3

Area of sphere = 2917 cm² conductivity loss factor = sum of
(thermal conductivity) (length) for the suspension and thermocouple
area

wires = 7.69×10^{-5} w/°K

TABLE III

TOTAL HEMISPHERICAL EMITTANCES OF POLISHED PLUTONIUM,
OXIDIZED ALPHA PLUTONIUM AND CANDLE SOOT

Material and Condition	Temperature of Determination, °C	Emittance
Polished alpha plutonium	88.7, 89.0	0.37
Polished alpha plutonium oxidized for:		
24 hours	79.0	0.47
66 hours	72.4	0.54
168 hours	63.9	0.70
Candle Soot	53.3	0.96

lists the total normal emittance of candle soot as 0.952 in the temperature range 0° - 100° C. Total hemispherical and total normal emittance are usually nearly the same. The total error in the emittances obtained in this study are estimated to be less than +3%.

DISCUSSION

The polished plutonium sample used in this study undoubtedly acquired a slight coating of oxide prior to the measurement of its equilibrium temperature. The data in Table III show the emittance of oxidized plutonium is dependent either upon the degree of oxidation, the temperature of determination, or upon both of these variables. The temperature of determination, at least in the range in this study, probably has the least affect on emittance. An extensive compilation of emittance data on metals⁵ shows that the temperature dependence of emittance is generally quite small. From these considerations, it may be inferred that a plutonium sample with a lesser quantity of oxide present on its surface will have a lower emittance. Also, the total hemispherical emissivity of plutonium, the intrinsic bulk property of the material determined from an optically polished unoxidized surface, is probably less than the emittance of a slightly oxidized surface and therefore less than 0.37. Continuation of this study will prove or disprove these contentions.

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