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A METHOD FOR DETERMINING THE CURIE LOADING
OF Kr⁸⁵ IN NUCLEAR BATTERIES

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ABSTRACT

This report describes a calorimeter which is used to determine the amount of Krypton 85 (in curies) in a nuclear battery. The construction of the calorimeter is described, and a sample measurement is included.

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A METHOD FOR DETERMINING THE CURIE LOADING OF KR⁸⁵ IN NUCLEAR BATTERIES

Introduction

When the problem of measuring the amount of Krypton 85 (in curies) contained in nuclear batteries was given to the Radiation Special Studies Section, a number of systems were investigated, but it was decided to use a calorimetric system using the Peltier effect. This system was first used by H. L. Callandar of the Imperial College of Science and Technology in London, England, in 1910. Callandar called his device a "Radiation Balance,"⁽¹⁾ and he used it to measure the sun's radiation, and in collaboration with Rutherford, he measured the heat output of several radioactive sources.

In 1928, F. E. Hoare in England⁽²⁾ used the system to accurately determine the Boltzmann Constant. Thereafter, no one used the idea to any great extent until in 1954 W. B. Mann of the National Bureau of Standards⁽³⁾ used it to cross-check the radium standards of Great Britain, Australia, France, and Canada.

Figure 1 is an electrical schematic of the Sandia calorimeter. Cups 1 and 2 are made of copper. Each cup is surrounded by a thermopile consisting of six chromel-constantan thermocouples in series. This thermopile is insulated from the cup. A Peltier junction made of a single chromel-constantan thermocouple is soft soldered to the bottom of the cup.

The thermopiles of cups 1 and 2 are connected in series with one another and with the galvanometer and are also connected in opposition to each other; thus, an equal rise in temperature in cups 1 and 2 will give a zero deflection. The Peltier junctions are also connected both in opposition and in series so that the same current flows through both and will cause heating in one cup and cooling in the other.

The calorimeter was used by Callandar in this manner in order to eliminate the Joule heating of the Peltier junctions. Assume that a radioactive source dissipating W_1 watts is placed in cup 1 and another source dissipating W_2 watts is placed in cup 2; assume also that W_1 is greater than W_2 and that a current C is passed through the Peltier junction in such a direction as to cool cup 1 and heat cup 2. The value of C is adjusted to give a galvanometer deflection of zero. Suppose, however, the residual galvanometer deflection is d_1 . The sources are now interchanged between cups 1 and 2, and the current C is reversed so that cup 1 is heated and cup 2 is cooled; a new residual galvanometer deflection d_2 is then noted. Under these conditions, Callandar showed that if W_1 and W_2 are heat sources in watts in cups 1 and 2 capable of being interchanged, P_1 and P_2 are coefficients of the Peltier

Figure 1. Electrical Schematic of the Radiation Balance

effect for couples soldered to cups 1 and 2, and R_1 and R_2 are effective resistances of Peltier couples for cups 1 and 2, then

$$P = \frac{P_1 + P_2}{2} = \text{mean of Peltier coefficients.}$$

Let: S_1 and S_2 = total deflection per watt transferred from cup 1 to cup 2,

d_1 = observed residual deflection with W_1 in cup 1 and W_2 in cup 2 balanced by a current cooling cup 1 and heating cup 2, and

d_2 = the corresponding deflection measured in the same direction when W_2 and W_1 are interchanged and the same current reversed.

Collecting the quantities of heat supplied to either cup, we have

$$(1) \quad S_1(W_1 - Cp_1 + C^2R_1) - S_2(W_2 + Cp_2 + C^2R_2) = d_1$$

$$(2) \quad S_1(W_2 + Cp_1 + C^2R_1) - S_2(W_1 - Cp_2 - C^2R_2) = d_2$$

Taking the difference between these two equations, the joule effects are exactly eliminated even if S_1 and S_2 and R_1 and R_2 are widely different, provided the current is unaltered on reversal. We thus obtain

$$(3) \quad (S_1 + S_2)(W_1 - W_2) - 2C(P_1S_1 + P_2S_2) = d_1 - d_2.$$

Since P_1 and P_2 are always very nearly equal, and S_1 and S_2 rarely differ by so much as 2 or 3 percent, we may write $(P_1S_1 + P_2S_2) = PS$, which gives the equation

$$(4) \quad W_1 - W_2 = 2 PC + \frac{(d_1 - d_2)}{S}.$$

In actual measurement of the Kr^{85} batteries a dummy nuclear battery was used for W_2 , and when the dummy and live battery were interchanged, the Peltier current C was reversed by means of a switch without altering the value of C . W_2 is then equal to 0 and $d_1 = 0$. The equation is now.

$$(5) \quad W_1 = 2 PC + \left(\frac{\pm d_2}{S} \right).$$

All the factors of equation (5) can be obtained directly with the exception of S .

The approximate value of S was determined by reversing a small current through the Peltier junctions connected in opposition with no source in either cup, and noting the change in galvanometer deflection. Since W_1 and W_2 are now zero, S can be determined directly from equation (4) by using the thermoelectric value of P determined from the formula

$$(6) \quad P = T \frac{de}{dt}.$$

The thermoelectric tables compiled by National Bureau of Standards can be used to determine $\frac{de}{dt}$

Since $T = \text{degrees Kelvin}$, we have now

$$(7) \quad S = \frac{d_1 - d_2}{2 PC} \cdot$$

The Construction of the Sandia Radiation Balance or Calorimeter

Figure 2 gives the construction details of the Sandia Corporation calorimeter. The whole assembly, including the cups, was made of copper. The dimensions of the cups can be obtained from Figure 3.

The Peltier junctions were made from two #24 gauge chromel-constantan wires silver soldered together after the junctions were constructed. They were then soft soldered to the bases of the cups.

The galvanometer thermopiles were fabricated from six #30 gauge chromel wires and six #30 gauge constantan wires. The wires were pushed through a lucite disc, and the ends were silver soldered together, a chromel to a constantan, making six junctions above and below the disc. The upper junctions were placed on a band of rubber hydrochloride which was wound around the cups. A band of adhesive tape was wound on top of the junctions, fastening them securely to the cups. The cups were placed in the calorimeter with the lower junctions immersed in vacuum oil.

The current for the Peltier junctions was obtained from a 9-volt Ray-O-Vac multiple ignition battery. It was controlled by a rheostat of 1000 ohms and was measured by taking the voltage drop across a 10-ohm resistor which was in series with the Peltier junctions. The IR drop derived was accurately measured with a Leeds and Northrup K3 potentiometer. A double-throw double-pole switch reversed the Peltier current when desired.

The electro-mechanical schematic is Figure 4. Both galvanometers are Kintel Model #204A. Because the room in which the calorimeter was placed was not temperature controlled, the calorimeter was placed on a radio chassis to insulate it from the bench top. It was surrounded by a lucite shield to protect it from drafts. If this is not done, it will be practically impossible to make any measurements. In a room which is temperature controlled, these precautions may not be necessary. A photograph of the calorimeter is shown in Figure 5.

The Determination of Kr^{85} in the Nuclear Battery

Because the power output of a nuclear battery is very minute, certain precautions must be taken. The instruments to be used must be turned on for at least

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Figure 3. Construction Details of the Copper Cups

Figure 4. Electro-Mechanical Schematic of the Sandia Corporation Calorimeter

Figure 5. Calorimeter

half a day before making any measurements; these are the two electronic galvanometers and the Leeds and Northrup K3 potentiometer.

When the instruments are sufficiently warmed up, the battery to be measured is placed in cup 1 and a dummy battery placed in cup 2. After a while the thermopile galvanometer will read some negative microvolt reading--for example, 20 microvolts.

When it has been established that the reading on the galvanometer is stable, the DPDT switch is thrown in such a way as to cool cup 1 and heat cup 2. The Peltier current is increased with the switch thrown this way until the galvanometer reads zero. The IR drop across the 10-ohm standard resistor is read on the K3 potentiometer. All these readings are recorded. When the galvanometer reaches zero, the nuclear battery is placed in cup 2 and the dummy is placed in cup 1. The current through the Peltier junctions is reversed by means of the DPDT switch. The current must be the same value as it was before reversal. The thermopile galvanometer will assume a value, after a while, somewhere in the vicinity of zero. The nuclear battery and the dummy are removed, the Peltier junction current is removed, and the thermopile galvanometer is allowed to come to some stable reading which is noted.

Sample Procedure and Calculations

1. Instruments allowed to warm up
2. Thermopile Galvanometer zeroed
3. Nuclear battery placed in cup 1
4. Dummy battery placed in cup 2
5. Galvanometer reading after 2 hours = -18 volts
6. Applied Peltier current and galvanometer bucked to zero,
Current = 18.92 ma.
7. Nuclear Battery placed in cup 2
8. Dummy battery placed in cup 1
9. Peltier Current reversed, Current = 18.92 ma
10. Thermopile galvanometer on reverse = +5.5 μ volts
11. Both cups emptied
12. Reading with both cups empty = + 1 μ volt

$$O_1 = 0$$

$$d_2 = +4.5 \mu\text{volts}$$

$$C = 18.92 \text{ ma}$$

$$T = 25^{\circ}\text{C} = 298^{\circ}\text{K}$$

$$\frac{de}{dt} = 60 \text{ } \mu\text{volts}$$

$$P = T \frac{de}{dt} = 17.880$$

$$S = .068$$

$$W_2 = 2 \text{ pc} + \frac{d_2}{S}$$

$$= 2(17880)1892 + \frac{4.5}{.068}$$

$$W_1 = 675.8 \text{ } \mu\text{watts}$$

$$W_2 = 741.8 \text{ } \mu\text{watts}$$

The amount of curies can be found by using the following formula:

$$\text{watts(H)} = 5.94 \times 10^{-3} \text{ A(curies) } \bar{E} \text{ (mev) (Reference 4).}$$

$$\text{Solving for curies} = \frac{\text{watts}}{5.94 \times 10^{-3} \text{ (.23 mev)}}$$

$$\text{curies} = \frac{\text{watts}}{130. \times 10^{-3}}$$

$$\text{curies} = \frac{741.8 \text{ } \mu\text{watts}}{1.36 \times 10^{-3}}$$

$$\text{curie} = 0.545$$

The average energy of the Beta particle emitted from the 9.4 year half life Krypton 85 is 0.23 mev $\pm$ 5%.

The total deflection per watt transferred from cup 1 to cup 2 (S) may be found by using equation (7).

The values of S given in the Table below were calculated using the given values of C, d_1 , d_2 and T.

C	d ₁ - d ₂	T	S
30 ma	72 μ volt	29.35	0.068
20 "	48 "	29.35	0.068
10 "	24 "	29.35	0.068
7.6 ma	18 "	29.35	0.068

The measurements on six nuclear batteries are now presented.

<u>Battery No.</u>		<u>Average curie</u>
269	=	0.663
271	=	0.613
242	=	0.557
299	=	0.552
344	=	0.370
361	=	0.529

These results were the averages of six separate determinations on each battery.

To find a constant in order to cross-check the curie loading and also to be able to evaluate future batteries, these six batteries were counted on a scintillation counter. The counts per minute were averaged for five minutes for each battery. The constant was obtained by dividing the curie loading determined from the calorimeter by the counts per minute obtained from the scintillation counter.

<u>Battery No.</u>		<u>< 0.35 mev Counts per minute</u>
344	=	43.6 x 10 ³
299	=	82.6 x 10 ³
269	=	85.2 x 10 ³
361	=	73.0 x 10 ³
271	=	79.5 x 10 ³
242	=	74.2 x 10 ³

Dividing the curie loading by the CPM gives the following results:

Battery No.	Calculation	K
344	$\frac{370 \times 10^{-3}}{43.6 \times 10^3}$	8.48×10^{-6}
299	$\frac{552 \times 10^{-3}}{82.6 \times 10^3}$	670×10^{-6}
269	$\frac{663 \times 10^{-3}}{85.2 \times 10^3}$	7.77×10^{-6}
361	$\frac{529 \times 10^{-3}}{73.0 \times 10^3}$	7.24×10^{-6}
271	$\frac{613 \times 10^{-3}}{79.55 \times 10^3}$	7.71×10^{-6}
242	$\frac{557 \times 10^{-3}}{74.2 \times 10^3}$	7.51×10^{-6}

Average K = 7.57×10^{-6}

The results after multiplying the CPM by the average K = 7.57×10^{-6} also the results derived from the calorimeter are given below.

I Battery No.	II Calorimeter curie	III CPM x 7.57×10^{-6} curie
269	0.66	0.64
299	0.55	0.62
271	0.61	0.60
242	0.55	0.56
361	0.52	0.55
344	0.37	0.33

The results in the third column are plotted in Figure 6, which is a plot of CPM versus curie loading. It can be seen that the plot is essentially a straight line.

Summary

The nuclear batteries have been successfully measured on the Sandia Corporation modification of the Radiation Calorimeter and cross-checked on a scintillation

Figure 6. Plot of Counts-Per-Minute Versus Curie Loading

counter. The scintillation counter method of counting the gamma rays above 0.35 mev seems to be the fastest way to evaluate a battery once the standards have been derived from the calorimeter. Each scintillation counting setup will have to be calibrated individually with standards previously derived.

Further improvements could be made on the calorimeter, such as more sensitive thermopiles, either by using more thermocouples in series or different thermocouples, or perhaps the new semi-conductor materials could be used. It is felt that with these improvements the determination of better standards would be possible.

It is also hoped that a few batteries can be calibrated by the National Bureau of Standards in order that the calorimeter standards can be tied in with the National Bureau of Standards.

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