International Journal of Applied Radiation and Isotopes, 1976, Vol. 27, pp. 43–45. Pergamon Press. Printed in Northern Ireland

Study of the Scattering Correction for Thick Uranium-Oxide and Other α-Particle Sources—II: Experimental

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(*Received* 23 *June* 1975)

Experimental values of the 2π α-particle counting rate, $C_{2\pi}$, including scattering, divided by the disintegration rate, N_0, are reported for uranium dioxide, ^{210}Po and ^{148}Gd sources mounted on platinum backings, and a ^{210}Po source on a U_3O_8 backing. The $^{235}UO_2$ source thicknesses varied from 0·05 to 1·1 mg/cm². The experimental values of the scattering are in agreement with theoretical values given in Part I (this issue), which take into account source thickness, α-particle energy, and composition of the backing material.

INTRODUCTION

The National Bureau of Standards has, for many years, measured the radioactivities of "weightless" α-particle sources deposited on monel or platinum surfaces. Recently, the activities of a number of thick (0·05 to 1·1 mg/cm²) sources, so-called fission deposits, of $^{235}UO_2$ and $^{238}UO_2$ were determined for the Neutron Standards Program. For the $^{238}UO_2$ sources in particular, the activities were too low to be accurately measured in a low geometry counter and therefore a $2\pi\alpha$ proportional counter was used. The 2π counting rate, $C_{2\pi}$, is not one-half of the disintegration rate, N_0, however, because some of the α-particles initially emitted downward are backscattered into the sensitive volume of the counter, while some of the α-particles initially emitted upward are scattered and/or absorbed in the source. As a result, the measured $C_{2\pi}/N_0$ ratios for these sources vary from approximately 0·52 to 0·47.

In order to determine accurately N_0 from a measurement of $C_{2\pi}$, the corrections for scattering and self-absorption must be well known. A few measurements and calculations of $C_{2\pi}/N_0$ for thick sources have appeared in the literature,[1–3] but, in general, the uncertainty in the reported values is unacceptably large for our purposes. We have measured the $C_{2\pi}/N_0$ ratios for ten $^{235}UO_2$ sources and one "weightless" ^{210}Po on U_3O_8 source in order to determine the dependence of this ratio on source thickness and composition of the backing, respectively, and to compare these results against the calculation developed in Part I (this issue). "Weightless" ^{148}Gd and ^{210}Po sources with α-particle energies of 3·1 and 5·3 MeV, respectively, were also measured to determine the dependence of $C_{2\pi}/N_0$ on α-particle energy. In our measurements three counters of known solid angle were used, namely (1) an approximately $0{\cdot}8\pi$ cesium-iodide scintillation detector, (2) a 2π proportional counter, and (3) a silicon surface-barrier detector with variable geometry, which can, however, be calibrated for any given source-to-detector distance.

EXPERIMENTAL

The $^{235}UO_2$ and $^{238}UO_2$ sources consisted of deposits that had been vacuum evaporated on platinum backings. The sources were prepared by Povelitis at the Los Alamos Scientific Laboratory, Los Alamos, New Mexico.[4] The deposits were 1·27 cm in diameter and varied in thickness from approximately 0·05 to 1·1 mg UO_2/cm². The isotopic compositions and specific activities of the $^{235}UO_2$ and $^{238}UO_2$ used were reported earlier.[5] The ^{148}Gd and ^{210}Po sources consisted of 3 mm diameter "weightless" (<1 μg/cm²) electroplated deposits on polished platinum backings. A U_3O_8 source consisting of a 1·8 cm diameter, 0·8 mg/cm² vacuum evaporated deposit of natural U_3O_8 on a platinum backing was counted in both the $0{\cdot}8\pi$ and 2π

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counters, and then a 1·6 cm diameter, ~4μg/cm^2 collodion film containing suspended ^{210}Po was placed on top of the U_3O_8 layer and the source recounted.

The disintegration rates, N_0, of the $^{235}UO_2$, ^{210}Po, and ^{210}Po on U_3O_8 sources were determined using the NBS 0·8$\pi\alpha$ counter. The detector in this counter is CsI(Tl) and is baffled to subtend a solid angle of 0·19748 × 4π steradians to the source. The baffles are so arranged that small changes in source position result in negligible changes in count rate.[6] Discrimination level plateaus of the 0·8$\pi\alpha$ counter had slopes of less than 0·1 % per 100 keV from 50 to 250 keV. Linear extrapolations to zero energy were made and the resulting count rates were corrected using the known geometrical factor to obtain the disintegration rates. For the $^{235}UO_2$ sources, the source thicknesses, d^*, in units of mass per unit area, were calculated from the following equation:

$$d^* = \frac{M(UO_2)}{M(^{235}U)} \cdot \frac{1}{A} \cdot \frac{1}{S} \cdot \frac{dN}{dt} . \qquad (1)$$

Here dN/dt is the disintegration rate, A is the source area, S is the specific activity (222·6 dps/mg ^{235}U for the 99·7 atom percent pure ^{235}U radioactivity in these deposits),[4] and $M(UO_2)$ and $M(^{235}U)$ are the molecular weights of UO_2 and ^{235}U, respectively.

The disintegration rate of the ^{148}Gd source was determined using a calibrated silicon surface-barrier detector. An electroplated ^{241}Am source, having the same diameter and backing material, was used as the transfer standard between the 0·8$\pi\alpha$ counter and the silicon surface-barrier detector. (The ^{148}Gd source could not be counted in the 0·8$\pi\alpha$ counter directly because the hydrogen gas used in this counter unduly attenuated the 3·18 MeV α-particles.)

Each source was also counted in the 6·0 cm radius, hemispherical, 2$\pi\alpha$ proportional counter using P–10 counting gas (90% argon, 10% methane) at atmospheric pressure.[7] The net count rates were linearly extrapolated to zero energy to obtain the values of the 2π counting rate, $C_{2\pi}$. Because the disintegration

Table 1. Experimental results

Source Number	Thickness (mg/cm^2)	d§	$C_{2\pi}/N_0$ Experimental	$C_{2\pi}/N_0$ Calculated From Part I
U235–1–1	0·0823	0·0080	0·5200 ± 0·0050	0·5158
U235–1–2	0·0850	0·0083	0·5188 ± 0·0036	0·5155
U235–2–4	0·1186	0·0115	0·5150 ± 0·0030	0·5142
U235–2–1	0·1234	0·0120	0·5146 ± 0·0022	0·5139
U235–2–3	0·1981	0·0192	0·5110 ± 0·0030	0·5108
U235–2–2	0·2218	0·0215	0·5034 ± 0·0033	0·5097
U235–5–2	0·4993	0·0485	0·4949 ± 0·0027	0·4978
U235–5–3	0·5360	0·0520	0·4979 ± 0·0021	0·4964
U235–5–1	0·5575	0·0541	0·4900 ± 0·0021	0·4954
U235–10–1	1·0911	0·1059	0·4753 ± 0·0019	0·4766
Gd 148–1 on Pt	0·001	~10^{-4}	0·5242 ± 0·0013	0·5241
Po 210–1 on Pt	0·001	~10^{-4}	0·5176 ± 0·0015	0·5176
U_3O_8–1	0·804		(†)	(†)
Po 210 on U_3O_8–1	0·004 (collodion)	~10^{-3}	0·5049 ± 0·0020	(‡)

§ Thickness expressed as a fraction of the range in the source material.

† Although the U_3O_8 source was counted in both the 0·8$\pi\alpha$ and the 2$\pi\alpha$ counters, the count rates obtained were used only as background measurements for the ^{210}Po on U_3O_8 determinations.

‡ Not calculated because σ is not known. However $C_{2\pi}/N_0$ agrees well with the value, 0·5060, obtained using σ for UO_2, which is expected because of the similarity in the chemical compositions of the two compounds.

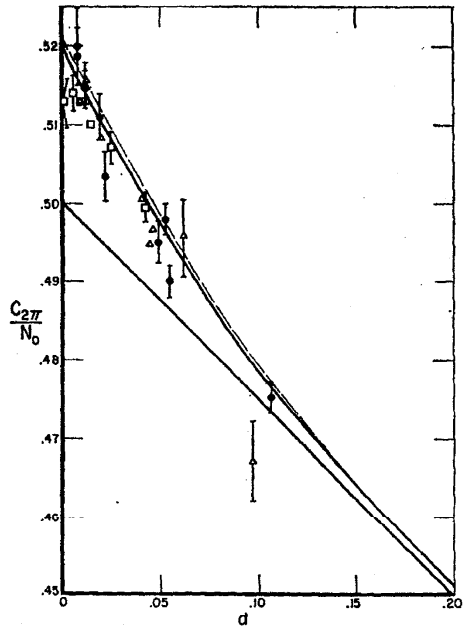

FIG. 1. Calculated and experimental values of $C_{2\pi}/N_0$ as a function of source thickness d, expressed as a fraction of the range in the radioactive material. The diagonal straight line corresponds to no scattering ($\sigma = 0$). The lower, heavy curve was calculated using equations (1) and (4) (Part I) with $b > 1$, $\sigma_b = 0{\cdot}107$, $\sigma_g = 0$, $\sigma_s = 0{\cdot}033$, and $t = 0$. These values correspond to the $^{235}UO_2$ and U_3O_8 sources. The upper, dashed curve was calculated with $\sigma_b = 0{\cdot}114$ and $\sigma_s = 0{\cdot}036$, corresponding to the $^{238}UO_2$ sources. The experimental points are from: (●), this work; (□), reference 2; (Δ), reference 3. (See Part I, this issue, for discussion of the parameters σ_b, σ_g and σ_s t is the thickness of a possible covering layer. $b > 1$ means that the backing thickness is greater than the range.) The difference between the experimental points and the straight line (corresponding to no scattering) is the scattering correction B. At $d = 0$, B is due entirely to scattering in the backing material, platinum. At other values of d, B is due to scattering in both the backing material and the source material.

rates of the $^{238}UO_2$ sources were too low to be measured accurately in a low-geometry counter such as the $0{\cdot}8\pi\alpha$ counter or the silicon surface-barrier detector, these sources were counted only in the lower background (~3 counts/min), higher-efficiency, $2\pi\alpha$ proportional counter. The disintegration rates for the $^{238}UO_2$ and U_3O_8 sources were calculated from the 2π counting rates, using the known specific activities and the scattering and absorption corrections obtained from the measurements on the $^{235}UO_2$ sources.

The results of the measurements for the $^{235}UO_2$, ^{210}Po, ^{148}Gd, and ^{210}Po on U_3O_8 sources are shown in Table 1. The error limits given are the sum of the random errors at the 68% confidence level and the estimated upper limit of the systematic errors.

The calculated values are in good agreement with the experimental values, and this suggests that the model discussed in Part I describes α-particle scattering within accuracies presently attainable experimentally.

These results are shown in the figure along with the theoretical curve calculated in Part I (this issue). Also shown are the results of GOLD and ARMANI[2], and WHITE[3] for U_3O_8 sources evaporated onto platinum backings.

Acknowledgements—We wish to thank J. A. GRUNDL and D. M. GILLIAM for making the UO_2 sources available to us.

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