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THE CROSS-SECTION OF THE REACTION U234 (d, 4n)Pu234

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ABSTRACT

The cross-section of the reaction $U^{234}(\alpha,4n)Pu^{234}$ has been determined by the recoil technique, using the reaction $Pu^{239}(\alpha,3n)Cm^{240}$ to monitor the beam and to determine the recoil efficiency. The maximum in the cross-section occurs at 42.6 Mev, where it is 0.98 ± 0.15 mb.

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1. The Recoil Technique.

The reaction studied was induced by bombardment of the target material at the Crocker Laboratory 60-inch cyclotron. The recoil technique used for the bombardments was similar in principle to that described by Harvey and co-workers.[1] The principal difference was that in these bombardments the external beam was used, thus considerably simplifying such problems as cooling and alignment with respect to the beam. The target assembly was designed by W. Wade and C. Corum for use in studies of the distribution of recoil fragments, but was easily adapted for use in determining cross-sections. The target assembly is shown in Fig. 1. The target was deposited as a very thin layer over a circular area 1.05 cm in diameter on 1-mil gold foil. The beam intensity, measured in the Faraday cup, varied from 2 to 6 microamps/cm^2. The beam was first passed through the gold foil and then through the target material, such that the reaction products were ejected from the target in the forward direction. They were caught on a 0.1-mil gold foil placed about 0.4 cm from the target foil. The energy of the helium-ions was varied by inserting aluminum degrading foils into the beam ahead of the target foil. The energy of the degraded beam was calculated from the range-energy data of Aron, Hoffman, and Williams.[2] The chamber containing the catcher foil was evacuated. Helium cooling was used between the degrading foils and the target foil.

The success of the recoil technique depends upon the ability of the heavy element recoiling nuclei to leave the target before being scattered or absorbed by the target material. Chetham-Strode[3] has measured the range of At^{211} recoils in bismuth resulting from the $Bi^{209}(\alpha,2n)At^{211}$ reaction. He found that for helium-ion energies of 20 to 40 Mev, high recoil-collection efficiency could be obtained if the thickness of the target deposit was less than about 30 $\mu g/cm^2$. In order to determine the recoil efficiency, the

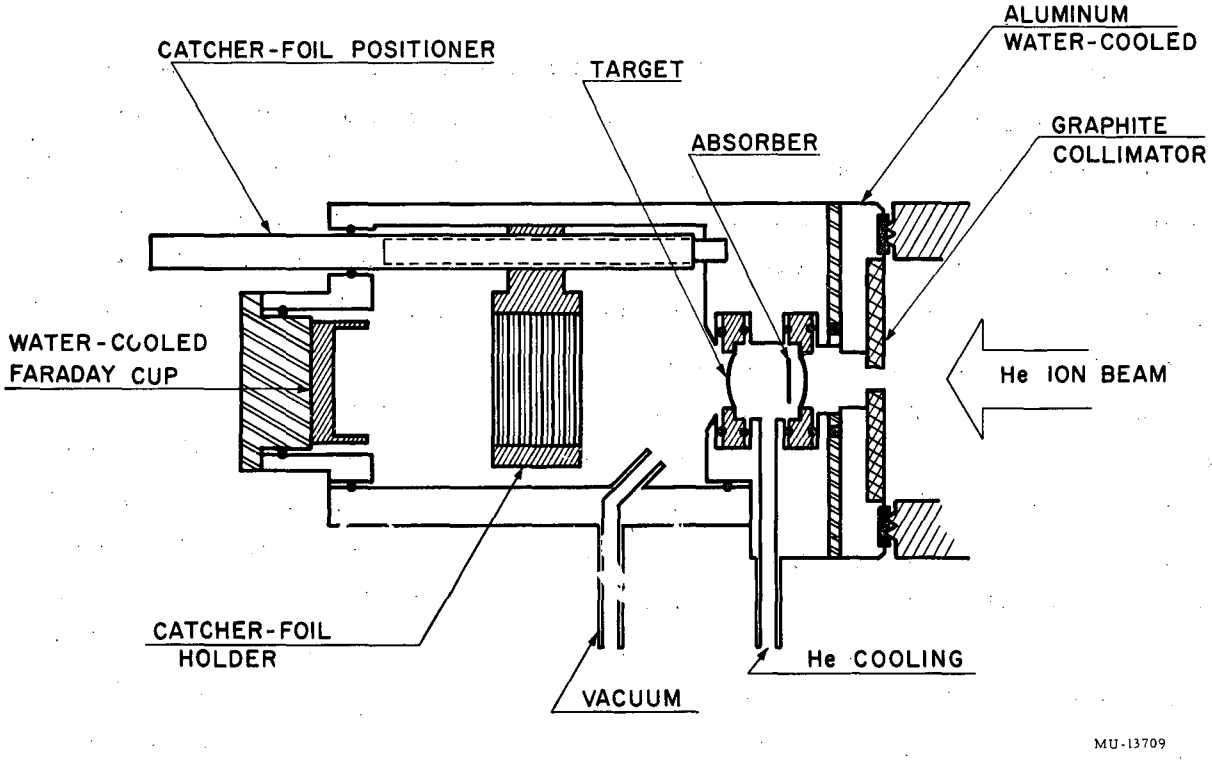

Fig. 1. The recoil block. The catcher foil holder is shown in retracted position. In the experiments described in this report it was moved up to 0.4 cm from the target.

$Pu^{239}(\alpha,3n)Cm^{240}$ reaction was used as a monitor. This reaction was chosen because the product is easy to separate chemically, and because it is believed that the $(\alpha,3n)$ and $(\alpha,4n)$ reactions proceed by similar mechanisms. A reaction proceeding by a different mechanism might lead to products that would have different recoil properties. The excitation function for the $Pu^{239}(\alpha,3n)Cm^{240}$ reaction has been reported by Glass and co-workers.[4]

2. Target Preparation.

The uranium used had an isotopic composition of 93.97% U^{234}, 4.33% U^{235}, and 1.70% U^{238}, as determined by mass spectroscopy. The target was prepared by electrodeposition of the uranium and plutonium onto a 1-mil gold foil, which served as the cathode, from a solution of 6 M ammonium chloride. The ammonium chloride buffer solution was adjusted to the methyl-red endpoint and the electrodeposition was made by plating at 6 volts with a current of 2 amps for 5 minutes. The target was found to contain 9.68 $\mu g/cm^2$ U^{234} and 1.93 $\mu g/cm^2$ Pu^{239}. This was determined by counting the radiations from the target in a low-geometry alpha counter and an alpha-pulse analyzer. In order to reduce the specific activity to levels which could be handled by the pulse analyzer, the target was covered with electromesh. The target was checked for uniformity by moving a thin foil with a small hole in it over the sample and alpha counting the exposed surface.

3. Chemical Separations.

The 0.1-mil gold catcher foil was dissolved in aqua regia containing Pu^{238} and Cm^{244} tracers. After dissolution, the solution was evaporated to dryness and the residue was taken up in a few drops of concentrated HCl and placed on an anion column packed with Dowex A-1 resin. The curium fraction, containing the Cm^{240} from the $Pu^{239}(\alpha,3n)$ reaction and the Cm^{244} tracer, passed through the column and was electrodeposited for counting. The plutonium (IV) and gold stuck to the column, which was washed with a few additional drops of concentrated HCl to remove the last traces of the curium. The plutonium (III) was then eluted with 1 N HCl and electrodeposited for counting. Because the only counting required was alpha pulse analysis, it was not necessary to have a high degree of purification from fission products.

4. Sample Preparation.

The fractions obtained from the column separation were evaporated down to a volume of approximately 1/2 ml, and buffered with a 6 M ammonium chloride solution before electrodeposition. The technique employed was the same as that used to prepare the target, with the exception that the samples were deposited on a 2-mil x 1-in. platinum disk. The disk was flamed to give a weightless sample suitable for alpha pulse analysis.

5. Counting.

The total disintegration rate of alpha-emitting samples and tracer solution assays was determined in a 52% geometry argon-flow ionization chamber. In order to differentiate between the alpha-emitting products and the tracers, it was necessary to use a 48-channel alpha-particle pulse-height analyzer. The instrument amplifies, electronically sorts, and records pulses of different energy produced in a methane-filled ionization chamber. The particular instrument used had a geometry of approximately 37% and was quite stable with respect to drift in energy scale.

Alpha pulse analysis was used to determine the amounts of alpha-emitting spallation products and to determine the chemical yield by comparison with the amount of activity from the tracers. The decay of the 9-hr Pu^{234} was followed and an alpha-branching of 6.16%[5] was used to calculate the total disintegration rate.

6. Calculation of Cross-Sections.

The formula defining the cross-section is:

$$N = In\sigma \qquad (1)$$

where N = number of atoms of the reaction product formed

n = number of target nuclei/cm^2

σ = cross-section in cm^2

I = number of incident bombarding particles.

Solving (1) for the cross-section:

$$\sigma = \frac{N}{In} \qquad (2)$$

The integrated beam is given in terms of microampere hours. Since one microampere hour is equivalent to 1.12×10^{16} helium-ions, (2) may be modified by replacing I by $q \times 1.12 \times 10^{16}$, where q is the integrated beam in microampere hours:

$$\sigma = \frac{N}{nq\ (1.12 \times 10^{16})} \qquad (3)$$

The number of atoms produced (N) is equal to the number of disintegrations per minute (d/m) divided by the decay constant (λ in minutes^{-1}) of the reaction product:

$$\sigma = \frac{d/m}{nq\ \lambda\ (1.12 \times 10^{16}} \qquad (4)$$

The alpha counting rate is measured in an alpha chamber having 52% geometry. Correcting for this, one obtains:

$$\sigma = \frac{c/m}{nq\ \lambda\ (.52 \times 1.12 \times 10^{16})} \qquad (5)$$

Where c/m is the number of counts per minute determined in the 52% geometry alpha counter.

Putting in numerical values for n and λ, one obtains the following equations:

$$\text{For } Pu^{239}(\alpha,3n)Cm^{240}, \quad \sigma = \frac{c/m\ Cm^{240}\ (1.97)}{q} \text{ in mb} \qquad (6a)$$

$$\text{For } U^{234}(\alpha,4n)Pu^{234}, \quad \sigma = \frac{c/m\ Pu^{234}\ (5.23 \times 10^{-3})}{q} \text{ in mb} \qquad (6b)$$

Equation (6b) must be modified by two corrections. First, equation (6b) assumes that all of the Pu^{234} decays only by alpha-emission. However, Hoff and Asaro[5] have found that it decays by alpha emission in only 6.16% of the decays. Secondly, it must be corrected for the recoil efficiency of the bombardment. Making these corrections, one obtains the equation used to calculate the cross section for the $U^{234}(\alpha,4n)Pu^{234}$ reaction:

$$\sigma = \frac{c/m\ Pu^{234}\ (5.23 \times 10^{-3})}{q \times B.R. \times R.E.}$$

where B.R. = branching ratio = .0616

R.E. = recoil efficiency.

The recoil efficiency was determined by dividing the apparent value for the cross section of the $(\alpha,3n)$ reaction on Pu^{239} (calculated using equation (6a), by the value found by Glass and co-workers[4] for the same reaction at the corresponding energy. An average recoil efficiency of 63.4% was obtained.

In determining the activity of Pu^{234} and Cm^{240} produced, it was necessary to correct for the decay which took place before the counting began and, in the first case, for decay during bombardment. The results of the calculations are given in Table 1, and a graph of the cross-section as a function of energy of the helium-ion is shown in Fig. 2.

Table 1

Energy of helium-ion (Mev)	σ $(\alpha,4n)$ on U^{234} (in mb)	Est. percent limit of error	Est. limit of error (in mb)
38.0	.30	± 25%	± .08
41.2	.63	10%	.06
41.8	.81	10%	.08
42.6	.98	15%	.15
43.5	.94	10%	.09
44.2	.83	10%	.08

7. Discussion.

The excitation function shows the shape characteristic of a compound nucleus reaction, as is to be expected for an $(\alpha,4n)$ reaction. The maximum cross-section gives a mean level width ratio (average neutron emission level width divided by the total de-excitation width for each of the four compound nuclei involved, Pu^{238}, Pu^{237}, Pu^{236}, and Pu^{235}, calculated by the method described by Glass, et al.[4]) of 0.17. The excitation function can be calculated to a good approximation through the use of Jackson's model,[6] as modified by Vandenbosch and co-workers,[7] by using 1.35 Mev as the nuclear temperature.

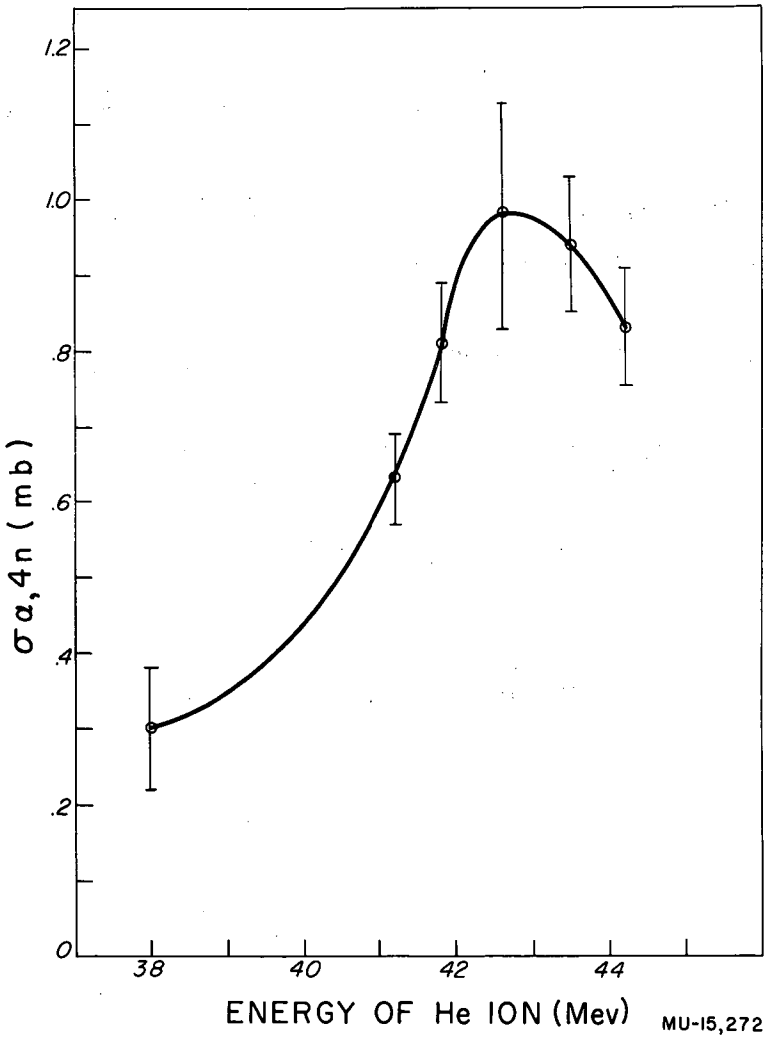

Fig. 2. Excitation function for the reaction $U^{234}(\alpha,4n)Pu^{234}$.

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