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CAPTURE CROSS SECTION OF Ac^{227} FOR THERMAL PILE NEUTRONS

by

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The neutron capture cross section of Ac^{227} has been determined as 500 ± 35 barns by H. Pomerance¹ using a pile oscillator technique, and 340 ± 75 barns by H. Kirby² by determining the ratio of Th^{228} to Ac^{227} using counting methods. Since the discrepancy between the two measurements is outside the experimental errors, another determination of the cross section seemed advisable. The cross section was redetermined by studying the following two series:

A sample of Ac^{227} , previously purified from its daughters, was irradiated in the Argonne heavy water pile (CP-3'), and several weeks after the irradiation a thorium fraction was chemically isolated from the bombarded actinium. The cross section of Ac^{227} was then determined from the ratio of Th^{228} , which is proportional to the amount of Ac^{228} formed in the bombardment, to Th^{227} , which is proportional to the amount of Ac^{227} originally present in the irradiation. The ratio $\text{Th}^{228}/\text{Th}^{227}$ was measured in a differential alpha pulse analyzer and the cross section was calculated from the following formula:

$$\sigma_{227} = \frac{\text{Th}^{228}}{\text{Th}^{227}} \frac{\lambda \text{Ac}^{227} (1 - e^{-\lambda \text{Th}^{227} t_2})}{\lambda \text{Th}^{227} n v t_1}$$

where t_1 = time of bombardment

t_2 = time of growth of Th^{227} since purification of the actinium.

The contribution to the cross section of neutrons above thermal energies was measured to see if there was any large resonance capture.

¹H. Pomerance, ORNL Progress Report ORNL-1415 (Sept. 1952).

²H. W. Kirby. (Private Communication).

Experimental - Three separate irradiations of actinium-227 were made in the central thimble of CP-3¹ for periods of two weeks. All of these samples were purified before irradiation in the manner of Hagemann³ and described as follows: 200-300 micrograms of an equilibrium solution of actinium-227 in 0.1 N HCl was contacted with 0.25 M thenoyltrifluoroacetone (TTA) in benzene. The RdAc (Th²²⁷) and AcC (Bi²¹¹) extracted into the benzene layer. The pH of the aqueous phase was then adjusted with NaOH to 5.5-6.0 and then contacted with 0.25 M TTA in benzene. The Ac²²⁷ extracted into the benzene layer and the AcX (Ra²²³) and AcK (Fr²²³) remained in the aqueous phase. The actinium-227 was reextracted from the benzene layer with 0.1 M HCl. Particular effort was made to remove all of the thorium-227 from the solution by making repeated contacts of the final 0.1 M HCl solution of actinium-227 with 0.25 M TTA in benzene. The time of the last contact to remove thorium-227 was noted as t_1 in equation (1). The actinium-227 was then evaporated to dryness in a quartz tube and the tube was sealed. To assure against breakage and the escape of actinon gas this tube was then sealed into another quartz tube. A neptunium-237 monitor was placed as close to the sample as possible.

In order to correct for the resonance contributions to the neutron captures in actinium-227, a cadmium shielded sample was irradiated in the second bombardment along with an unshielded sample. The actinium sample was chemically purified as described above, sealed into a quartz capsule, and placed in a small, 1/16-inch thick cadmium tube. This sample was placed three inches away from the unshielded sample so that no effect of the cadmium on the unshielded sample would be encountered.

After bombardment the actinium and its decay products were dissolved in 0.1 N HCl and the thorium isotopes extracted with 0.25 M TTA in benzene. The benzene solution was washed seven times with 0.1 N HCl to reextract any actinium which may have carried into the benzene layer. As soon as possible after the thorium separation a pulse analysis was run to determine the ratio of thorium-228 to thorium-227. The benzene layer containing the thorium isotopes was periodically purified of the decay products of the thorium and a sample was pulse analyzed. This was essential as a check on the purity of separation from the actinium and to obtain better accuracy on the ratios of the peak heights as the 18.6 da. thorium-227 decayed. A typical set of pulse analysis curves is shown in Figure 1.

The possibility of the original actinium-227 solution containing thorium-228 was checked by pulse analyzing a thorium fraction extracted from the actinium before bombardment. Pulse analyses have been made as the thorium-227 decays and after 18 half-lives no thorium-228 has been detected.

³F. Hagemann. Chap. 2 Nuclear Energy Series Volume 14A, The Actinide Elements.

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The neptunium-237 monitors were prepared by depositing a nitric acid solution of neptunium-237 on both platinum and aluminum plates using tetraethylene glycol as a spreading agent. To check the effect of epithermal neutrons on the neptunium-237, cadmium shielded samples were also irradiated. These were wrapped in 1/16-inch cadmium and placed three inches from the actinium sample in bombardment 3. In this same bombardment four unshielded monitors were spaced along the actinium capsule as a check on the uniformity of the flux.

Results - The ratio of thorium-228 to thorium-227 was measured by pulse analysis at varying times after the separation of the thorium from the actinium. This gave a check on the cross section and on the completeness of separation from the actinium. The results of these analyses are shown in Table I. The first pulse analysis following the thorium separation from the actinium has been neglected in the averaging of the cross section values in bombardments 1 and 3. Inspection of these two pulse analyses shows the accuracy of the values to be poor due to the large size of the thorium-227 peaks in comparison to the small size of the thorium-228 peak. The final cross section for capture of thermal neutrons becomes 516 ± 50 barns.

Table I

UNIFORMITY OF FLUX ALONG CAPSULE IN
BOMBARDMENT 3

Np ²³⁷ Monitors	A Results by counting	B Results by pulse analysis
A	3.62×10^{12}	3.51×10^{12}
B	4.41×10^{12}	4.34×10^{12}
C	3.50×10^{12}	3.55×10^{12}
D	3.87×10^{12}	3.97×10^{12}
Average	3.85×10^{12}	3.84×10^{12}

The resonance contributions to the neutron captures in actinium-227 were measured by using a cadmium shielded sample of actinium. It was found that 1.6% of the thorium-228 activity was due to the capture of neutrons whose energy was above 0.3 e.v. The cross section of the actinium-227 as shown above has been corrected for this fast neutron effect.

The principal cause of uncertainty in the measurements is the flux determination, for the measurement of the neptunium-237 cross section⁴ has not been carried out to an accuracy of better than 15 percent. The variation of the flux along the capsule as determined in bombardment 3 is shown in Table II. The results as found by counting are shown in column A and those found by pulse analysis of the same monitor plates in column B. Sample D corresponds to the position of the bulk of the actinium-227. The spacing of the plates were 0.75 inches center to center of each plate. It may be noticed that a variation in flux of ten to twenty percent over the distance of 0.75 inch was found. The average value of the four readings was used as the flux value for the bombardment 3. All of the flux values used have been corrected for the epithermal neutron contribution. The ratio of plutonium-238 to neptunium-237 was found to be 0.62, in the cadmium shielded sample and this value was used in correcting all of the flux values.

Table II

RESULTS OF CROSS SECTION VALUES AS FOUND IN
BOMBARDMENTS 1, 2, AND 3

Bombardment	Flux	Number of days after thorium separation from actinium	$\sigma_{Ac^{227}}$
1	3.99×10^{12}	1	692* $\pm$ 299
		24	595 $\pm$ 111
		46	539 $\pm$ 94
		76	535 $\pm$ 92
		124	542 $\pm$ 94
		Av.	553 $\pm$ 98
2	3.83×10^{12}	5	548 $\pm$ 87
		35	505 $\pm$ 78
		53	493 $\pm$ 80
		76	499 $\pm$ 79
		Av.	511 $\pm$ 81
3	3.84×10^{12}	1	706* $\pm$ 260
		21	495 $\pm$ 95
		65	488 $\pm$ 76
		121	527 $\pm$ 83
		154	531 $\pm$ 84
		Av.	510 $\pm$ 85

*Cross section not used in average value

⁴A. H. Jaffey. ANL-4030.

6

FIGURE 1
TYPICAL PULSE ANALYSIS OF THORIUM FRACTION SHOWING CHANGING $\text{Th}^{228}/\text{Th}^{227}$
RATIO AS 18.6 DAY Th^{227} DECAYS.