

of $112 \Omega^{-1}\text{m}^{-1}$), the concentration $1.2 \times 10^{26} \text{ m}^{-3}$ is calculated, which is in good agreement with this Hall value. For other samples a similar agreement was obtained.

The very high conductivity of the reduced ceramics, and the fact that the Hall measurement indicates that all of the electrons are free to contribute to conduction at room temperature, show that, in strongly reduced TiO_2 samples, the activation energy has become very low in agreement with earlier data obtained during the war in this Laboratory.⁶

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Thermal Neutron Capture Cross Section of A^{40} and Observation of $\text{A}^{42}\dagger$

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The thermal neutron absorption cross section of A^{40} was determined by measuring in calibrated proportional counters the amount of 110-min A^{41} produced from samples of argon irradiated in a neutron flux whose magnitude was measured by β - γ coincidence counting of gold monitors. The result was 0.53 ± 0.03 barn.

A^{42} was produced from A^{40} by two successive neutron captures and it was detected through its 12.5-hr K^{42} daughter. A lower limit of 3.5 years was set on its half-life. From this result the thermal neutron absorption cross section of A^{41} is calculated to be greater than 0.06 barn.

It was also shown that the thermal neutron absorption cross section of A^{38} is 0.8 ± 0.2 barn.

INTRODUCTION

THE thermal neutron cross section of A^{40} for the formation of 110-min A^{41} was measured by Kern¹ and by Wattenberg and West.² They found values of 1.24 barns and 0.72 barn, respectively. In both cases the activity was measured by placing the argon in a cell external to a Geiger counter. At about the same time the total neutron absorption cross section of argon was measured by the "danger coefficient" method³ using cadmium wire of known length and

diameter as standard. This value, which also measures an upper limit for the activation cross section of A^{40} , was 0.62 barn. In a similar experiment Colmer and Littler⁴ used the pile oscillator at Harwell and obtained the same value, 0.62 ± 0.04 barn. The total thermal neutron cross section (absorption plus scattering) for argon was first determined by Carroll⁵ from transmission measurements. He used a Rn-Be in paraffin source and an argon sample which transmitted 92 percent of the neutron beam. His value was 2.5 barns. Melkonian *et al.*,⁶ employed the Columbia University slow neutron velocity spectrometer and a sample of argon which transmitted 79 percent of the beam at 0.025 ev. Their result for the total cross section was 1.4 barns. If one subtracts from this the thermal neutron scattering cross section of 0.80 ± 0.08 barn determined by Harris⁷ one obtains 0.6 barn for the absorption cross section, in good agreement with the direct measurements.^{3,4}

In the present work the thermal neutron activation cross section of A^{40} was determined by measuring the A^{41} activity within a proportional counter and measuring the neutron flux by coincidence counting of gold

TABLE I. Activity per cc as function of counter length.

Length of cathode (cm)	Total counter volume (cc)	Cathode volume (cc)	Activity at a given time (counts/min)	Activity per cc of cathode volume (counts/min)
7.0	31.8	19.0	3460	182
17.0	59.6	47.1	8400	178
29.7	98.5	85.0	15,700	185
29.8	101.6	87.6	16,000	183
45.2	137.7	125.7	23,700	189
75.8	222.2	211.0	38,600	183
Average				183 ± 4

[†] Research carried out under the auspices of the AEC.

¹ B. D. Kern, unpublished Plutonium Project Report CP-772 (1943).

² A. Wattenberg and J. West, unpublished Plutonium Project Report CP-781 (1943).

³ Lichtenberger, Fowler, and Wattenberg, unpublished Plutonium Project Report CP-781 (1943).

⁴ F. C. W. Colmer and D. J. Littler, Proc. Phys. Soc. (London) **A63**, 1175 (1950).

⁵ H. Carroll, Phys. Rev. **60**, 702 (1941).

⁶ Melkonian, Rainwater, Havens, and Dunning, Phys. Rev. **73**, 1399 (1948).

⁷ S. P. Harris, Phys. Rev. **80**, 20 (1950).

monitors. The result is 0.53 ± 0.03 barn, considerably lower than previous direct determinations,^{1,2} but consistent with the results of references 3 and 4, and of 6 combined with 7.

MEASUREMENT OF A⁴⁰ CROSS SECTION

Small quartz ampules of measured volume (2 cc) were filled with pure argon at a carefully measured temperature (near 23°C) and pressure (near 500 mm Hg). These were irradiated in the Brookhaven pile for one minute with 10 mg gold monitors. Then each ampule was attached to a calibrated vacuum system and the irradiated argon was allowed to expand through a break-off seal into one or more glass bulbs. After allowing sufficient time for the pressure to equalize, additional carrier argon was added to one of these calibrated bulbs. It was mixed thoroughly with the irradiated argon by several cycles of cooling with liquid nitrogen and heating with a flame. When this gas was again at room temperature an aliquot was removed into a cylindrical proportional counter whose

TABLE II. Activity per cc as function of counter diameter.

Diameter of cathode (cm)	Counter gas cm argon	Operating voltage	Total volume (cc)	Cathode volume (cc)	Activity at a given time (counts/min)	Activity per cc of cathode volume (counts/min)
1.9	0.6	76	3800	98.5	85.0	1.16
1.9	75	10	2300	101.6	87.6	1.30
2.7	75	10	2500	205	164	1.34
4.9	75	10	3050	667	544	1.33
6.8	75	10	3500	1250	1044	1.32
8.7	75	10	3600	1990	1681	1.32
Average						1.33 ± 0.02

total volume and cathode volume had been measured previously. Methane was added to bring the total pressure up to one atmosphere.

The counting, which was done with proportional counters, was usually begun two hours after the irradiation and continued for about 3 half-lives. Coincidence loss corrections were negligible. When the background was subtracted, the decay was exponential with a half-life of 110 ± 1 minutes for A⁴¹, in good agreement with previous determinations. Two sets of experiments were done which indicated that the counting rate was actually equal to the disintegration rate of the A⁴¹ contained within the volume bounded by the counter cathode. In one series of experiments, the counter length was varied, and in the other series, the diameter was varied. The volumes were measured by passing in toluene from a buret. In each experiment, all the counters were opened to one manifold and filled with the irradiated argon at the same time. The data and results are shown in Tables I and II. All the counters of Table I have a diameter of 1.9 cm, a pressure of 16 cm of argon and 58 cm of methane, and they were

TABLE III. Thermal neutron absorption cross section of A⁴⁰.

No. of run	Thermal neutron flux (n/cm ² sec)	Number of irradiated argon atoms in cathode volume	Activity at end of irradiation (disint/sec)	Thermal cross section of A ⁴⁰ (cm ²)
1	4.92×10^{12}	15.12×10^{16}	2409	0.514×10^{-24}
2	4.42	6.36	893	0.505
3	4.19	0.95	141	0.562
4	3.20	3.07	322	0.520
5	3.90	14.63	1908	0.531
6	3.76	5.21	674	0.546
7	3.84	4.70	614	0.540
Average				0.531 ± 0.020

operated at 3600 volts (middle of 200-300 volt plateau); the counters of Table II are 30 cm long and they were filled with argon and methane as indicated in columns 2 and 3. It is seen from these results that the specific counting rate (activity per unit of cathode volume) for the 1.2-Mev β -rays of A⁴¹ is independent of counter length or counter diameter, within the experimental error of 2 percent. Bernstein and Ballentine⁸ found similar results for the 0.155-Mev β -rays of C¹⁴. Apparently the sensitive length of the counter is shorter than the cathode by a small amount which just compensates for the β -rays which enter the sensitive volume from the ends of the counter. This amounts to about one cm for the standard counters whose diameter is 19 mm.

The neutron flux in each irradiation was measured by making a determination of the absolute disintegration rate of the gold monitor by means of β - γ coincidence counting. The detectors were crystals of anthracene and of thallium activated NaI mounted on No. 5819 photomultiplier tubes. The random coincidence rate was measured by introducing a 1.3 μ sec delay which is long compared to the 0.3 μ sec resolving time. This was subtracted from the total coincidence rate to obtain the true rate. The counts recorded by the β -ray counter were reduced by 3 percent to correct for the γ -rays it records. Another small correction was applied for the effect of the 411-kev γ -rays which are converted 3 percent in the K shell and one percent in the L shell, the conversion electrons raising the counting rate in the β -ray counter by 4 percent. The γ -ray counter was covered with 600 mg/cm² of aluminum to remove the β -rays and the discriminator was set so that 2 percent of the counts were from the K x-rays. The disintegration rate A of the gold monitor was then determined from the formula, $A = C_{\beta}C_{\gamma}/1.02C_{\beta-\gamma}$, where C_{β} is the counting rate in the β -counter corrected for γ -rays, C_{γ} is the counting rate in the γ -counter, and $C_{\beta-\gamma}$ is the true coincidence rate. In one run the disintegration rate was also measured by comparing a small aliquot of the gold with a *RaE* standard by means

⁸ W. Bernstein and R. Ballentine, Rev. Sci. Instr. **21**, 158 (1950).

of an end-window Geiger counter.⁹ The results from both methods agreed within 4 percent.

By irradiating gold monitors in the same position in the pile both with and without cadmium shielding, it was found that 70 ± 2 percent of the Au^{198} is produced by thermal neutrons. The thermal neutron flux was calculated using 0.70 of the observed Au^{198} disintegration rate, 64.5 hr for the Au^{198} half-life, and 95 barns for the Au^{197} cross section. No correction was made for the small branches to the 1.09-Mev level and to the ground state because these are completely negligible for the purpose of the present calculation. The flux for each run is listed in Table III. In one case, the flux was also determined from the amount of a fission product produced in a uranium monitor. A quantitative radiochemical analysis was done for 12.8-day Ba^{140} . Its activity was measured with an end-window Geiger tube whose efficiency was measured with a uranium "geometry" standard. The usual corrections were applied and the flux calculated from the Ba^{140} disintegration rate, the fission yield, the fission cross section of uranium, the weight of the monitor, and the irradiation time. The result was within 5 percent of that derived from the coincidence measurements on the gold monitor.

The observed A^{41} disintegration rate was decreased by 3 percent to correct for the activation produced by neutrons above the cadmium cutoff. This was determined from two special runs. The thermal neutron activation cross section of A^{40} calculated from each run is given in Table III. The average is 0.53 ± 0.02 barn. The error indicated is the standard deviation for the seven values given in the table. In addition, the following uncertainties must be considered: 3 percent in the fraction of the Au^{198} activity that is produced by neutrons below the cadmium cutoff; 2 percent in the gold cross section; and one percent in the A^{41} half-life. The over-all standard deviation then becomes 6 percent and the value of the cross section is taken as 0.53 ± 0.03 barn. The purity of the argon was kindly checked with a mass spectrometer by Dr. O. A. Schaeffer. About 0.7 percent of foreign gas (principally nitrogen) was found and the necessary correction was made.

A^{39}

In order to compare the above result with the neutron absorption cross section of natural argon as reported in the literature,^{3,4} the other two stable isotopes must be considered. A^{36} , whose abundance is 0.337 percent, has an isotopic cross section of 6 barns,¹⁰ thus contributing 0.02 barn to the total absorption. The sum for A^{36} and A^{40} (99.600 percent abundance) is 0.55 ± 0.03 barn. Subtracting this from 0.62 ± 0.04 , the value of

Colmer and Littler⁴ for the total absorption cross section, one obtains 0.07 ± 0.05 barn for the contribution of A^{38} . The upper value, 0.12 barn, specifies an upper limit of 190 barns for the isotopic absorption cross section of A^{38} whose natural abundance is only 0.063 percent.

A direct determination of this cross section by the activation method is not feasible because only a lower limit has been established¹¹ for the half-life of A^{39} (>15 years). Nevertheless, some A^{39} was prepared by irradiating 1.56 liters of argon in a quartz tube near the center of the Brookhaven pile for 12 days in a flux of 4×10^{12} neutrons/cm²/sec. A small aliquot was put into a cell with a mica window (5 mg/cm²) and counted with an end-window G-M tube about one year after the irradiation. The very soft radiations of 34-day A^{37} were completely absorbed by the windows. The A^{39} was identified by means of the characteristic absorption curve of its 0.565-Mev β -rays in aluminum. The total activity corrected for backscattering, absorption, aliquot, and counter efficiency was 7600 disintegrations/sec. By combining this value with the minimum half-life of 15 years, a lower limit of 0.05 barn can be placed on the cross section of A^{38} . By using the upper limit of 190 barns (calculated in the previous paragraph) an upper limit of 6×10^4 years can be set for the A^{39} half-life.

Note added in proof:—Very recently H. Zeldes *et al.*, Phys. Rev. **86**, 811 (1952), reported 265 ± 30 years for the A^{39} half-life. Based on this value and the data given above, the thermal neutron absorption cross section of A^{38} is 0.8 ± 0.2 barn.

A^{42}

Most of the 1.56-liter argon sample that had been irradiated for 12 days was used to investigate A^{42} which can be formed from A^{40} by two consecutive neutron captures. No attempt was made to detect the A^{42} radiations directly because of the greatly preponderant activity of A^{39} . Rather, the A^{42} was detected by successive extractions of its 12.5-hr K^{42} daughter. The first extraction was made two days after the end of the irradiation; growth times were varied from 4 hours to 131 days. Initial counting rates were about 12 counts/min above the background and the natural K^{40} activity. The decay curves corresponded to the known K^{42} half-life, 12.5 hours. In 13 extractions over a period of 400 days, the corrected activity of A^{42} showed no apparent decrease; consideration of the possible errors indicates that it could not have gone down by more than 20 percent. This sets a lower limit of 3.5 years on the half-life of A^{42} and a lower limit of 0.06 barn on the thermal neutron absorption cross section of 110-min A^{41} .

⁹ B. P. Burt, Nucleonics **5**, 28 (1949).

¹⁰ G. E. McMurtrie and D. P. Crawford, Phys. Rev. **77**, 840 (1950).

¹¹ Brosi, Zeldes, and Ketelle, Phys. Rev. **79**, 902 (1950).