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Neutron Absorption Cross Sections of Radioactive La^{140} and Two Stable Cerium Isotopes*

S. KATCOFF,** J. A. LEARY, K. A. WALSH, R. A. ELMER, S. S. GOLDSMITH, L. D. HALL, E. G. NEWBURY, J. J. POVELITES, AND J. S. WADDELL
Los Alamos Scientific Laboratory, University of California, Los Alamos, New Mexico
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Two multi-curie samples of 40-hr. La^{140} were irradiated with slow neutrons at the Los Alamos homogeneous pile for 24 hours. The small amount of 3.7-hr. La^{141} formed was allowed to decay to 28-day Ce^{141} which was then radio-chemically extracted and counted. From the activity measured, the neutron flux, and the known half-lives, a value of 3.1 ± 1.0 barns was calculated for the cross section of 40-hr. La^{140}. The thermal neutron absorption cross sections of stable Ce^{140} and Ce^{142} were also measured and found to be 0.27 ± 0.06 and 0.105 ± 0.020 barn, respectively, per atom of the naturally occurring element.

INTRODUCTION

THE activation method, which has been used extensively for the determination of cross sections of stable nuclei,[1] has also been applied in several cases to the measurement of neutron absorption cross sections of radioactive isotopes.[2] One method of attack is to irradiate in a high neutron flux a stable isotope, S, which by a single neutron capture gives rise to active isotope J, whose cross section is to be measured. Further capture of neutrons by J yields radioactive isotope B which frequently has an active daughter, C:

$$\text{stable } S \xrightarrow{(n,\gamma)} \text{active } J \xrightarrow{(n,\gamma)} \text{active } B$$
$$\Big\downarrow \beta^- \qquad\qquad \Big\downarrow \beta^-$$
$$K \qquad \xrightarrow{(n,\gamma)} \text{active } C.$$

The saturation number of atoms of J is given by

$$N_J = N_S(nv)\sigma_S\left(\frac{1}{\lambda_J}\right), \tag{1}$$

where N_S is the number of S atoms, σ_S is the thermal neutron activation cross section of S, λ_J is the disintegration constant of J, and (nv) is the neutron flux. If the half-life of J is short compared to the time of bombardment and also to the half-life of B, then the activity of B at the end of bombardment time, t, is given by

$$A_B = (1/\lambda_J)N_S(nv)^2\sigma_S\sigma_J(1-\exp(-\lambda_B t)). \tag{2}$$

By measuring A_B, (nv), N_S, and t, the cross section of J, σ_J, can be calculated from this equation. The cross section σ_S is known or can easily be measured. In cases where the time of irradiation is not long compared to the half-life of J, the above equations must be replaced by more general ones.[2] It should be noted that the activity of B is proportional to the square of the neutron flux. Since it is usually more advantageous to analyze radiochemically for C rather than for its parent B, account must be taken of the amount of C which is formed by the alternate process involving neutron capture by K, the daughter of J[2]. However, in the favorable cases where the half-life of C is short compared to that of its parent B, this alternate process does not interfere because C can be separated radiochemically from B at periodic intervals. Only the first extraction can contain C which originated from neutron capture by K. All succeeding extracts can contain only that C activity which comes from β-decay of its parent B.

By applying the above method, upper limits have been set on the cross sections of a few fission products,[2,3] and the thermal neutron capture cross section of 85-min. Ba^{139} was determined to be 3.8 ± 1.0 barns.[4]

A slightly different method for measuring cross sections of unstable nuclei involves the isolation of as much radioactive J as possible from its source of supply and then irradiating it in a high neutron flux. The product of neutron capture, B (or its daughter C), can be measured as above. In this case the activity of B at the end of bombardment time t is given by[2]

$$A_B = (N_J{}^0 T_J\sigma_J(nv)/T_J - T_B) \times(\exp(-\lambda_J t)-\exp(-\lambda_B t)), \tag{3}$$

where $N_J{}^0$ is the number of atoms of J present at the beginning of the irradiation, and T_J and T_B are the half-lives of J and B, respectively. The activity of C arising from neutron capture by stable K (the daughter of J) is given by the following equation:

$$A_C = (N_J{}^0\sigma_K(nv)/T_J - T_C)[T_J(1-\exp(-\lambda_J t)) - T_C(1-\exp(-\lambda_C t))], \tag{4}$$

* Based on AEC Report LA-630 (May 26, 1947).
** Now at Brookhaven National Laboratory, Upton, Long Island, New York.
[1] L. Seren, H. N. Friedlander, and S. H. Turkel, Phys. Rev. 72, 888 (1947).
[2] S. Katcoff, Plutonium Project Record, IX B, 7.59.1 (1946).
[3] N. Sugarman and A. Turkevich, Manhattan Project Report CC-2485 (Dec. 15, 1944); Plutonium Project Record, IXB, 7.59.2 (1946).
[4] S. Katcoff, Manhattan Project Report CC-2908 (April 7, 1945); Plutonium Project Record IXB, 7.59.4 (1946).

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where σ_K is the neutron absorption cross section of K. Equation (3) can be used to calculate the cross section of J. However, when the activity of C is measured instead of the activity of B and when the half-life of C is long compared to that of its parent B, then the activity of C, which is formed by neutron activation of K, must be subtracted from the total activity of C. This amount is calculated by use of Eq. (4).

This method has been applied in work with some of the transuranic isotopes, in a rough determination[5] of the cross section of long-lived I^{129}, and in measuring the cross section of 40-hr. La^{140} as reported here. The nuclear processes involved in the latter were:

$$\begin{array}{ccc} \text{40-hr. } La^{140} & \xrightarrow{(n,\gamma)} & \text{3.7-hr. } La^{141} \\ \downarrow \beta^- & & \downarrow \beta^- \\ \text{stable } Ce^{140} & \xrightarrow{(n,\gamma)} & \text{28-day } Ce^{141}. \end{array}$$

A quantity of 40-hr. La^{140}, corresponding to a number of curies, was irradiated at the Los Alamos homogeneous pile for a period of 24 hours. Several days later radiochemical analysis was made for the 28-day Ce^{141} resulting from the irradiation.

EXPERIMENTAL PROCEDURE AND RESULTS

Before starting the major portion of the experiment, a determination was made of the neutron absorption cross sections of the two major stable cerium isotopes, Ce^{140} (88.5 percent abundance) and Ce^{142} (11.1 percent abundance). The former had not been measured previously, and the latter had been measured only roughly.[6] For this determination 2.0 g of Ce, as $Ce(NO_3)_3 \cdot 6H_2O$, was irradiated for one hour at a point in the graphite 8″ from the edge of the "water boiler" sphere. A piece of uranium was used as a monitor for the neutron flux. Three weeks of cooling was allowed for the 33-hr. Ce^{143} to decay to a negligible value. Then the cerium was dissolved, and aliquots were withdrawn for radiochemical analyses. Two samples were analyzed for 28-day Ce^{141} and two others for 13.8-day Pr^{143}, the daughter of 33-hr. Ce^{143}. The decay of the samples was followed for several weeks by means of a mica-window bell-shaped Geiger counter. Aluminum absorption curves were also taken of the activities in order to check their purity and also for making corrections to zero absorber. Correction was also made to 100 percent counter geometry (by means of a standard), to 100 percent chemical yield, and for decay. The neutron flux was measured by analyzing the uranium monitor for the 12.8-day Ba^{140} fission product. From the latter's fission yield and the thermal fission cross section of uranium the flux was calculated. Then by applying the parent daughter relation and Eq. (1), the neutron absorption cross sections which are given in Table I were calculated. These values are for thermal neutrons if the fission cross section of uranium and the cross sections measured here change by the same ratio when a purely thermal neutron flux is substituted for the approximately thermal flux actually used. This assumption is probably valid within the limits of error given. The value listed here for Ce^{142} checks the tentative value of 0.1 barn reported earlier[6] for the natural atom cross section.

TABLE I. Neutron capture cross sections of stable Ce.

	Isotopic abundance	Natural atom cross section	Isotopic cross section
Ce^{140}	88.5 percent	0.27 ±0.06 barn	0.31±0.07 barn
Ce^{142}	11.1 percent	0.105±0.020	0.95±0.18

Two runs were made to determine the cross section of 40-hr. La^{140}. In the first one, a solution of 12.8-day Ba^{140} was used which had been milked of its 40-hr. La daughter several times before. After the last milking four days were allowed for more 40-hr. La to grow in. Then 10 mg of stable La carrier was added and precipitated with NaOH as $La(OH)_3$. This was filtered, redissolved, and precipitated twice more as $La(OH)_3$ in order to purify it from 12.8-day Ba. Then a final precipitation was made as lanthanum "fluo-oxalate."[7] These operations were performed by means of remote control apparatus which was made available for this work by Dr. R. W. Spence. Most of the radioactive La^{140} was contained in a stainless steel container of 0.020-in. wall thickness. It was transported to the pile in a thick lead shield and then transferred quickly by means of two 6-foot rods and a string to a position in the graphite 9″ from the "water boiler" sphere. Two small uranium foils were used as neutron flux monitors. The sample was irradiated for 24 hours at maximum pile power. However, a portion of this La^{140} sample was not bombarded but reserved for comparison with the irradiated sample.

After a two-week cooling period, both samples were radiochemically analyzed, in duplicate, for 28-day Ce^{141}, 40-hr. La^{140}, and 12.8-day Ba^{140}. The La^{140} analyses served as assays for the size of the samples. In the Ce analyses it was necessary to reprecipitate $Ce(IO_3)_4$ eight times to decontaminate completely from the vastly greater activity of 40-hr. La. Aluminum absorption curves of the radiations from the Ce samples indicated the presence of both 28-day Ce^{141} and 275-day Ce^{144}. It became obvious that these isotopes were present as small but im-

[5] S. Katcoff, Phys. Rev. **71**, 826 (1947).
[6] S. Katcoff, Manhattan Project Report CC-2739 (February 23, 1945).
[7] G. Friedlander, R. Spence, and R. G. Steinhardt, Jr., Manhattan Project Report LA-557 (May 6, 1946).

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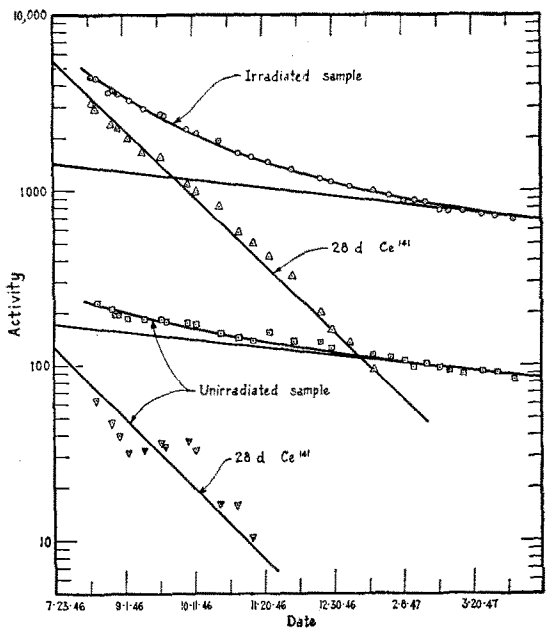

FIG. 1. Decay of Ce activity extracted from irradiated and unirradiated 40-hr. La^{140}.

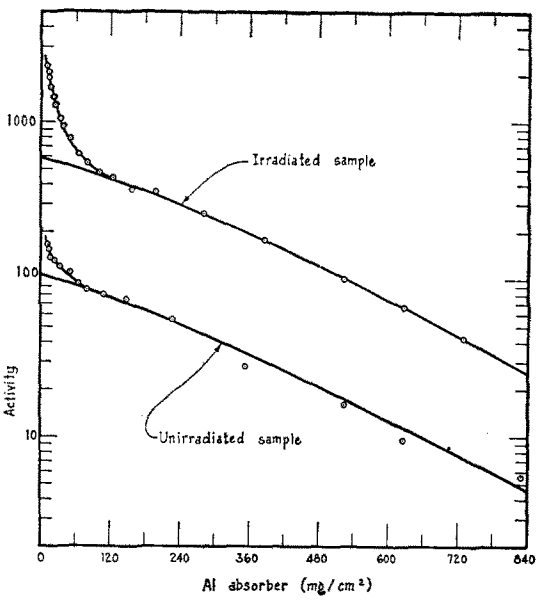

FIG. 2. Aluminum absorption curves of Ce activity from irradiated and unirradiated 40-hr. La^{140}.

portant impurities in the original active La^{140} samples. A small increase in the 28-day Ce^{141}, however, was found in the irradiated sample. This was used to calculate a rough upper limit of about 2 barns on the neutron absorption cross section of 40-hr. La^{140}. The barium analyses indicated that an appreciable quantity of 12.8-day Ba^{140} was also present. This isotope can also absorb neutrons to give 18-min. Ba^{141} which decays to 3.7-hr La^{141} and then to 28-day Ce^{141}. Therefore the Ba^{140} had not been removed sufficiently in this run.

In the final run an attempt was made to overcome the above difficulties. A Ba^{140} solution similar to the first one was scavenged three additional times by precipitating $Fe(OH)_3$ four days before finally extracting the La^{140} sample. These precipitations were designed to remove any radioactive Ce that may have been present. For the final La^{140} extraction 10 mg of stable lanthanum carrier was used again, but this time the two $La(OH)_3$ reprecipitations were done in the presence of about 500 mg of stable Ba carrier. This was designed to reduce further the amount of 12.8-day Ba^{140} carried along. Subsequent operations were essentially identical with those of the previous run. The radio-chemical analyses for La^{140} showed that 22.6 curies of this isotope were present at the beginning of the neutron irradiation.

The radio-chemical analyses for active Ce in both the irradiated and unirradiated samples were performed in duplicate on two aliquots (0.400 of total) from each sample. A considerable improvement over the previous run was observed. Although active Ce was not completely eliminated from the La^{140} samples before irradiation, a large increase in the amount of 28-day Ce^{141} was observed in the irradiated sample. This is seen from the decay curves and aluminum absorption curves shown in Figs. 1 and 2. The soft part of the radiation is from 28-day Ce^{141} and 275-day Ce^{144}; the hard beta-ray is from 17.5-min. Pr^{144}, daughter product of 275-day Ce. In Table II the data are presented. The time of irradiation was 24.0 hours, starting 7.9 hours after the La^{140} was separated from the Ba^{140}.

The data in both the second and third columns represent an average of two aliquot samples; column 4 represents an average of four aliquots, and the last column an average of three aliquots. The data of column 3 are based on the very poor decay curve shown at the bottom of Fig. 1, but fortunately these data affect the final result to only a small extent. The activity of 28-day Ce^{141} which was formed by the neutron irradiation of the La sample was (968−221) or 747 disintegrations per second. From this must be subtracted 115 d/s, which is the calculated activity of 28-day Ce^{141} formed by neutron activation of the stable Ce^{140} arising from the beta-decay of 40-hr. La^{140} before and during the irradiation. It is assumed that essentially all of the stable cerium was removed from the Ba^{140} solution when the latter was scavenged with $Fe(OH)_3$ precipitations, and that the Ce^{140} which grew into the solution in the subsequent four days was carried down completely with the final separation of 40-hr. La^{140}. Of the 115 d/s, 101 d/s was formed from the Ce^{140} which grew in before the bombardment, and 14 d/s from the Ce^{140} which grew in during the bombardment. The latter quantity was calculated by means of Eq. (4). Thus the net activity of 28-day Ce^{141}

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TABLE II. Data for La^{140} cross-section determination.

	28-day Ce from irradiated La sample	28-day Ce from un-irradiated La sample	40-hr. La from irradiated La sample	40-hr. La from unirradiated La sample
Observed activity corrected to 100% chemical yield and extrapolated to end of bombardment	5250 c/m	113 c/m	1.05×10^7 c/m	0.989×10^7 c/m
Correction to zero absorber	1.42	1.42	1.04	1.04
Correction to 100 percent counter geometry	3.12	3.12	3.04	3.04
Correction for aliquot (and to equal amounts of 40-hr. La for column 3)	2.50	26.5	10×10^5	1×10^5
Corrected activity	968 d/s	221 d/s	5.53×10^{11} d/s	0.521×10^{11} d/s
Corrected number of atoms at *beginning* of bombardment			1.74×10^{17}	0.164×10^{17}

which arose from neutron capture by 40-hr. La^{140} was (747 − 115) or 632 d/s. Then the cross section of 40-hr. La^{140} was calculated from Eq. (3) after substituting the subscript *C* for the subscript *B*. This is permissible because the half-life of B (3.7 hours) is very short compared to the half-life of C (28 days). The neutron flux (5.87×10^{10} n/cm² sec.) was derived from the radio-chemical analyses of the uranium monitors for the 12.8-day Ba^{140}, the fission yield of the latter (6.1 percent), the weights of the monitors, and the known thermal fission cross section of normal uranium. The value for the neutron absorption cross section of La^{140} is then 3.1 barns, probably reliable to within one barn.

DISCUSSION

The radiochemical analysis for 12.8-day Ba^{140} in the La^{140} sample of the final experiment indicated that much less of it carried through than in the preliminary experiment. By comparing the amounts from the two experiments, it can be shown that the maximum amount of 28-day Ce^{141} which could have resulted from neutron capture by Ba^{140} was only a few percent of the amount actually observed. Thus the 28-day Ce^{141} formed in both the final and preliminary runs must have resulted mostly from neutron capture by 40-hr. La^{140}.

If the lanthanum carrier used in the experiments had contained 25 percent stable cerium as an impurity, the same amount of 28-day Ce^{141} would have been formed as that observed. Therefore a separate neutron bombardment was made on a sample of the carrier, and then this was analyzed for 28-day Ce^{141}. Only a very small amount was found, proving that the lanthanum carrier was of sufficient purity. Another possible source of 28-day Ce^{141}, which could have interfered with the measurements, is a small uranium or plutonium impurity. By neutron irradiation 275-day Ce^{144} would be formed as well as 28-day Ce^{141}. In the final sample the 275-day Ce activity would have increased by 30 percent. Instead, a decrease of about 22 percent was found from a comparison of the cerium decay curves for the irradiated and unirradiated samples. This apparent decrease may be involved in measuring the soft radiations of the 275-day Ce^{144}. The relative amounts could have been estimated somewhat more accurately by carefully measuring the activity through 180 mg/cm² of aluminum so that essentially only the hard radiations of 17.5-min. Pr^{144} would be measured. The absorption curves of Fig. 2 cannot be applied accurately for making this comparison because the two samples were at slightly different distances from the Geiger counter. Furthermore, the distance had to be considerably greater for the absorption curves than for the decay curves in order to make room for the absorbers. That resulted in a low counting rate for the Pr^{144} from the unirradiated sample. If the necessary chemical yield and (inaccurate) geometry corrections are applied, an apparent decrease of about 14 percent is found for the Pr^{144} after irradiation. It seems improbable, therefore, that the La^{140} sample was contaminated by an important amount of a fissionable isotope.

The probable error in the cross section of La^{140} is estimated as large as 33 percent mainly because of the difficulty in measuring neutron flux and absolute disintegration rates accurately and because of the uncertainty introduced by the 275-day Ce impurity. In addition, it is still possible that some part of the induced 28-day Ce activity came from a small fissionable impurity. Circumstances did not permit the repetition of the experiment so as to further minimize these difficulties.

This paper is based on work performed at the Los Alamos Scientific Laboratory of the University of California under Government Contract W-7405-eng-36, and the information contained therein will appear in the National Nuclear Energy Series as part of the contribution of the Los Alamos Laboratory.

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