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NEUTRON ABSORPTION CROSS SECTIONS OF RADIOACTIVE La^{140}, Ba^{140}, AND TWO STABLE Ce ISOTOPES

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ABSTRACT

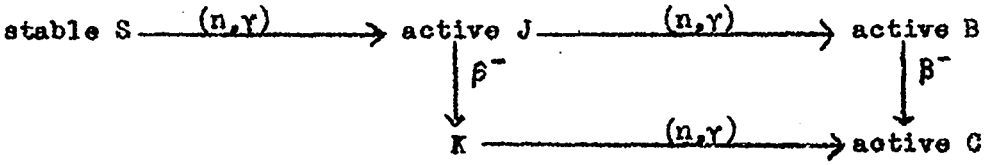
Two large samples (many curies) of 40h La^{140} were irradiated with slow neutrons at the Los Alamos homogeneous pile for 24 hours. The small amount of 3.7h La^{141} formed was allowed to decay to 28d Ce^{141} which was then radiochemically extracted and counted. From the activity measured, the neutron flux, and the known half-lives, a value of 3.1 ± 1.0 barns was calculated for the cross section of 40h La^{140}. An upper limit of about 400 barns was found for the cross section of 12.8d Ba^{140}.

The thermal-neutron absorption cross sections of stable Ce^{140} and Ce^{142} were also measured and found to be $0.27 \pm .06$ barns and $0.105 \pm .018$ barns, respectively, per atom of the naturally occurring element.

Neutron Absorption Cross Sections Of Radioactive* La^{140}, Ba^{140}, and Two Stable Ce Isotopes

Introduction

The activation method, which has been used extensively for the determination of cross sections of stable nuclei, has also been applied in several cases to the measurement of neutron absorption cross sections of radioactive isotopes(1). 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:



The saturation number of atoms of J is given by

$$N_J = N_S (nv) \sigma_S (1/\lambda_J) \qquad (1)$$

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

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

*Previous papers in this series on the cross sections of unstable nuclei are listed in references 1 - 6.

(1) S. Katcoff, Plutonium Project Record, Vol. IX B, 7.59.1 (1946).

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[1]. 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[1]. 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.

The above method has been applied to several radioactive isotopes. The thermal-neutron capture cross section of 85m Ba^{139} was determined to be 3.8 ± 1.0 barns[2]; upper limits of 470 barns[3], 200 barns[4] and 108 barns[5], have been set for the cross sections of 75m Kr^{87}, 17.5m Rb^{88}, and 55d Sr^{89}.

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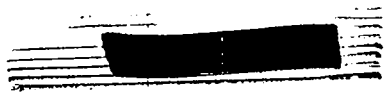
(2) S. Katcoff, Manhattan District Report CC-2908, (7 April 1945); Plutonium Project Record, Vol. IX B, 7.59.4 (1946).

(3) N. Sugarman and A. Turkevich, Manhattan District Report CC-2485 (15 December 1944); Plutonium Project Record, Vol. IX B, 7.59.2 (1946).

(4) S. Katcoff, Manhattan District Report CC-2739 (23 February 1945); Plutonium Project Record, Vol IX B, 7.59.3 (1946).

(5) S. Katcoff, Manhattan District Report CC-3059 (15 April 1945); Plutonium Project Record, Vol. IX B, 7.59.5 (1946).



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(1)

$$A_B = \frac{N_J^o T_J \sigma_J (nv)}{T_J - T_B} (e^{-\lambda_J t} - e^{-\lambda_B t}) \qquad (3)$$

where N_J^o 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:

$$A_C = \frac{N_J^o \sigma_K (nv)}{T_J - T_C} \left[T_J (1 - e^{-\lambda_J t}) - T_C (1 - e^{-\lambda_C t}) \right] \qquad (4)$$

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 toal activity of C. This amount is calculated by use of equation (4).

This method has been applied in experiments with the heavy isotopes, in setting limits on the cross section of long-lived I^{129} (0.8 barns to 80 barns)(6), and in measuring the cross section of 40h La^{140} as reported here.

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(6) S. Katcoff, Manhattan District Report CC-389 (29 May 1946): Phys. Rev. (in press).

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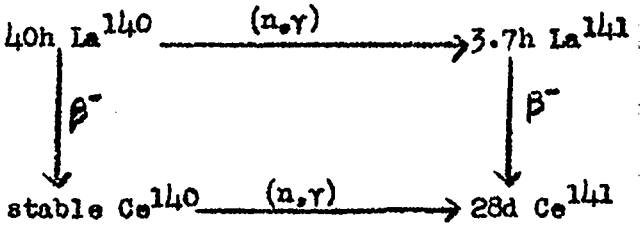
The nuclear processes involved in the latter were:



A quantity of 40h La^{140}, corresponding to a number of curies, was irradiated at the Los Alamos "water boiler" for a period of 24 hours. Several days later radiochemical analysis was made for the 28d Ce^{141} resulting from the irradiation.

Experimental Procedure and Results

Two experiments were performed: a preliminary one and a final one. A determination was also made of the neutron absorption cross sections of the two major stable cerium isotopes, Ce^{140} (89% abundance) and Ce^{142} (11% abundance), because the former had not been measured previously and the latter had been measured only roughly(7). For this determination 2.0g 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 weighing 170mg was used as a monitor for the neutron flux. Three weeks of cooling was allowed for the 33h 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 28d Ce^{141} and two others for 13.8d Pr^{143}, the daughter of 33h 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% counter

(7) S. Katcoff, Manhattan District Report CC-2739 (23 February 1945).

geometry (by means of a standard), to 100% chemical yield, and for decay. The neutron flux was measured by analyzing the uranium monitor for the 12.8d 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 equation (1) the neutron absorption cross sections which are given in Table I were calculated

Table I. Neutron Capture Cross Sections of Stable Ce

	Natural Atom Cross Section	Isotopic Cross Section	Isotopic Abundance
Ce^{140}	0.27 ± .06 barns	0.30 ± .06 barns	89 %
Ce^{142}	0.105 ± .020 barns	0.95 ± .18 barns	11 %

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[7] for the natural atom cross section.

For the preliminary experiment to determine the cross section of 40h La^{140}, a solution of 12.8d Ba^{140} was used which had been milked of its 40h La daughter several times before. After the last milking four days were allowed for more 40h La to grow in. Then 10mg 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 12.8d Ba. Then

a final precipitation was made as lanthanum "fluo-oxalate".(8) These operations were performed with several hundred curies of activity by means of remote control apparatus which was made available for this work by R. W. Spence. Most of the radioactive La^{140} was contained in a stainless-steel container of 0.020" wall thickness. It was transported to the "water boiler" in a thick lead shield and then transferred quickly by means of two 6' 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 the La^{140} was not irradiated and reserved for comparison with the irradiated sample.

After a two-week cooling period both samples were radiochemically analyzed, in duplicate, for 28d Ce^{141}, 40h La^{140}, and 12.8 Ba^{140}. The La analysis indicated that at the beginning of the irradiation 155 curies of 40h La had been in the pile and 6.5 curies had been reserved as a blank. In the Ce analyses it was necessary to reprecipitate $Ce(IO_3)_4$ eight times to decontaminate completely from the vastly greater activity of 40h La. Aluminum absorption curves of the radiations from the Ce samples indicated the presence of both 28d Ce^{141} and 275d Ce^{144}. It became obvious that these isotopes were present as small but important impurities in the original active La^{140} samples. A small increase in the 28d Ce^{141}, however, was found in the irradiated sample. This was used to calculate an upper limit of around 2 barns on the neutron absorption cross section of 40h La^{140}. The barium analyses indicated that an appreciable quantity of 12.8d Ba^{140} was also present. This isotope can also

(8) G. Friedlander, R. Spence, and R. G. Steinhardt, Jr., Manhattan District Report LA-557 (6 May 1946).

absorb neutrons to give 18m Ba^{141} which decays to 3.7h La^{141} and then to 28d Ce^{141}. Thus an upper limit on the cross section of 12.8d Ba^{140} could be calculated too. This was about 700 barns.

In the final experiment an attempt was made to remove as much radioactive cerium as possible from the 12.8d Ba^{140} solution prior to extraction of the final 40h La^{140} daughter activity. A Ba^{140} solution very similar to the one used before was scavenged three additional times by precipitating $Fe(OH)_3$ four days before finally milking the La^{140}. For this, 10mg of stable La carrier was again used but this time the two $La(OH)_3$ reprecipitations were done in the presence of about 500mg of stable Ba carrier. This was designed to further reduce the amount of 12.8d Ba^{140} carried along. Subsequent operations were essentially identical with those of the previous experiment. Although measurement with a radiation meter at the end of the milking operation indicated the presence of about 230 curies, a similar measurement made about two weeks later indicated that only about 1/10 of this had been irradiated at the water boiler. The radiochemical analyses showed that actually only 22.6 curies of 40h La^{140} was present at the beginning of the irradiation. The most probable explanation is that most of the active La^{140} was shaken out of the stainless-steel container into the lead shield during the nine mile journey over rough roads from the remote control equipment to the "water boiler". Fortunately, however, the remaining 22.6 curies was sufficient for measuring the cross section.

The radiochemical analyses for active Ce in both the irradiated and un-irradiated samples indicated a considerable improvement over the previous experiment. Although active Ce was not completely eliminated from the La^{140} samples before irradiation, a large increase in the amount of 28d Ce^{141} was

observed in the irradiated sample. This is clearly seen from the decay curves and aluminum absorption curves shown in Figs.1 and 2. The soft part of the radiation is from 28d Ce^{141} and 275d Ce^{144}; the hard beta ray is from 17.5m Pr^{144}, daughter product of 275d 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}.

Table II

	20d Ce from Irradiated La Sample	28d Ce from Unirradiated La Sample	40h La from Irradiated La Sample	40h La from Unirradiated La Sample
Observed activity corrected to 100% chemical yield and extrapolated to end of bombardment.	5250 c/m	113 c/m	1.05×10^6 c/m	0.989×10^6 c/m
Correction to zero absorber	1.42	1.42	1.04	1.04
Correction to 100% counter geometry	3.12	3.12	3.04	3.04
Correction for aliquot (and to equal amounts of 40h La)	2.50	26.5	1×10^6	1×10^3
Corrected activity	968 d/s	221 d/s	5.53×10^{11} d/s	5.21×10^{10} d/s
Corrected number of atoms at beginning of bombardment			1.74×10^{17}	1.64×10^{16}

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 28d Ce^{141} which was formed by the neutron irradiation of the La sample was (968 - 221) or 747 disintegrations/second. From this must be subtracted 115d/s which is the activity of 28d Ce^{141} formed by neutron activation of the stable Ce^{140} arising from the beta decay of 40h 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 40h La^{140}. Of the 115d/s, 101d/s was formed from the Ce^{140} which grew in before the bombardment and 14d/s from the Ce^{140} which grew in during the bombardment. The latter quantity was calculated by means of equation (4). Thus the net activity of 28d Ce^{141} which arose from neutron capture by 40h La^{140} was (747 - 115) or 632d/s. Then the cross section of 40h La^{140} was calculated from equation (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 (nv = 5.87 X 10^{10} $n/cm^2 sec$) was derived from radiochemical analyses of the uranium monitors for the 12.8d Ba^{140}, the fission yield of the latter (6.1%), 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 about one barn.

Discussion

The radiochemical analysis for 12.8d Ba^{140} in the La^{140} sample of the final experiment indicated that much less of it carried through than in the preliminary experiment. Using the upper limit of 700 barns for the cross section of Ba^{140} determined in the preliminary run, the maximum amount of 28d Ce^{141} which could have resulted from neutron capture by Ba^{140} is only 14d/s or only 2% of the amount actually observed (632d/s). Thus it is clear that the 28d Ce^{141} formed in both the final and preliminary runs must have resulted mostly from neutron capture by 40h La^{140}. Therefore the upper limit for the cross section of Ba^{140} can safely be lowered to about 400 barns, at least.

If the lanthanum carrier used in the experiments, had contained 75% stable cerium as an impurity the same amount of 28d Ce^{141} would have been formed. A separate neutron bombardment was therefore carried out on a sample of the carrier which was then analyzed for 28d Ce^{141}. Only a very small amount was found, indicating that the La carrier was of sufficient purity. Another possible source of 28d Ce^{141} which could have interfered with the measurements is a small uranium or plutonium impurity. However, this would also yield 275d Ce^{144} as well as 28d Ce^{141}. In the final sample the 275d Ce activity would have increased by 30% after the irradiation. Instead, a decrease of about 20% was observed. (This apparent decrease may be caused by the errors involved in measuring the soft radiations of the 275d Ce). It seems improbable, therefore, that a significant amount of a fissionable isotope could have contaminated the La^{140} sample.

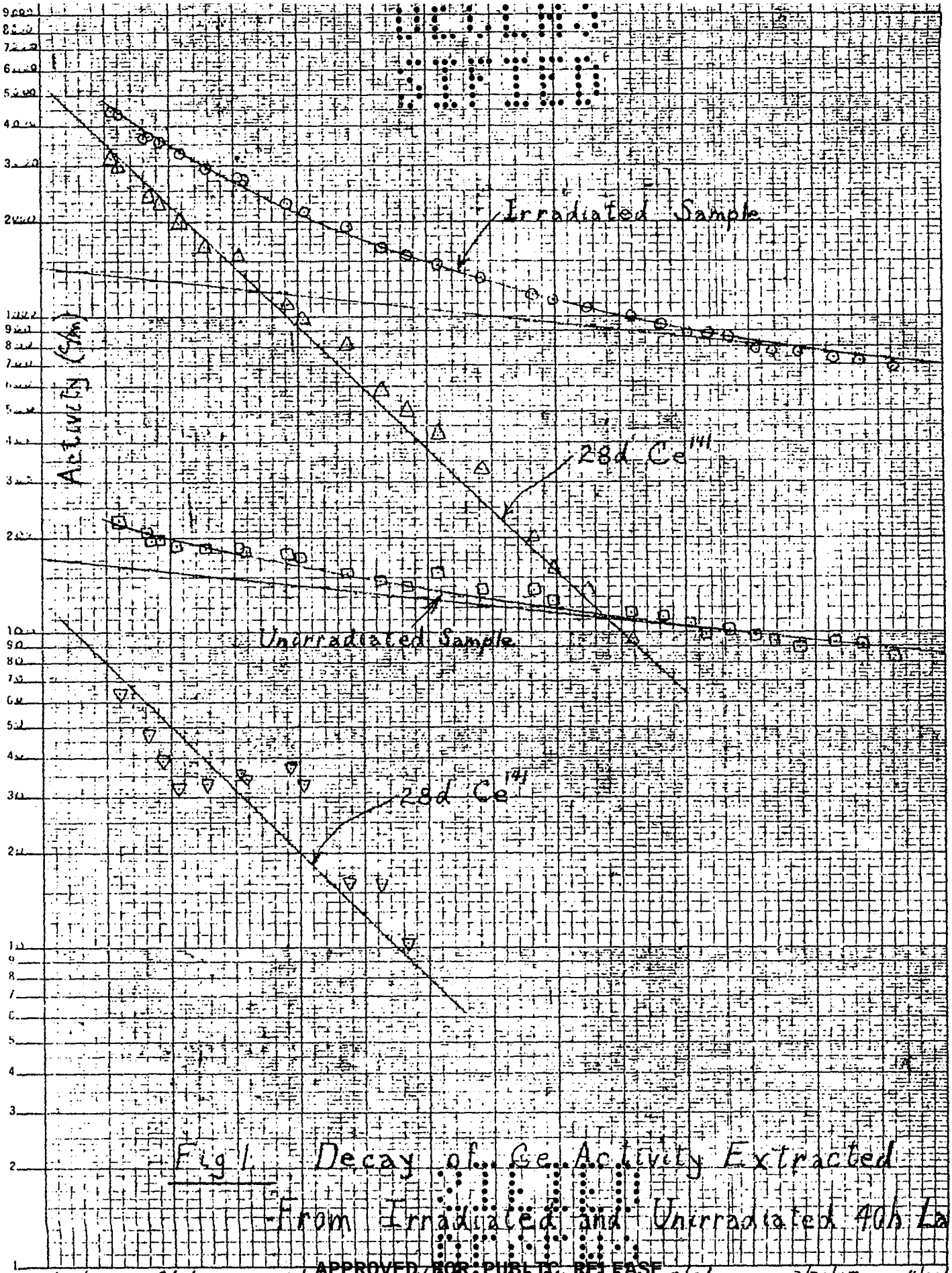


Fig 1. Decay of Ce Activity Extracted From Irradiated and Unirradiated 40h La

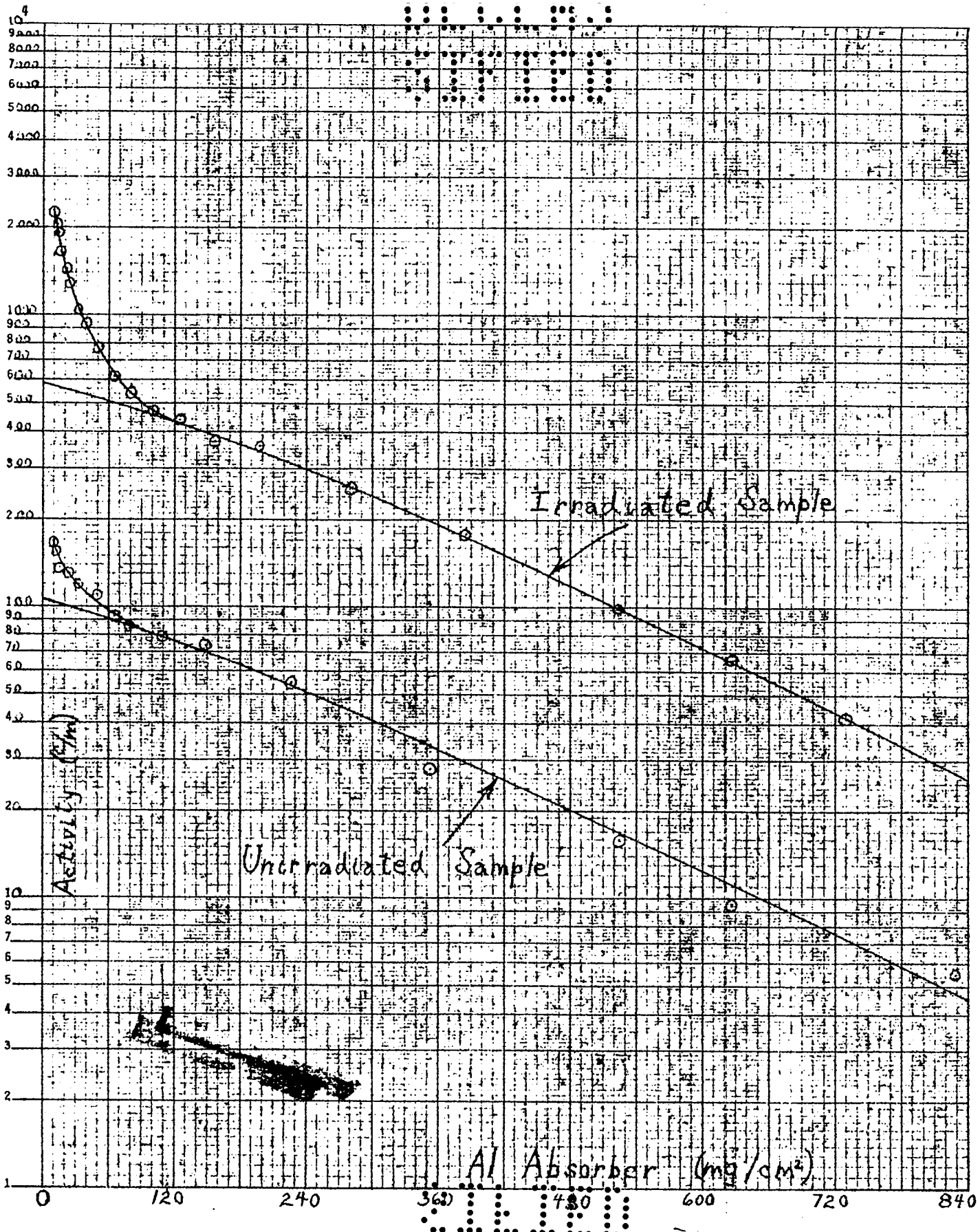


Fig. 2. Aluminum Absorption Curves of Ce Activity From Irradiated & Unirradiated 40h La^{140}

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