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AEC RESEARCH AND DEVELOPMENT REPORT

LOS ALAMOS SCIENTIFIC LABORATORY
OF THE UNIVERSITY OF CALIFORNIA ○ LOS ALAMOS NEW MEXICO

A MEASUREMENT OF THE $Li^{7}(n, n'\alpha)T$ CROSS SECTION FOR SEVERAL NEUTRON ENERGIES FROM 4 TO 15 MEV BY ABSOLUTE COUNTING TECHNIQUES

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A MEASUREMENT OF THE $Li^7(n, n'\alpha)T$ CROSS SECTION FOR SEVERAL NEUTRON ENERGIES FROM 4 TO 15 MEV BY ABSOLUTE COUNTING TECHNIQUES

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by

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Contract W-7405-ENG. 36 with the U. S. Atomic Energy Commission

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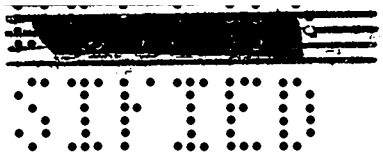

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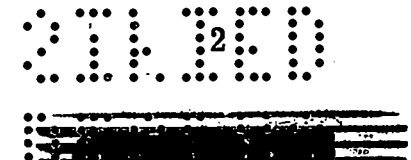
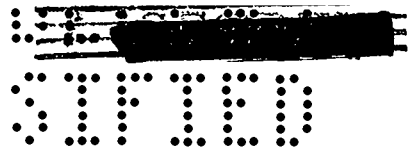


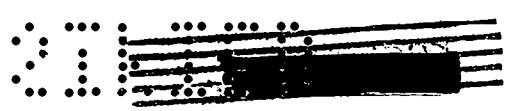
ABSTRACT

A measurement of the $Li^7(n,n'\alpha)T$ reaction cross section by absolute counting of the tritium betas is described. The cross section was measured at several neutron energies between 4 Mev and 15 Mev. A comparison with results by emulsion techniques is given.

ACKNOWLEDGMENT

The authors wish to thank the many people of LASL Groups P-3, P-4, and P-9 who willingly operated their machines to supply the neutrons for the irradiations. We especially wish to thank S. J. Bame of P-3, E. Haddad, R. L. Henkel, J. E. Perry, Jr., and R. K. Smith of P-9, and J. H. Coon and R. W. Davis of P-4, who measured the neutron fluxes.





INTRODUCTION

One way of measuring the cross section for the $Li^7(n,n'\alpha)T$ reaction is to determine the tritium yield by absolute counting of its beta activity. The half-life for the beta decay of tritium is 12.262 ± 0.004 years,[1] and the end point energy of the beta is 18.0 kev.[2] These properties require rather long irradiations for the expected cross sections and demand that the activity be internally counted.

EXPERIMENTAL TECHNIQUES

A low background system was built using an ethane proportional counter. Figure 1 is a schematic sketch of this counter. By the use of Teflon plugs on the ends of a 9 in. long, 1 in. OD copper tube, counting losses in the end regions were minimized. The center wire (0.002 in. platinum wire) was threaded through the 1/8 in. copper tubes held in the Teflon plugs and crimped. The steel ends were screwed down tight against the Teflon plugs to produce a vacuum seal for the counter. The platinum and copper materials were chosen because they have lower solution rates for hydrogen than most structural materials. Hence, the increase in counter background due to internal contamination should be reduced to a minimum. The counter had a volume of 80 cc and was filled with ethane to a pressure of 20 cm Hg.

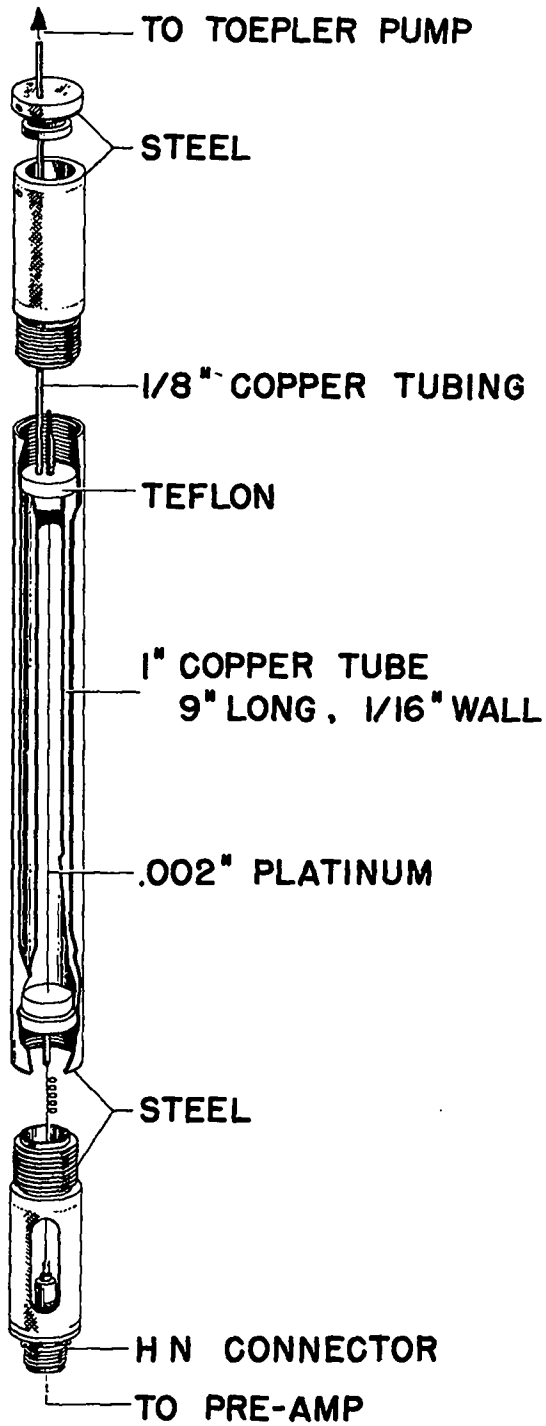


Fig. 1. Teflon-ended counter for counting the tritium beta activity.

The counter proved to be about 95% efficient by comparison with a counter of known efficiency.[3,4]

An anticoincidence counter surrounded the tritium counter and was built as an annular ring containing ten 0.003 in. Kovar wires. The counter and anticoincidence counter were placed in a 3 in. thick steel sleeve and then surrounded by 2 tons of lead as shown in Fig. 2.

The signals from each of these two counters were fed into amplifiers which had been modified to accept pulses 30 times greater than normal saturation without blocking. The amplifier gains were set at about 10^4. By feeding the outputs of these amplifiers into the anticoincidence circuit with the discriminators set at about 4 volts, it was possible to get voltage plateaus for each counter with less than 1% change in 100 volts at 2000 volts. The plateau characteristics were not altered by the addition of hydrogen up to 25% of the counter filling. The tritium counter had a background counting rate of about 55 counts/min, but when used with the anticoincidence circuit the rate was reduced to about 2.5 counts/min.

Samples of metal Li^7 were loaded into copper capsules 1/2 in. OD and 1/2 in. high. The bases and covers were drawn from 0.007 in. electrolytic copper stock. Dry helium flowing continuously into an inverted box gave a sufficient working volume for cutting the metal lithium and pressing it into these capsules by hand. The capsules were covered and soldered shut to form the required seal. Care was required since the tiniest hole would permit the lithium to react with the water vapor in

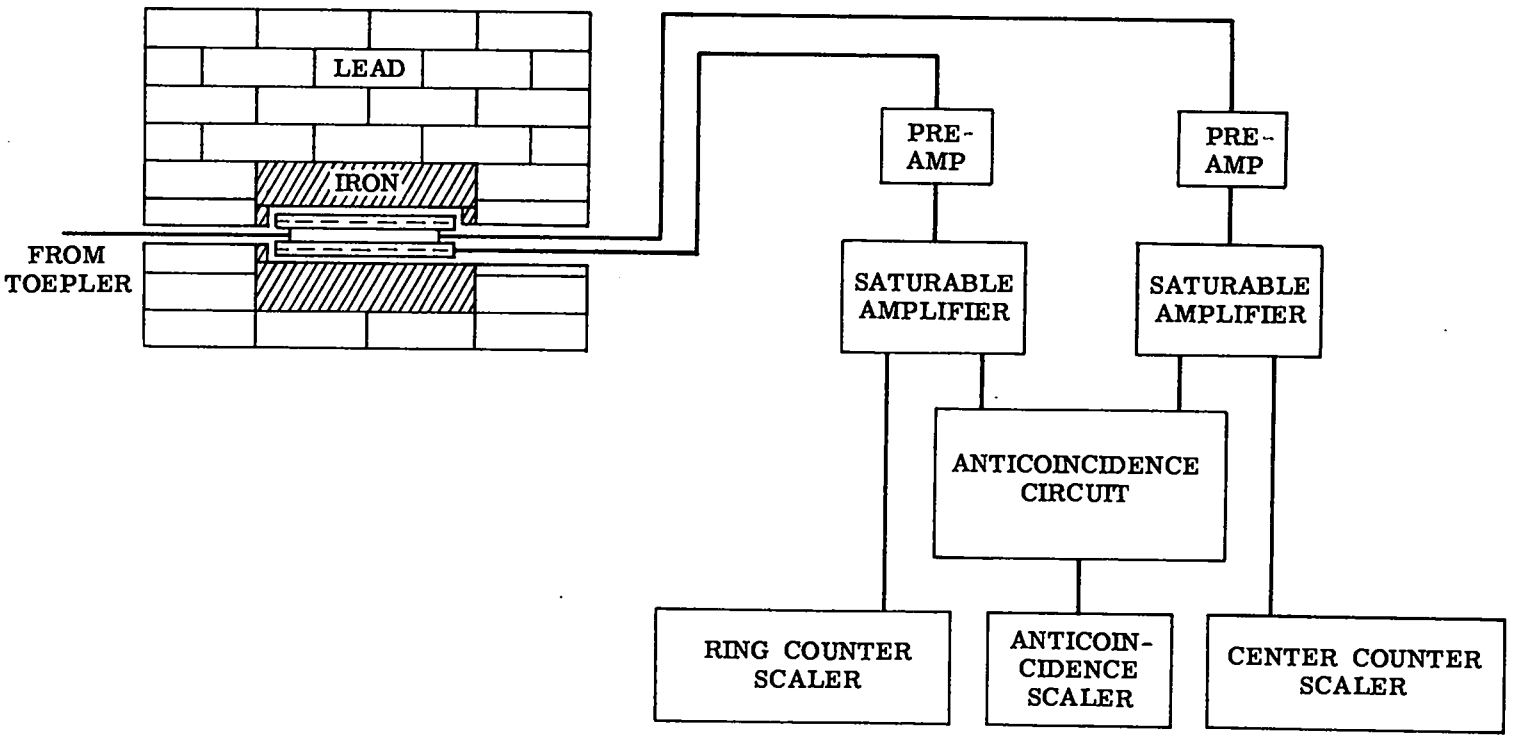


Fig. 2. The shield and anticoincidence circuit.

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the air, forming LiOH which released water vapor during the extraction process. This water vapor destroyed the counter plateau properties and made the tritium analysis very difficult and subject to large errors.

The samples were irradiated in monoenergetic neutron fluxes supplied by the P-3 and P-9 Van de Graaffs and the P-4 Cockcroft-Walton.

ANALYSIS

In order to extract the tritium, it was necessary to heat the capsules until the solder seal was broken and then to heat the capsules above the hydride decomposition point for any of the possible hydrides created. Lithium metal is a very reactive material, especially at temperatures above its melting point. The following procedure was adopted. The capsule was placed in a steel sleeve 3/4 in. long, 5/8 in. ID, and with a 0.035 in. wall. Loose fitting steel discs covered the ends, confining the capsule but permitting evolved gases to escape. This sleeve was used as a thermal radiator in the induction heating system. Surrounding the sleeve was an open-ended quartz tube 6 in. long and 20 mm OD. This assembly was placed in an 8 in. long, 24 mm OD quartz tube which had one end closed. Finally the entire assembly was inserted in an 18 in. long, 28 mm OD quartz tube which was waxed (Apiezon W) to the vacuum line analysis circuit. This three layer quartz system was surrounded by a water cooling jacket around which the induction heating coils were wound. The system is sketched in Fig. 3.

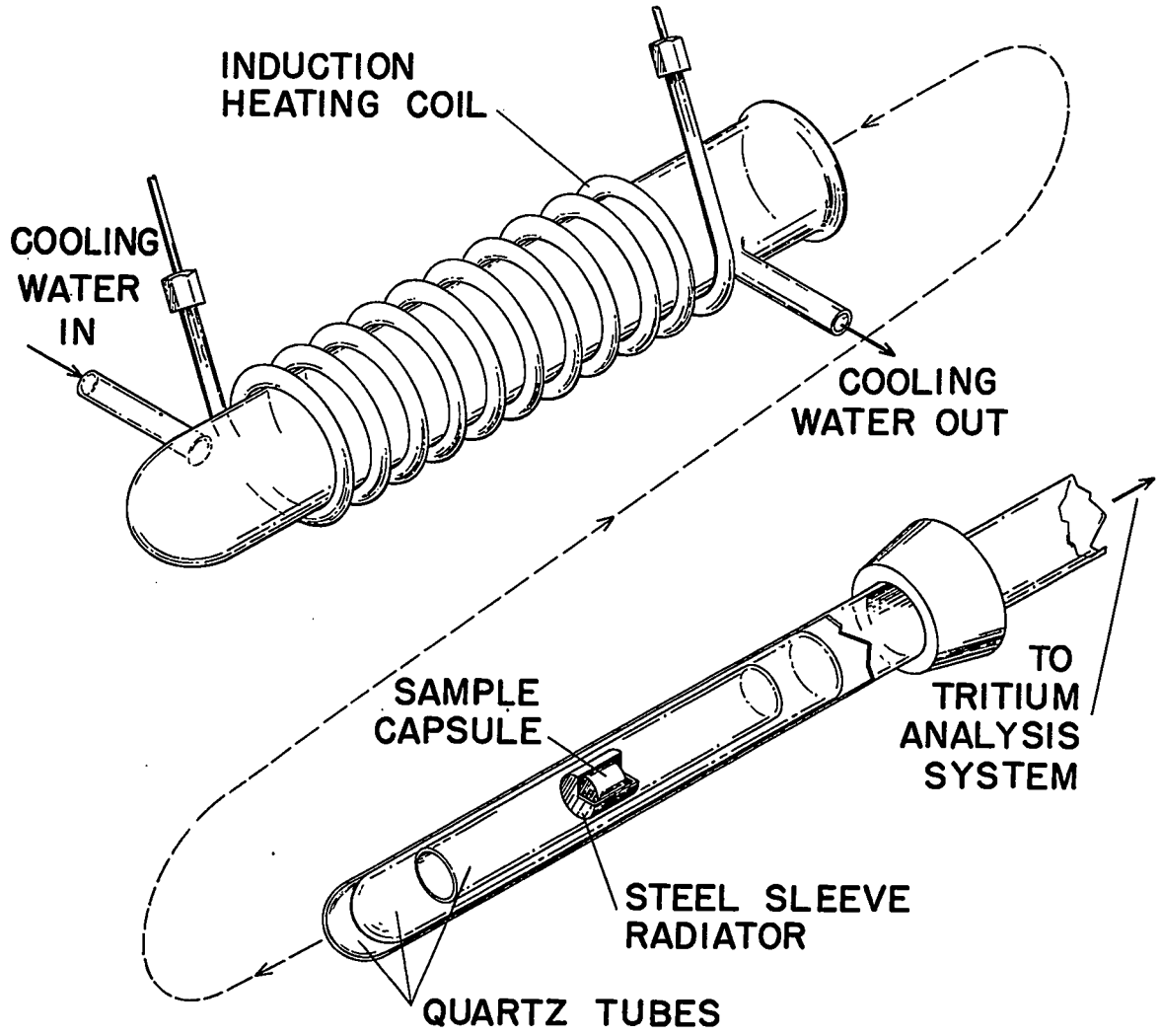

Fig. 3. Quartz evolution system.

The induction heating of the steel sleeve caused the enclosed capsule to rise in temperature above the solder melting point. The lithium and the solder normally reacted exothermically but the explosive response was confined by the steel sleeve. Much of the tritium was released at this time. The temperature of the steel sleeve was raised to 1200°C, which caused the lithium to react with the quartz and usually destroyed the inner 6 in. tube (sometimes the 8 in. tube also). Occasionally during this operation some lithium metal was deposited on the water-cooled outer quartz tube. Because some tritium was carried along with the lithium, a second extraction was made using a muffler furnace. The coils and water jacket were removed and the quartz system heated to 1100°C for an hour. This evolved most of the remaining tritium (5 to 10% of that extracted during the induction evolution).

The reason we used the water-cooled induction heating system was to force the lithium reactions with the quartz to occur at a place which would not destroy the "vacuum" of the analysis line. The second furnace heating permitted lithium reactions with the quartz vacuum jacket but considerably fewer reactions occurred because of the first induction heating. Even if the vacuum quartz tube broke during the second heating, less than 5% of the tritium would remain unanalyzed.

The evolved gases were collected in a Toepler pump (Fig. 4) and added to the ethane of the counter. When the evolved gases were added successively to the counter, the measured activity increased and reached a limiting value, indicating that all the tritium had been extracted. A

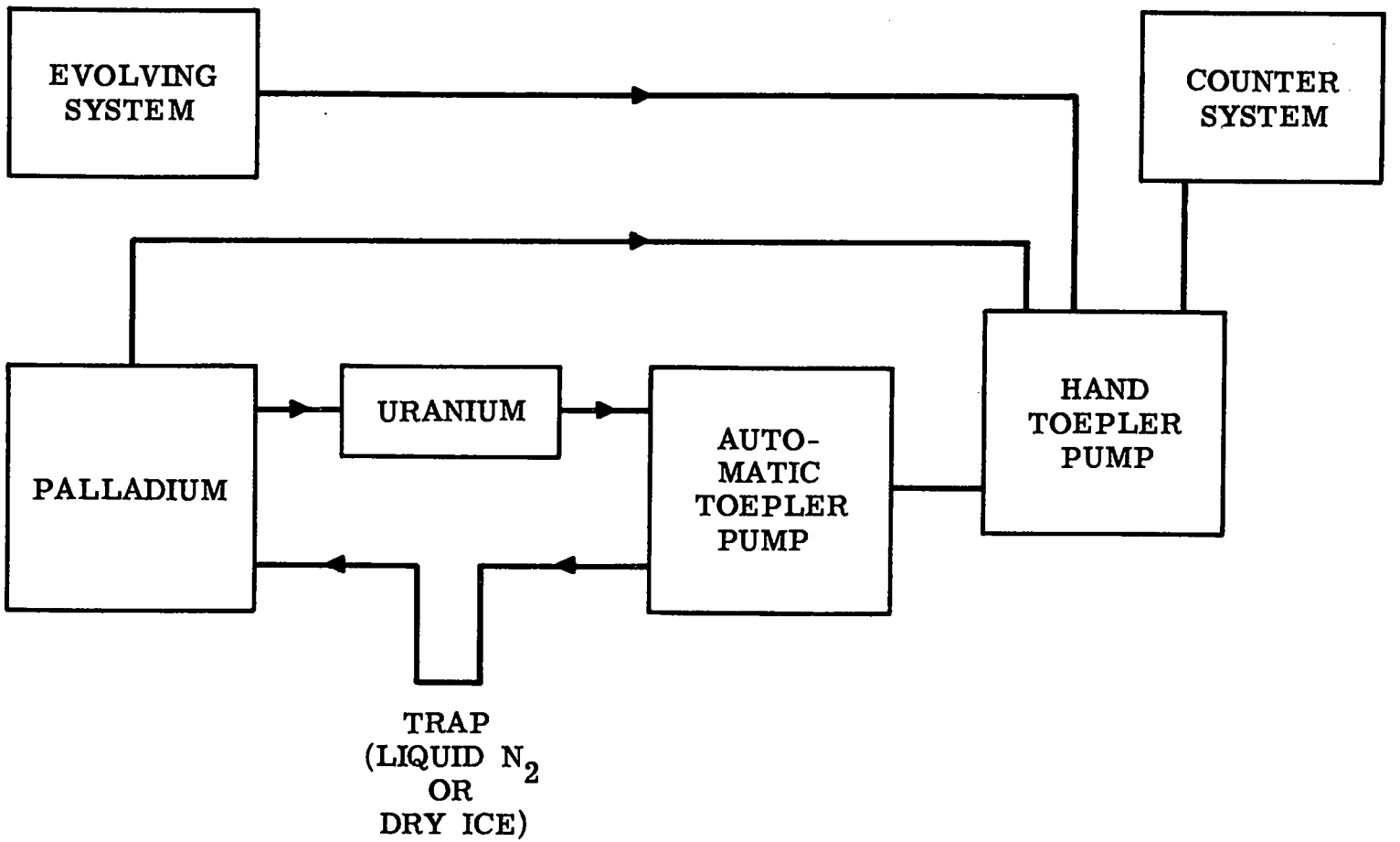

Fig. 4. Analysis system.

small amount of hydrogen was added to the system before the evolution to act as a carrier. This prevented any loss of tritium in the transfer.

In order to identify the activity as that due to tritium, the gas was passed through a palladium window. The counter filling, including the evolved tritium, was transferred by the hand Toepler pump to the automatic Toepler pump loop. A liquid nitrogen trap condensed the ethane, leaving the tritium (and hydrogen carrier) to pass through the heated palladium (500°C). The counter was refilled with new ethane and the gases which had passed through the palladium were added to the counter. Normally all the original activity could be recovered in 1 to 2 hours. If some of the tritium was collected as water vapor, it condensed in the liquid nitrogen trap. When this trap was replaced with a dry ice trap, the water vapor was held but the ethane released. Permitting the remaining water vapor to pass over uranium at 900°C released the hydrogen and tritium. These gases could be passed through the heated palladium and added to the counter to check for the remaining activity. With this system it was possible to verify the activity as that due to tritium to better than 95% in all cases checked.

CORRECTIONS

The source of neutrons was really a line rather than a point and corrections were made to account for this. Because the samples were not located on the target axis in all cases, an adjustment was made for the angular distribution of the neutrons. Neutron flux attenuation in the samples was taken into consideration.

The enriched Li^7 material was analyzed mass spectrographically. A chemical analysis was made on several typical capsules to determine the amount of lithium they contained. This permitted a correction to be made to account for any nitride or hydroxide formation during the capsule loading.

The tritium activity measurement was corrected for any time delay between irradiation and extraction. Empty capsules were irradiated at each energy in the same flux and analyzed. Although only a trivial amount of activity (~2 counts/min) was found, the tritium counting rates from the samples were adjusted for it.

RESULTS AND RELIABILITY

4.0 Mev Neutrons

Five 0.5 g samples of metal Li^7 were irradiated at the P-3 Van de Graaff. The 4.0 Mev neutrons were supplied by 1.50 Mev deuterons irradiating a deuterium gas target 3 cm long (the deuteron energy at the center of the target was 0.90 Mev). The ring of capsules (Fig. 5) was located so that the distance from the effective center of the neutron source to the capsules was about 6 cm. The neutron flux was determined by the deuteron beam and the D-D reaction cross section.[5] One of the samples was improperly sealed and could not be analyzed. The other four showed a spread of 20% with an average deviation of 8%. The uncertainty in the neutron flux was estimated to the ±10%. The measured cross section was 105 ± 15 mb. The neutron energy was 4.0 ± 0.2 Mev.

Fig. 5. Capsule irradiation holder.

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5.6 Mev Neutrons

Five 0.5 g samples of Li^7 were irradiated in a flux of 5.6 Mev neutrons at the P-9 Van de Graaff. The neutrons were supplied by 6.7 Mev protons bombarding a 6 cm long gas target of tritium (the proton energy at the center of the target was 6.38 Mev). The capsules containing the lithium were located in a ring (Fig. 5) in front of the tritium gas target so that the distance from the center of the capsules to the effective center of the neutron source was about 10 cm. The results from the five samples showed a spread of 10% with an average deviation of 3%. The neutron flux was measured with a counter telescope[6] and should have a reliability of ±7%. The extraction and counting systems should have a reliability of ±6%. The cross section was measured to be 340 ± 35 mb. The neutron energy was 5.60 ± 0.05 Mev.

8.0 Mev Neutrons

Six 0.5 g samples of Li^7 were irradiated in a flux of 8.0 Mev neutrons at the P-9 Van de Graaff. The neutrons were supplied by 4.9 Mev deuterons bombarding a deuterium gas target 6 cm long. The deuteron energy at the center of the target was 4.75 Mev. The capsules containing the lithium were located in a ring (Fig. 5) in front of the deuterium gas target so that the distance from the effective source of neutrons to the center of the capsules was about 10 cm.

The extraction system leaked during the analysis of one of the samples. The other five gave results which had a spread of 50% of the average value but which had an average deviation of 12%. The neutron

flux was calculated from the D-D cross section[5] and the geometry. The flux should have a reliability of ±8%.

The cross section was measured to be 450 ± 60 mb. The neutron energy was 8.0 ± 0.1 Mev.

9.6 Mev Neutrons

Five 0.5 g samples of Li^7 were irradiated in a flux of 9.6 Mev neutrons at the P-9 Van de Graaff. The neutrons were produced by 6.6 Mev deutrons bombarding a deuterium gas target 6 cm long. The deuteron energy at the center of the target was 6.4 Mev. The capsules containing the lithium were located in a ring (Fig. 5) so that the distance from the effective source of neutrons to the center of the capsules was about 10 cm.

The five samples gave results which had a total spread of 14% of the average value, but they had an average deviation of only 5%. The neutron flux was measured by a counter telescope[6] with an uncertainty of ±7%. The cross section was measured to be 390 ± 40 mb. The neutron energy was 9.6 ± 0.15 Mev.

14 Mev Neutrons

Five 0.4 g samples of Li^7 and six 0.4 g samples of normal lithium were irradiated by neutrons from the D-T source at the P-4 Cockcroft-Walton. Because the thermal neutron flux near the Cockcroft-Walton is relatively higher than that near the Van de Graaffs, a check on its effect was deemed advisable. Since normal lithium has about 100 times more Li^6 than did the enriched Li^7, the normal lithium samples would

give significantly higher values for the cross section than the enriched Li^7 samples if thermal neutrons were significant. Half the samples were located at 20 cm from the center of the neutron source and the remainder at 10 cm.

Figure 6 shows a graph of the results. The more distant normal lithium samples gave higher values of the cross section than the closer ones, indicating an effect of the lower energy neutrons on the Li^6. It would appear that there is a thermal neutron background which is relatively constant. Since the enriched Li^7 samples had only about 1% as much Li^6 as in normal lithium, the correction was trivial. Positioning the samples at various angles with respect to the deuteron beam made it possible to have a neutron energy variation from 13.4 Mev to 14.9 Mev.

The samples of normal lithium proved to be of some assistance in determining the change in the cross section with energy for Li^7 since the change would be the same except for a displacement upwards due to the Li^6 contribution. The listed values for the cross section are from the Li^7 curve in Fig. 6 rather than from the individual measured values.

The neutron flux was measured by counting the associated α particle in the D-T reaction and has an uncertainty of about $\pm 5\%$ at 13.5 and 14.8 Mev, and $\pm 3\%$ at 14.1 Mev. With this system, the cross sections are as follows: 13.4 ± 0.2 Mev, 325 ± 20 mb; 14.1 ± 0.1 Mev, 310 ± 15 mb; and 14.8 ± 0.2 Mev, 305 ± 20 mb.

It must be remembered that this technique measures only the tritium production and will not distinguish the method of creation. Even though

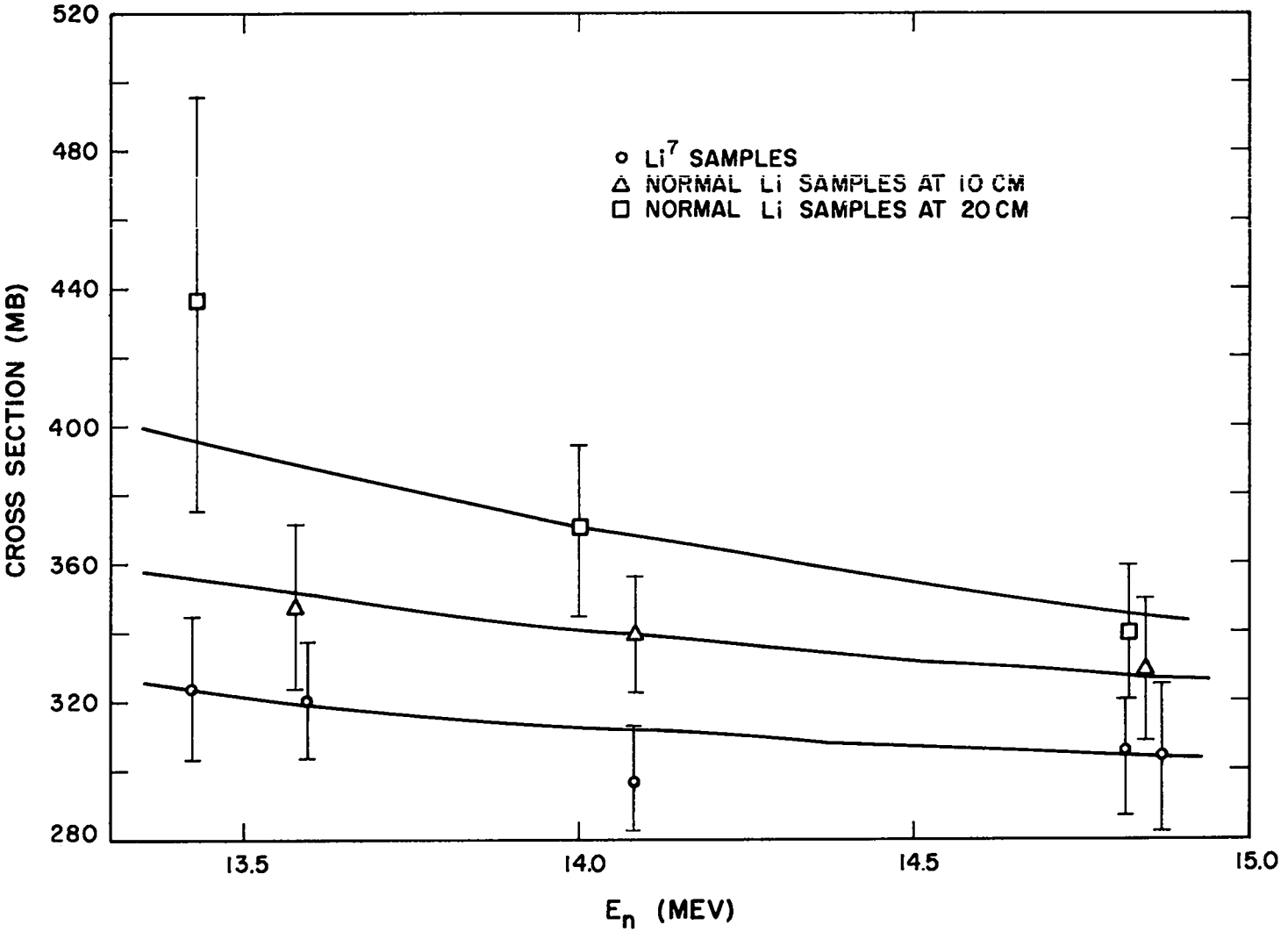

Fig. 6. The $Li^7(n,n'\alpha)T$ reaction cross section for 14 Mev neutrons. The variation in neutron energy was obtained by positioning the samples at various angles with respect to the deuteron beam.

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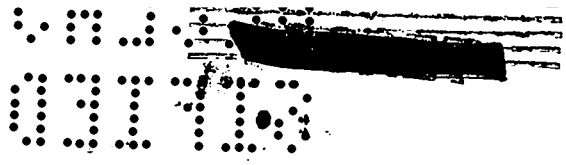


the cross section is listed as $Li^7(n,n'\alpha)T$, some fraction of the tritium yield may come from $Li^7(n,t)He^{5*}$. Since the He^5 breaks up into a neutron and an α, the final products are the same as in the inelastic scattering case.

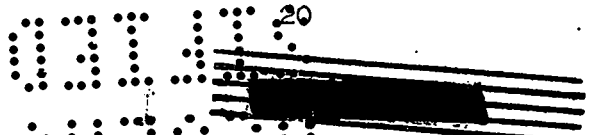
The results of these cross section measurements are listed in Table I. Typical counting rates from the samples are given there.

TABLE I. $Li^7(n,n'\alpha)T$ CROSS SECTION AS A FUNCTION OF THE NEUTRON ENERGY

Neutron Energy (Mev)	No. of Samples	Typical Counting Rate (counts/min)	Cross Section for Tritium Production (mb)
4.0 ± 0.2	5	100	105 ± 15
5.6 ± 0.05	5	220	340 ± 35
8.0 ± 0.1	6	70	450 ± 60
9.6 ± 0.15	5	400	390 ± 40
13.4 ± 0.2	4	1200	325 ± 20
14.1 ± 0.1	3	1200	310 ± 15
14.8 ± 0.2	4	1200	305 ± 20

COMPARISON WITH EMULSION TECHNIQUE

Lithium-7 loaded emulsions were irradiated by various energy neutrons and analyzed by members of LASL Group P-10. These results[7] are shown in Fig. 7, along with the present work for comparison. The smooth curve drawn through the points shows a reasonable variation in the cross



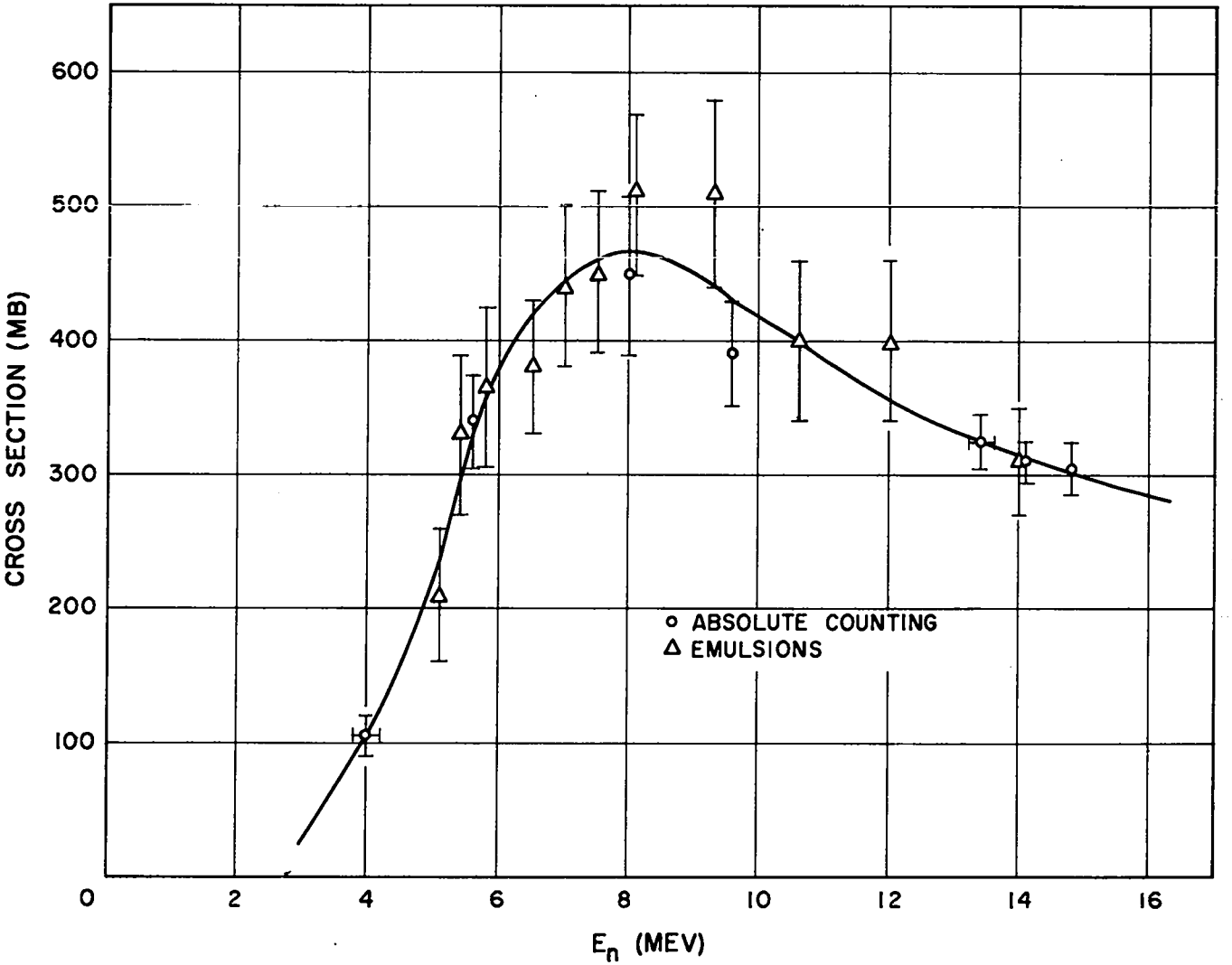


Fig. 7. The $Li^7(n,n'\alpha)T$ reaction cross section as a function of neutron energy, showing the results from both absolute counting and emulsion techniques.

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section. The rapid increase in yield corresponds to the excitations of the 4.6 Mev and 6.5 Mev levels in Li^7.

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