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Lithium Neutron Cross Sections During the Manhattan Project and the Quest for the H-Bomb

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Abstract — *Neutron cross sections of the stable lithium isotopes ^{6}Li and ^{7}Li were of interest in the 1940s and 1950s in part because of their reactions, which form tritium using moderated neutrons on ^{6}Li and higher-energy neutrons on either isotope. Lithium remains of interest today for use as a blanket and shielding material in fusion reactors, where it can be used to breed tritium for a self-sustaining fuel cycle. During the Manhattan Project, the resonance in the ^{6}Li(n,t) reaction was discovered and later became important for enhancing tritium production for nuclear technologies. The dominant natural isotope ^{7}Li was and remains of interest because of the expense of enriching ^{6}Li. It has been oft reported that the 1954 Castle Bravo nuclear test had a yield twice as large as expected because the nuclear explosive device designers had not properly accounted for the benefits from the ^{7}Li isotope in the fuel; we note that this explanation is false.*

Keywords — *Lithium, history, cross sections.*

Note — *Some figures may be in color only in the electronic version.*

I. INTRODUCTION

The disintegration of ^{6}Li on neutron bombardment into an alpha particle and a triton was first reported by Chadwick and Goldhaber in *Nature* in January 1935,[1] with a Q-value of approximately 5 MeV. By the time Bethe published his seminal work on nuclear theory in 1937, ^{6}Li was identified as a 1/v absorber with a large thermal absorption cross section and a cross section for tritium production with thermal neutrons of 900 barns (b),[2] as compared to 938 b today.[3] Cross sections at energies up to 800 keV were studied in more detail during the Manhattan Project.[4]

The potential benefits of ^{6}Li as a thermonuclear fuel were discussed in the United States from a very early date, certainly by 1942, and probably earlier. In the earliest references, ^{6}Li was often considered in terms of its beneficial exothermic energy production, rather than for its tritium breeding potential. For example, Oppenheimer's "Memorandum on Nuclear Reactions," dated July 1942, addressed the former aspects, whereas Manley's September 1942 document appears to discuss the latter, tritium breeding (see Fig. 17 in Ref. [5]).

In the case of ^{7}Li, proton reactions were of particular interest, as these were more readily accessible and allowed for the study of resonance capture behavior in a simple compound nucleus.[2] In addition, this reaction

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was recognized in 1949 as a viable source of quasi-monoenergetic neutrons.[6] It was not until 1953 that significant interest arose in the cross-section details of both isotopes at 14 MeV, likely, in part, because of the limited availability of deuterium-tritium (DT) neutron sources.[7] On March 1, 1954, one of the first peer-reviewed papers on these reactions was published,[8] which estimated a possible production path for tritium via ^{7}Li with 14-MeV neutrons of 50 mb, a factor of roughly 7 smaller than the modern value, although this was for only a single production channel.

Following the Castle nuclear test series in the Pacific that same month, Los Alamos undertook further study of the lithium cross sections. By July, leveraging predominately existing measured data, they had an estimated value for ^{7}Li(n, n′) tritium production with 14-MeV neutrons of 325 ± 75 mb.[9] This was very close to today's ENDF/B-VIII.0 (and IRDFF-II) value of 302 mb. Also, this early report quotes a measured ^{7}Li(n, n′t) cross section of 410 mb for the 14-MeV total inelastic cross section, which agrees favorably with today's ENDF-VIII.0 value of 369 mb.

These measurements were further refined later in the 1950s across a variety of energies for both isotopes using a combination of techniques, including nuclear track emulsion measurements, absolute counting of tritium beta particles, and measurement of the inelastic neutrons. Evaluations based on these measurements often relied on the use of transmission data from the pulsed sphere approach, originally published by Graves and Rosen in January 1953.[10] This approach has persisted as a key method for benchmarking neutron cross-section data at high energies,[3] including in the MCNP 6.3[11] validation suite.

To orient readers to the modern understanding of these cross sections, Fig. 1 shows the International Atomic Energy Agency's recent IRDFF-II evaluation[12] of the tritium production cross sections from natural lithium and the isotopes. Below 3 MeV, only ^{6}Li produces tritium and is dominated by the large resonance at 239 keV, while at 14 MeV, tritium production from ^{7}Li exceeds that from ^{6}Li by an order of magnitude. At the beginning of the Manhattan Project in 1943, none of these details were known.

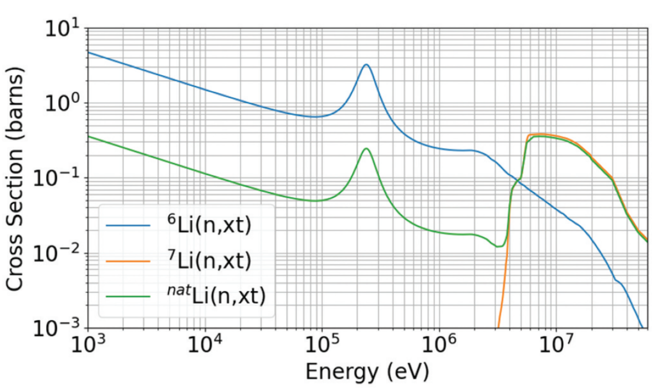

Fig. 1. IRDFF-II evaluated cross sections for tritium production from naturally occurring lithium isotopes and the resulting natural abundance cross section.[12]

II. MANHATTAN PROJECT LITHIUM MEASUREMENTS

The first Manhattan-era measurements of lithium neutron cross sections relied on the development of both neutron sources and detectors. Most used a neutron source, along with moderators of polyethylene and boron carbide. Kinematics were used to select neutrons of a particular energy to interrogate a target. The neutron source consisted of either a proton beam impinging on a natural lithium target or a deuterium beam impinging on a deuterium target. Early efforts to measure neutron outputs relied on the measurement of proton recoils from thin polyethylene foils; however, these were replaced by what we today would call fission chambers, which are designed to have a nearly uniform response from 10 keV to over 2 MeV by first thermalizing the neutrons in paraffin prior to them interacting in the chamber. In either case, the general approach was to use the most accurately measured absolute cross section (^{235}U fission) and take the ratio of the count rate to a different sample of known mass thickness. Similar approaches underpin many nuclear data measurements today.

In 1944, Los Alamos report LA-150[4] identified the ^{6}Li neutron resonance leading to tritium and alpha production and found it to occur at ~280 keV. The measurement was made on a natural lithium target and gave a magnitude of 0.23 b (but the high-resolution set indicated a higher cross section closer to 0.275 b). By 1946, this value had been revised downward to 0.21 b based on improved measurements of the ^{235}U fission cross section.[13] Since ^{6}Li makes up 7.59% of natural lithium, the measured cross section corresponds to a ^{6}Li(n,t) cross section of 3.3 to 2.8 b. Both measurements are in fair agreement with the modern value of 0.244 b at 239 keV shown in Figs. 1 and 2. The relevant parts of LA-150 are reproduced in Figs. 3 and 4.

In 1947, Goldsmith et al. from Brookhaven published an early version of the "barn book" in their paper in "Review of Modern Physics."[14] This compilation included a figure showing the 1944 Los Alamos data on Li(n,t), identified as "Los Alamos unpublished, Blair et al." Blair was a contributor to LA-150.

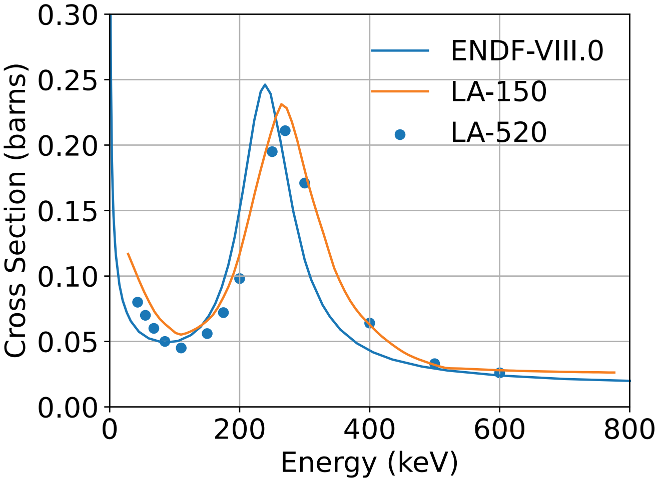

Fig. 2. Natural lithium cross sections reported in LA-150, LA-520, and ENDF/B-VIII.0.

Two independent sets of data were obtained and are shown in Fig. 14. The higher-energy data were acquired with neutrons whose energy was less well-defined than for the case of the lower-energy data. The results shown are admittedly preliminary and incomplete but do indicate quite clearly that a marked resonance in the cross section for this process occurs in the neighborhood of 280 Kev. The magnitude and width of this resonance are not well-established as witnessed by the discrepancy between the two sets of data taken with different resolving powers.

[redacted]

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Fig. 3. Los Alamos documented the discovery of the resonance in ^{6}Li(n,t) in 1944 (reproduced from LA-150).

III. PULSED SPHERE METHOD

Many different techniques have been developed to measure energy-dependent cross sections for secondary neutron production from high-energy neutron sources, but the pulsed sphere approach was highly reliable with minimal bias. It relied on a deuterium beam impinging on a tritium target placed in the middle of a sphere of the isotope of interest. The total nuclei per square centimeter of the target material was chosen to be approximately one-quarter of the estimated cross section, in other words, one-quarter of a mean free path thick. It was then possible to measure the neutron spectra with and without the sphere of the isotope of interest present to determine the relative neutron production rate (or multiplication) and transmission as a function of energy.

A variety of techniques that do not require a pulsed beam have been used to measure neutron energy spectra in the pulsed sphere setup, beginning with the use of

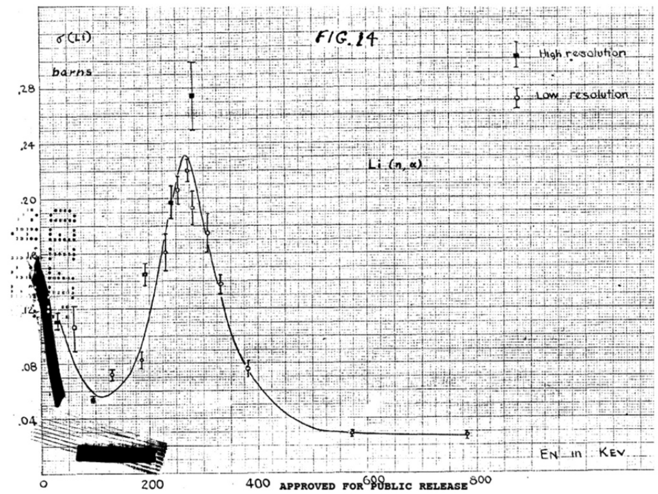

Fig. 4. First data measured for the ^{6}Li(n,t) resonance. The target was natural lithium, so the ^{6}Li cross section was about 13 times larger than shown here (reproduced from LA-150).

proton recoils in nuclear track plates. Even with nuclear track plates, it was possible to measure neutron fluxes with an assessed error in absolute intensity of 12%. This method was key to improving the understanding of ^{7}Li cross sections in the mid-1950s.

IV. TRITIUM PRODUCTION FROM ^{7}Li

Lithium-7 has a particular value as a breeding material that is appreciated in controlled fusion energy concepts today. Higher-energy neutrons (e.g., 14 MeV) can cause a (n,n′t)α reaction, which creates tritium and a secondary neutron. This benefits overall neutron economy since the emitted neutron with a degraded energy can still create more tritium in subsequent reactions with ^{6}Li. This reaction was not realized until >5-MeV neutron sources were readily available.

By 1954, sufficient information about the decay of excited states of ^{7}Li to tritium was known[15] such that the production of tritium via inelastic scattering should have been expected. Yet a review of the literature available at the time indicates that the first observation of tritium through this channel (one portion of it) was reported in a March 1954 paper by Frye at Los Alamos.[8] As noted previously, a follow-on report by Thomas in July 1954 made a surprisingly accurate prediction of the ^{7}Li(n,n′t) cross section (325 mb at 14.2 MeV) by analyzing available data and evaluating the implied relative magnitudes of different channels based on theory and a combination of experiments.[9] According to Rosen and Stewart,[16] this prediction was confirmed experimentally in late 1954 by Wyman, who used absolute beta counting to measure a cross section of ~300 mb at the same energy. Thorpe and Wyman documented the technique in their 1958 report, LA-2235.[17]

The absolute beta counting approach offered a reliable absolute measurement of total tritium production from neutron-induced reactions on ^{7}Li as a function of energy. With a convenient half-life of ~12 years, tritium is sufficiently radioactive to be readily measurable in 0.5-g samples, and its half-life is long enough that decay corrections have minimal effect on the measurement. A special lithium target was designed and fabricated for the measurement so that under the appropriate thermal conditions its packaging would fail in a controlled manner and release the tritium. This target was irradiated with a known neutron flux and energy. The tritium was then extracted and pumped into a low-background proportional counter estimated to be 95% efficient at detecting any tritium decay in its volume.

Using this method with a combination of deuterium-deuterium and DT sources along with different accelerating potentials, total tritium production was measured at incident neutron energies of 4 to 15 MeV, as shown in Fig. 5, and compared to more traditional emulsion counting techniques. Note that the measured cross section is in fair agreement with the modern IRDFF-II (and ENDF/B-VIII.0) estimate shown in Fig. 1, which gives 380 mb for the maximum tritium production located at 8 MeV versus ~450 mb shown in Fig. 5.

By 1956, differential neutron cross sections and neutron production for the two lithium isotopes were measured at 5 to 14 MeV, as reported in LA-2643.[16] These measurements were done with nuclear track emulsion by irradiating emulsion detectors loaded with lithium and observing the resulting ionization tracks to deduce reaction rates. Using the known Q-values and masses of the

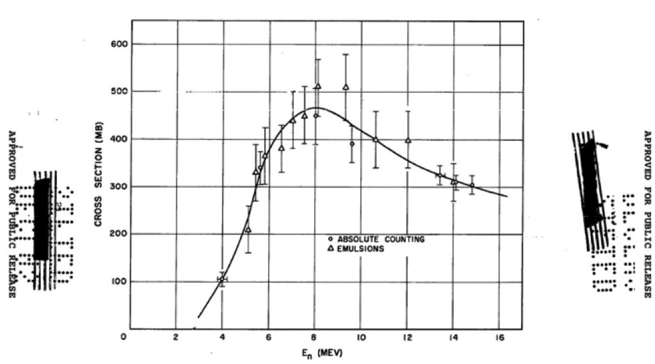

Fig. 5. Absolute tritium production cross section for ^{7}Li as a function of neutron energy (reproduced from LA-2235).

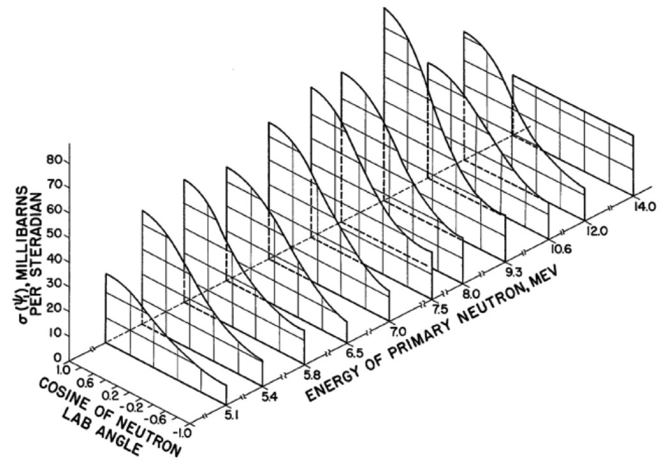

Fig. 6. Laboratory angular distributions of secondary neutrons in ^{7}Li(n,n`t) reactions (reproduced from LA-2643).

products, the reaction rates were determined as a function of energy and angle, and the outgoing neutron energy spectrum for inelastic scattering events was inferred using kinematic arguments.

Figure 6 provides an example of this work. Although these cross-section measurements were further refined in the following decades, this 1956 report contained the key measurements needed to understand the neutron cross sections of lithium at energies relevant to fission and fusion energy systems. The results reported in LA-2643 are the endpoint for this review of early cross-section measurements.

V. CASTLE BRAVO, 1954

The 1954 Castle series of nuclear tests was a landmark in the development of practical, deliverable thermonuclear devices. The first Castle shot, named Bravo, produced 15 megatons, roughly twice the yield that had been expected based on preshot calculations. Bravo remains the highest yield test in U.S. nuclear test history.

Over the years, it has been widely reported that Los Alamos scientists missed the full yield because they had not appreciated the importance of certain ^{7}Li cross sections that contributed to thermonuclear energy production. This sentiment appears in both internal Los Alamos documents and popular books on the history of the H-bomb, including Rhodes's *Dark Sun* (quoting Harold Agnew) and Tarter's *The American Lab*.[18,19] As can be seen by the previous review of early cross-section measurements and evaluations, the production of tritium via this mechanism had not been studied in detail prior to this test and the known size of the production cross section did change by a factor of more than 6 between March and July 1954.

However, recent calculations[20] with our modern Los Alamos codes do not support the claim that the poor prediction of Bravo was the result of improperly accounting for ^{7}Li nuclear cross sections. Indeed, our modern calculations show that ^{7}Li reactions did not contribute very significantly to the yield of Bravo. It is the case that the computational treatment of neutron reactions on ^{7}Li were very crude in the early 1950s, but that does not imply that this led to a large yield underprediction by a factor of 2.

After realizing that our modern calculations contradicted the oft-reported "folklore" about the role of ^{7}Li reactions in Bravo, we asked our Livermore colleagues for an independent check. Peter Rambo has run modern Lawrence Livermore National Laboratory codes on the same problem and obtained similar results to those of Los Alamos.

We are left to speculate that other deficiencies in the preshot calculations, perhaps in the material equations of state, led to the underprediction. Given the rapid nature of progress in thermonuclear weapons development in the mid-1950s, limited documentation exists explaining how the yield discrepancy was resolved at the time. The real reason for the underprediction may never be fully understood.

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Disclosure Statement

No potential conflict of interest was reported by the authors.

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