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NEUTRON SOURCE INVESTIGATIONS IN SUPPORT OF THE CROSS SECTION PROGRAM AT THE ARGONNE FAST-NEUTRON GENERATOR

by

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PREFACE

This report contains the text of a paper presented at the Consultants' Meeting on Neutron Source Properties, Debrecen, Hungary, 17-21 March 1980. This conference was sponsored by the International Atomic Energy Agency, Vienna, Austria. The manuscript was prepared under the assumption that it would be published in the conference Proceedings. However, the decision was made at this meeting to publish only the abstracts of the contributed papers in the Proceedings. Therefore, the original manuscript is presented here in its entirety.

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NEUTRON SOURCE INVESTIGATIONS IN SUPPORT OF THE CROSS SECTION PROGRAM AT THE ARGONNE FAST-NEUTRON GENERATOR*

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ABSTRACT

Experimental methods related to the production of neutrons for cross section studies at the Argonne Fast-Neutron Generator are reviewed. Target assemblies commonly employed in these measurements are described, and some of the relevant physical properties of the neutron source reactions are discussed. Various measurements have been performed to ascertain knowledge about these source reactions which is required for cross section data analysis purposes. Some results from these studies are presented and a few specific examples of neutron-source related corrections to cross section data are provided.

I. GENERAL

The Fast-Neutron Generator at Argonne National Laboratory is an 8-MeV Tandem Dynamitron accelerator which has been in operation since ~1970 [1]. This facility has been used primarily for the production of monoenergetic fast neutrons for neutron data research in support of fast-fission technology. Neutrons are produced using charged-particle reactions which are convenient for this purpose. The accelerator is routinely operated in both steady-beam and pulsed modes ($\lesssim$ 1 nanosecond to ~ several milliseconds) depending upon the experimental requirements. In some instances, experiments have been performed using the continuous neutron spectra which are produced by bombarding thick targets with charged particles.

Table I provides information about the energetics of several neutron sources which are useful in the production of fast neutrons for nuclear data research [2]. Figure 1 gives the relative yields for several of these reactions and indicates the neutron energy ranges of applicability.

No source of neutrons is truly monoenergetic. Even for a discrete transition, finite resolution of the accelerated charged particle beam, charged-particle energy loss in the target and kinematic effects all

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contribute to a broadening of neutron energy. Secondary reactions, which can compete significantly with the primary reactions at energies above their thresholds, produce either discrete groups or broad distributions of neutrons with energies differing from the primary-group energy (see Table I). As long as the yield of secondary neutrons is significantly smaller than the primary-group yield, it is usually possible to deal with the secondary neutrons by experimental methods (e.g. time-of-flight or pulse-height discrimination) or by the application of calculated corrections. When the yield of secondary neutrons begins to dominate the total yield, the reaction ceases to be useful for many applications. Figure 2 indicates the apparent useful ranges for several neutron sources from Table I and shows the regions where the neutron yield is approximately monoenergetic. The neutron-energy interval from ~8-14 MeV is a difficult one for neutron cross section investigations because of limitations in the available neutron source reactions. This fact is evident from Fig. 2, and it is also reflected in the generally poor quality of cross section data for this region (e.g. Ref. 3).

The neutron energy range < 10 MeV is of greatest concern for the work in our laboratory since it is the region of importance for fast-fission reactor applications. We have not employed any tritium targets in our laboratory since lithium and deuterium reactions have proved to be satisfactory for our requirements. Tritium targets are less convenient to use than lithium and deuterium because of the safety precautions which must be observed. The advantages of the T(p,n) reaction have not been sufficient to justify the additional experimental difficulties which would be encountered in using this reaction instead of Li(p,n) in our laboratory.

A knowledge of the neutron yield from various target reactions as a function of energy, of angular distributions, and of spectral shapes for continuous distributions are often required in data processing. Much of the requisite information on source reactions was either not available or was of questionable accuracy when we began to investigate this matter in the early 1970's. The important evaluations of source reactions by Liskien and Paulsen were not available [4,5]. We have not undertaken a comprehensive program of investigating source reactions because of the higher priority of other research goals. Nevertheless, we have made various measurements during the last decade to determine those specific properties of the Li(p,n) and D(d,n) reactions which are required for our cross section studies. These included some measurements on yield, group ratios, background, and angular distributions at the forward angles of interest for our applications, at energies relevant to our work. Portions of this work have been reported [6,7] and some of the results will be mentioned in later sections of the present paper.

The importance of correcting experimental cross sections for the effects of the other neutrons besides the primary group, when using Li(p,n) or D(d,n) sources, is indicated by the example shown in Fig. 3 and is discussed in the caption. The other-group corrections are positive in this example because ^{232}Th fission has a threshold while ^{235}U fission does not. Other examples from our work are also informative: The correction for D(d,np)D neutrons increases from a negligible level < 8 MeV neutron energy to ~ + 5% for 9-MeV neutron energy in measurements of the ratio of (n,p) activation for ^{52}Cr and ^{19}F and (n,α) activation for ^{19}F relative to ^{238}U fission. On the other hand, the corrections for ^{7}Li(p,n^{3}He)^{4}He neutrons in measurements of the ratio of ^{115}In(n,n')^{115m}In activation relative to ^{238}U fission

range from a negligible level < 3.5 MeV neutron energy to - 4% for 5.8 MeV neutron energy. The corrections in this case are negative because ^{115}In activation has a lower effective threshold than ^{238}U fission.

II. PROTON BOMBARDMENT OF LITHIUM

The lithium target assembly used in our laboratory is shown in Fig. 4 and is discussed in the caption. The target thickness for proton energies of ~1.88 MeV can be deduced from the measured neutron yield curve at threshold. We have used targets with thicknesses in the range ~2 keV to ~200 keV (at threshold) in our measurements. A knowledge of specific-energy-loss data for protons in lithium enables us to deduce the effective thickness at other energies. A comparison of measured vs. calculated resolution for primary neutrons from the $^{7}Li(p,n)^{7}Be$ reaction appears in Fig. 5 and is discussed in the caption.

Neutron production from the tantalum cup used in lithium targets is low; however, it can produce a few percent of the total yield for a low-threshold reaction when E_p is above ~5 MeV. We measured the total yield of neutrons from a clean tantalum cup using ^{235}U and a fission chamber as a flat-response detector. The results appear in Fig. 6.

Second-to-first group ratios were measured at several energies and forward angles for the Li(p,n) source using a calibrated scintillator and time-of-flight techniques. Representative results appear in Fig. 7 where comparison is made with the evaluation of Liskien and Paulsen [5]. Our data were considered in their evaluation.

We found no useful results in the literature for the $^{7}Li(p,n^{3}He)^{4}He$ breakup reaction so we performed measurements at zero degress to determine the relatve yield and spectrum shapes at a few energies. A fission chamber containing ^{235}U and time-of-flight techniques were used in this experiment. Measurements were made with 50- and 100-cm flight paths. Some of our results appear in Figs. 8 and 9. The neutron spectra can be fitted reasonably well with the empirical formula

$$N(E_n) = AE_n \left(1 - \frac{E_n^2}{E_{max}^2}\right) \exp(E_{max} - E_n) \quad , \tag{1}$$

where

A = normalization constant,

$$E_{max} = (E_1^{1/2} + E_2^{1/2})^2, \tag{2}$$

$$E_1 = E_p \frac{m_1^2}{(m_1 + m_2)^2}, \tag{3}$$

$$E_2 = \frac{m_2}{(m_1 + m_2)} \left[\frac{m_2}{(m_1 + m_2)} E_p + Q \right] \qquad (4)$$

with

m_1 = proton mass,

m_2 = ^{7}Li nuclear mass,

and

Q = reaction Q-value (-3.23 MeV).

Measurements with the scintillator and the fission detector also provided useful information on the total neutron intensity from lithium targets. Typical results are given in Table II and Ref. 6.

In time-of-flight measurements using long flight paths and scintillation detectors, one can usually distinguish between various neutron groups (e.g. Ref. 8). In other types of measurements (e.g. Refs. 9 and 10), no attempt is made to distinguish between the groups experimentally and corrections must be determined analytically. Procedures for determining other-group corrections for the Li(p,n) source have been reported (e.g. Refs. 6, 9, 11 and 12).

Scattering of neutrons from the lithium target cup produces a rather well-defined group of neutrons with lower energy than the direct neutrons. The magnitude of this scattered flux has been estimated by Monte-Carlo methods. The relative intensity is not large as is evident from Table III.

III. DEUTERON BOMBARDMENT OF DEUTERIUM

We routinely use a gas target rather than a deuterated-metal target because the former provides superior yield and resolution performance and much lower background for the energies encountered in our laboratory. Figure 10 is a schematic diagram of the target cell in current use. Details about the design and use of these targets appear in the caption.

We find that the yield and angular distribution data from an evaluation by Liskien and Paulsen [4] provide a description of the primary group which is adequate for our purposes. However, there is very little useful information available in the literature on the D(d,np)D breakup reaction. We have performed measurements at several energies and forward angles to determine the yield and spectra for the deuteron breakup reaction relative to the D(d,n)^{3}He reaction. A calibrated scintillator and time-of-flight techniques were used in this work. Figure 11 is a typical raw time-of-flight spectrum. Background neutrons from the target assembly are in evidence. Figure 12 shows a background-subtracted spectrum. The relative importance of background from the empty cell was investigated, and the results appear in Fig. 13. Figure 14 summarizes the results of zero-degree yield measurements for D(d,np)D relative to D(d,n)^{3}He. Figure 15 shows some typical zero-degree breakup spectra deduced from this work.

Neutron distributions from deuteron bombardment are rather forward-peaked and the neutron-energy spectrum varies dramatically with angle. Therefore, even more care is required in calculating source-related corrections for D(d,n)-source than for Li(p,n)-source measurements (e.g. see Ref. 7). Although the gas target assembly we have been using is relatively massive, detailed Monte-Carlo calculations have shown that the scattered-neutron flux is fairly modest and is dominated by neutrons scattered from the beam stop (Table III). Forward peaking of the direct neutron flux reduces the likelihood of scattering from other components of the target assembly.

IV. THICK-TARGET APPLICATIONS

Routinely, relatively thick targets of lithium ($\lesssim$ 200 keV) have been used to produce pseudo "white-source" neutron fields for total and capture cross section studies at low energies < 500 keV (e.g. Ref. 13); occasionally, they have been used for similar measurements at higher energies. Energy analysis is accomplished using time-of-flight methods.

We have also found that the bombardment of "infinitely thick" targets of lithium or beryllium with deuterons (the beam stops in the target material) provides intense fast-neutron fields which are useful for certain applications (Table I). One application is the activation of research samples to levels high-enough to permit coincidence measurements to be performed. We have made comparisons of total neutron yield for several sources using a flat response detector. Some of our results appear in Table II. For E_d = 7.6 MeV, the yield from the Be(d,n) reaction (the beam stops in the target) is a factor of 200 larger than from a gas target containing 4 atm-cm of deuterium, when a comparison is made at zero degrees. Figure 16 shows raw time-of-flight spectra for very-thick Be and Li targets at E_d = 7.6 MeV. These sources produce broad neutron-energy distributions with $\bar{E}_n \approx 0.4E_d$ [14].

V. CONCLUDING REMARKS

Our investigations of the Li(p,n), D(d,n), Be(d,n), and Li(d,n) reactions have provided the information we need for our current research programs, but these results do not begin to satisfy the general need for a comprehensive investigation of the source reactions commonly used for fast-neutron cross section research. More detailed angular distribution measurements are needed to reduce the uncertainties in experiments where the relative neutron yield away from zero degrees is of considerable importance. Systematic measurements of breakup neutron spectra and yield, relative to the primary group, should be performed as a function of energy and angle. Although the Be(d,n) and Li(d,n) reactions are under investigation at higher energies ($E_d \geq$ 10 MeV) as required for fusion and medical research applications [14], there has not been much effort devoted to developing these source reactions for nuclear data studies at lower energies.

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TABLE I.

Several Useful Neutron Sources: Energetics

1. Li + p

Reaction	Q-Value (MeV)	Threshold (MeV)
$^{7}Li + p \rightarrow n + {}^{7}Be$ (gnd. state)	-1.644	1.881
$^{7}Li + p \rightarrow n + {}^{7}Be^{*}$ (1st state)	-2.079	2.380
$^{7}Li + p \rightarrow n + {}^{7}Be^{**}$	-6.18	7.06
$^{7}Li + p \rightarrow n + {}^{3}He + {}^{4}He$	-3.23	3.68
$^{6}Li + p \rightarrow n + {}^{6}Be$	-5.07	5.92
$^{6}Li + p \rightarrow n + p + {}^{5}Li$	-5.67	6.62

2. D + d

Reaction	Q-Value (MeV)	Threshold (MeV)
$D + d \rightarrow n + {}^{3}He$	+3.268	0
$D + d \rightarrow n + p + D$	-2.225	4.45
$d + d \rightarrow 2n + 2p$	-4.45	8.90

3. T + p

Reaction	Q-Value (MeV)	Threshold (MeV)
$T + p \rightarrow n + {}^{3}He$	-0.765	1.020
$T + p \rightarrow n + p + D$	-6.258	8.342
$T + p \rightarrow 2n + 2p$	-8.483	11.31

4. T + d

Reaction	Q-Value (MeV)	Threshold (MeV)
$T + d \rightarrow n + {}^{4}He$	+17.639	0
$T + d \rightarrow 2n + {}^{3}He$	-2.990	4.98
$T + d \rightarrow 2n + p + D$	-4.653	7.75
$T + d \rightarrow 3n + 2p$	-10.708	17.84

5. Be + d

Principal Reaction	Q-Value (MeV)	Threshold (MeV)
$^{9}Be(d,n)^{10}B$	+4.4	0

6. Li + d

Principal Reaction	Q-Value (MeV)	Threshold (MeV)
$^{7}Li(d,n)^{8}Be$	+15.0	0

TABLE II.

Comparison of 0° Neutron Yields[a]

Target, Beam	Principal[b] Reactions	Yield[c] (neutrons/sr/μ Coulomb)
200-keV[d] natural lithium	$^7Li(p,n)^7Be$ $^7Li(p,n)^7Be^*$ $^7Li(p,n^3He)^4He$	7.0(7) for E_p = 2.3 MeV[e] 1.2(7) for E_p = 7.6 MeV
2-cm long cell with 2 atm of D_2 at 30°C	$D(d,n)^3He$ $D(d,np)D$	1.4(8) for E_d = 7.6 MeV
Thick beryllium metal	$^9Be(d,n)$ All channels	3.6(10) for E_d = 7.6 MeV
Thick lithium metal	$^7Li(d,n)$ All channels	3.0(10) for E_d = 7.6 MeV

[a] Measured with a ^{235}U fission detector.

[b] Minor source and target component reactions not listed.

[c] All neutron energies

[d] Measured at threshold energy E_p = 1.881 MeV.

[e] Maximum yield at resonance in $^7Li(p,n)^7Be$.

Table III.

Ratio of Neutrons Scattered from Target Beam Stop to the Unscattered Neutrons at 0°[a]

Lithium target:		Deuterium-gas target:	
E_p (MeV)	Ratio (x100)	E_d (MeV)	Ratio (x100)
1.95	1.86	2.0	1.05
2.05	2.71	3.0	0.71
2.15	2.08	4.0	0.43
2.3	0.89	5.0	0.35
2.7	1.02	6.0	0.29
3.7	1.30	7.0	0.26
4.7	1.13		
5.7	1.28		

[a] Beam stop is 0.0254-cm tantalum for each target.

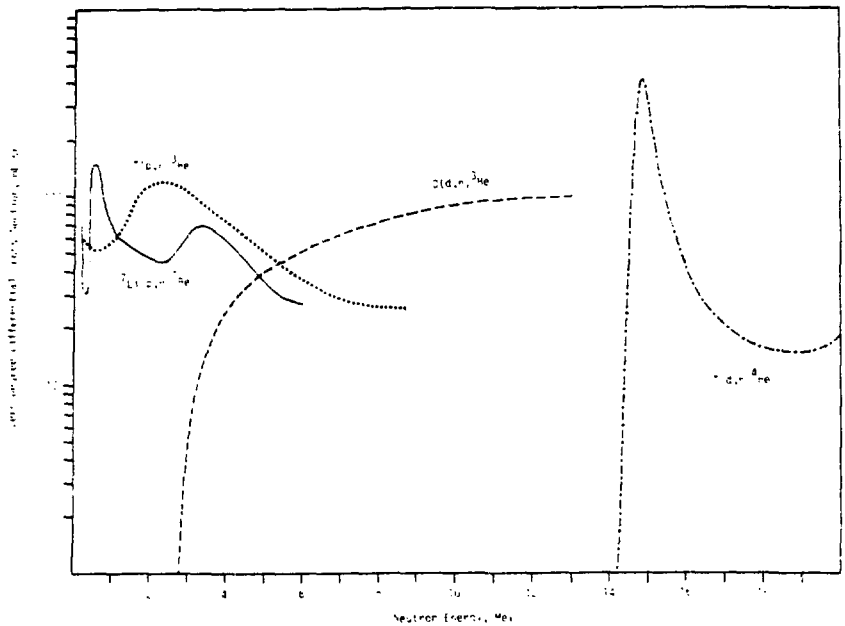

Fig. 1. Neutron yields vs. neutron energy for several important monoenergetic neutron source reactions. (see Ref. 15).

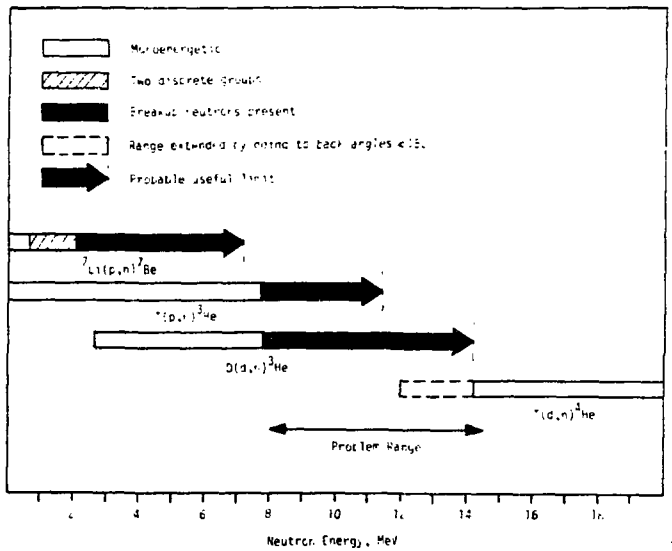

Fig. 2. Approximate usable energy ranges for neutron source reactions from Fig. 1. Regions where secondary neutrons appear are indicated.

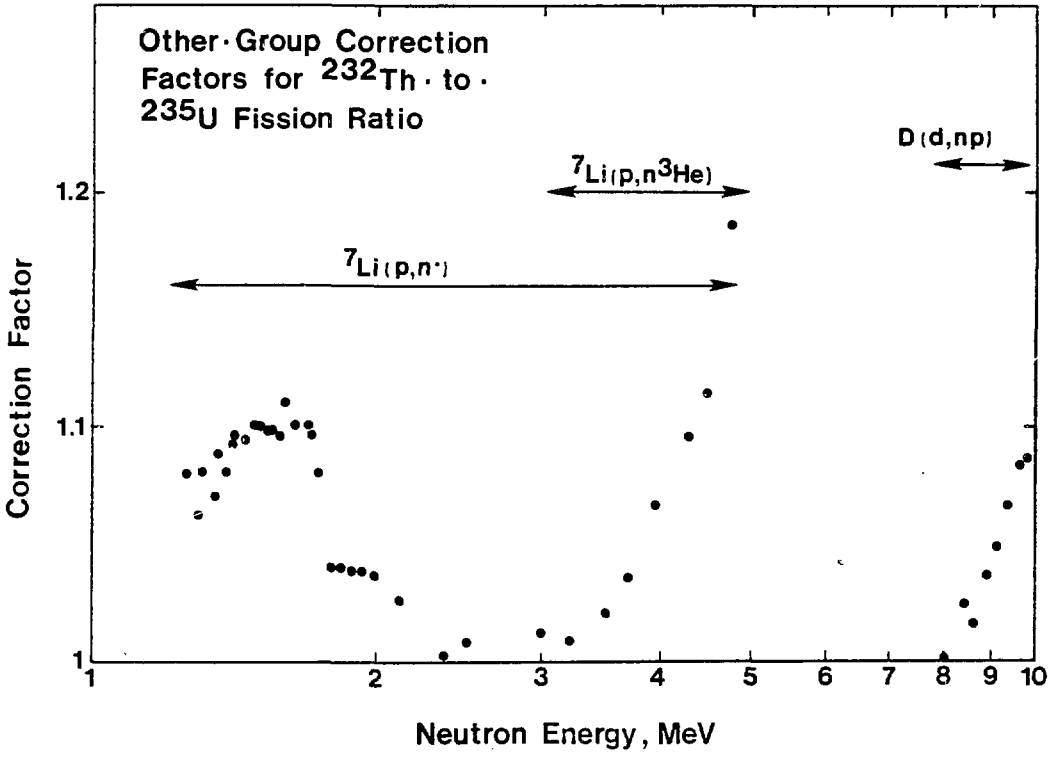

Fig. 3. Calculated corrections for the effect of secondary neutrons in the measurement of ^{232}Th to ^{238}U fission cross section ratios are presented. At energies below 5 MeV, the experiment utilizes proton bombardment of natural lithium as a neutron source. The corrections for second-group neutrons from the $^{7}Li(p,n)^{7}Be^{*}$ reaction are significant below 2.5 MeV. From 2.5-5 MeV, both the first- and second-group neutron energies fall in a region where ^{232}Th and ^{235}U fission cross sections do not vary rapidly with energy. Therefore, the corrections are very small. Above 3 MeV, the corrections for neutrons from $^{7}Li(p,n^{3}He)^{4}He$ reactions are significant. Deuteron bombardment of deuterium is used as a neutron source above 5 MeV, however the corrections for secondary neutrons from the D(d,np)D reaction are negligible below 8 MeV. The scatter of the points is caused by structure in the $^{232}Th(n,f)$ cross section and by the spacing of the grid used in calculating the corrections.

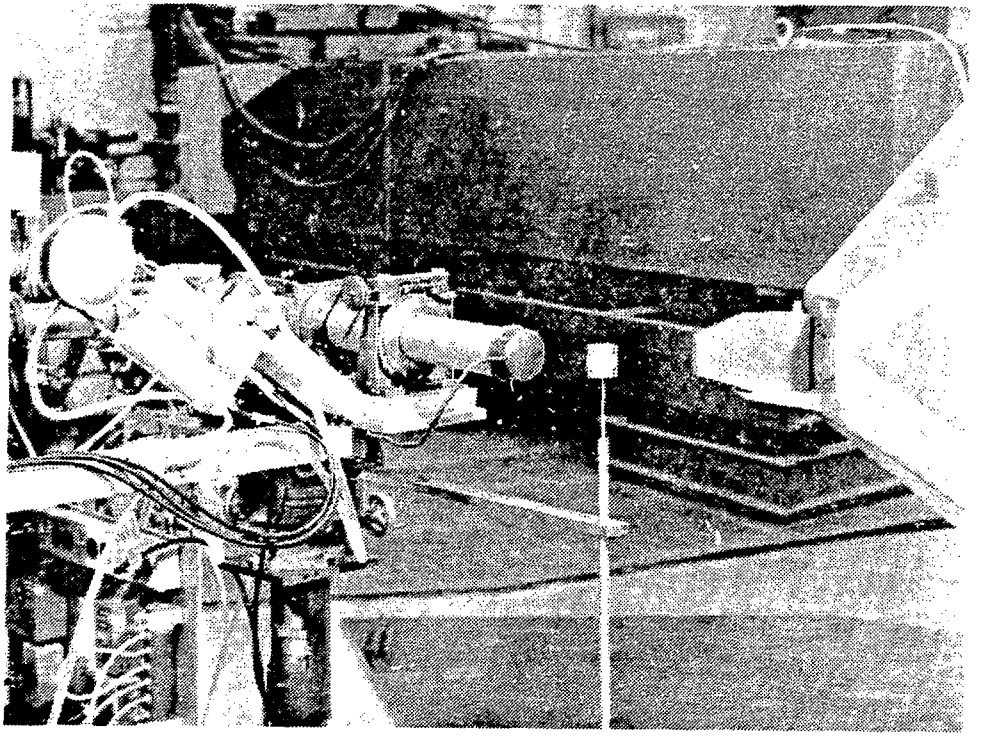

Fig. 4. Lithium targets are prepared by evaporating a thin metallic-lithium film onto a cup which is fabricated from 0.0254-cm thick tantalum sheet. This cup is mounted at the end of a flight tube on a mechanism which continuously moves the cup in such a way that the collimated proton beam traces a circular path on the lithium film. The combination of this motion and air-jet cooling enables targets to dissipate as much as 100 watts/cm^2 of beam power for several hours without serious degradation of the lithium film. This photograph shows the apparatus in the vicinity of the target for an experimental station which is used in gamma-ray production cross section measurements.

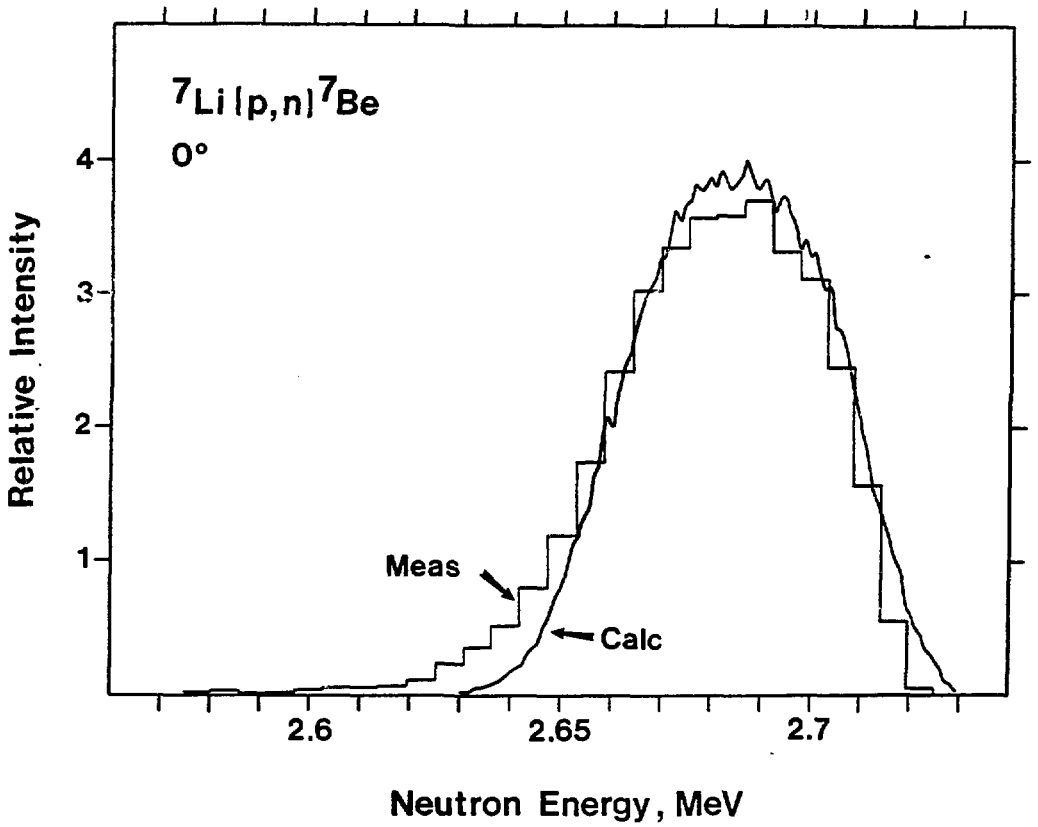

Fig. 5. The measured and calculated first-group neutron energy profile for a nominal 100-keV lithium target are compared. Measurements were made using a scintillator at 40 meters and pulsed-beam time-of-flight techniques. The calculation took into consideration the energy dispersion of the proton beam from bunching, the specific energy loss of protons in lithium metal, and proton straggling effects. The proton beam used in the measurements was not magnetically analyzed. It is speculated that the difference between the calculated and measured spectra is due to an energy tail on the proton beam which was not specifically identified or taken into consideration in the analysis. Approximately 5% of the neutron yield appears in the tail.

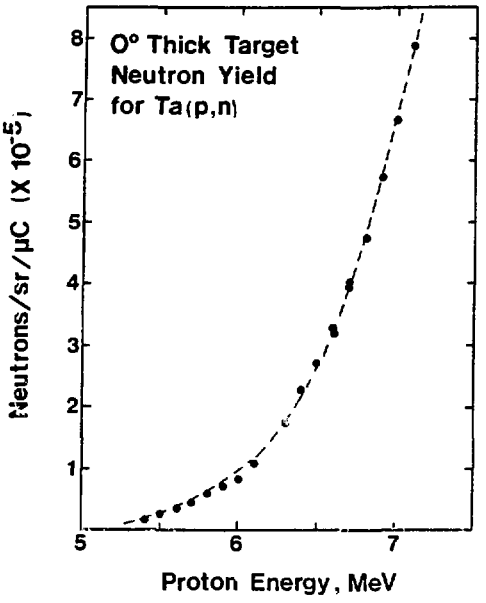

Fig. 6. Neutron yield from the tantalum target backing.

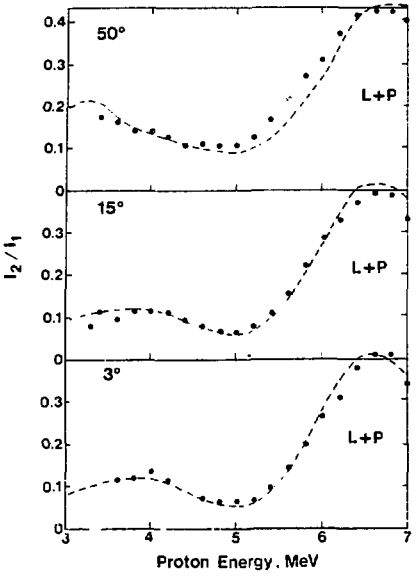

Fig. 7. Group ratios for the $^{7}Li(p,n)$ reactions. Some results from our measurements are compared with the Liskien and Paulsen evaluation (Ref. 5).

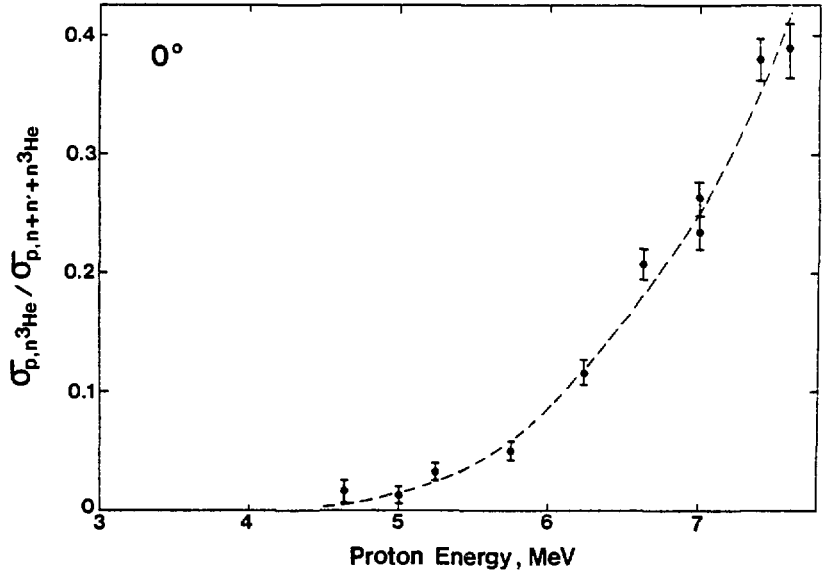

Fig. 8. Measured values for the fraction of all neutrons from proton bombardment of lithium which can be attributed to the reaction $^{7}Li(p,n\ ^{3}He)^{4}He$.

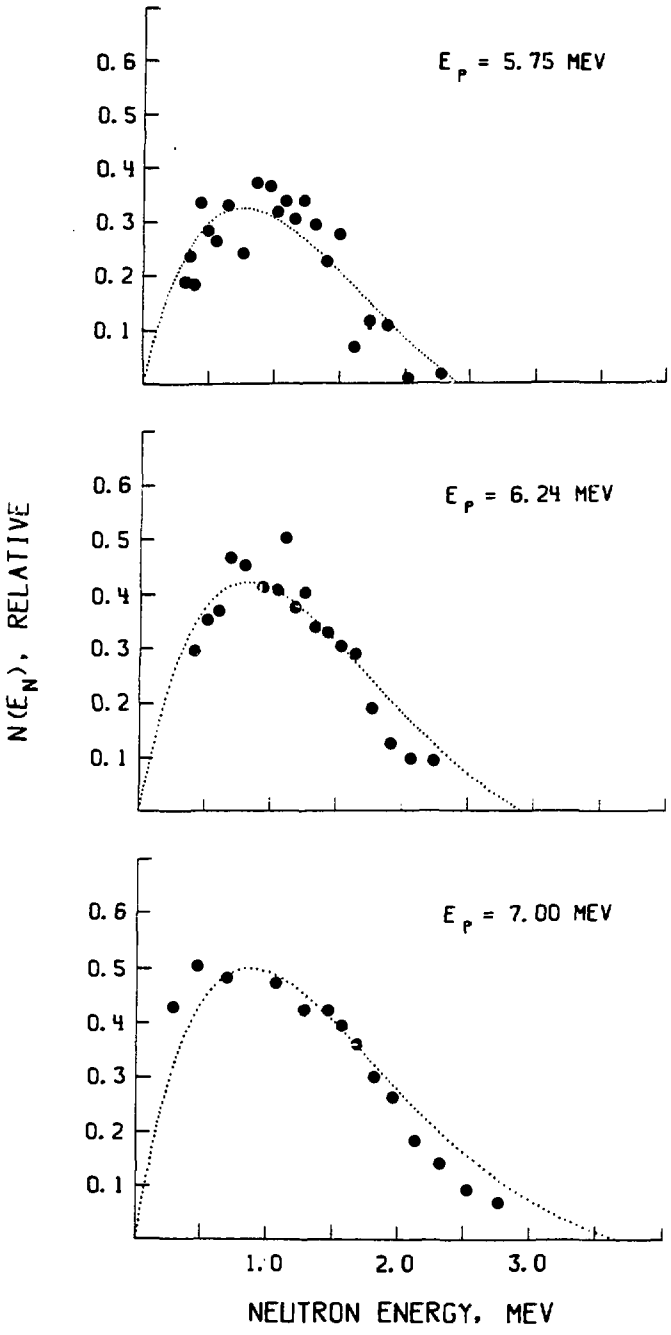

Fig. 9. Examples of measured neutron energy spectra for the reaction $^{7}Li(p,n\ ^{3}He)^{4}He$. Curves are best fits of Eq. (1) to the data.

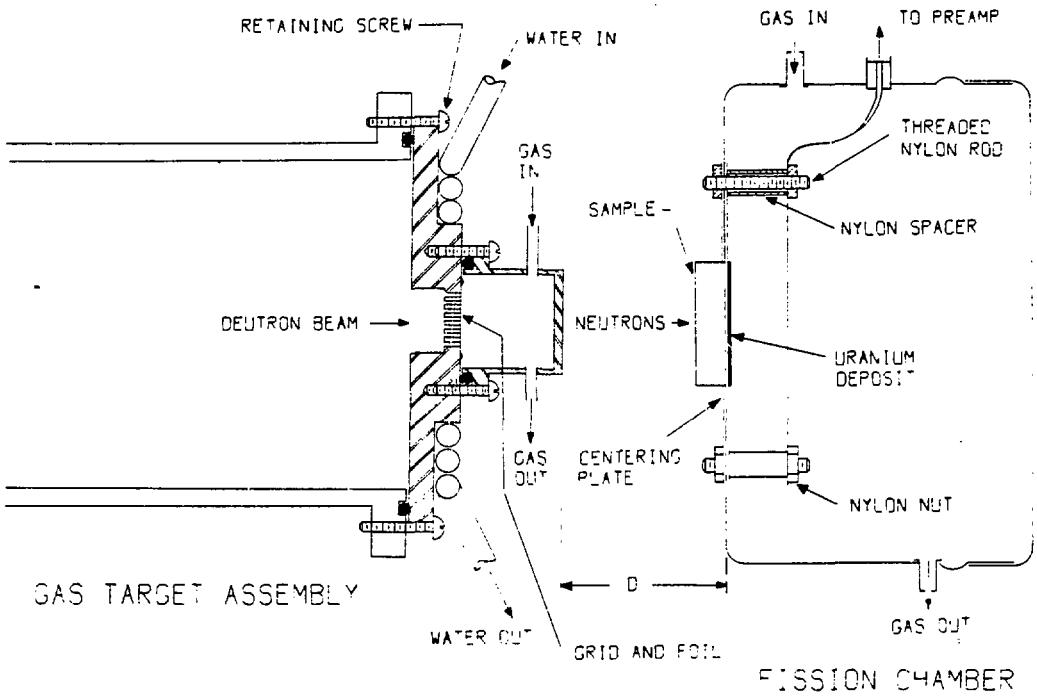

Fig. 10. Schematic diagram of the target apparatus used for neutron production from deuteron bombardment of deuterium gas. Apparatus used for measurement of activation cross sections relative to standard fission cross sections is also shown. The target has a water-cooled copper plate attached to the end of a beam tube. A gold grid with about 75% transmission is imbedded into the center of this plate. Thin nickel metal foils (2-3 mg/cm^2) are fastened to the grid plate with epoxy cement to provide a vacuum seal as well as a thin entrance window to the gas cell. The deuteron beam is stopped by a gold or tantalum end cap to the cell. The entire target cell is cooled by an air jet as well as by water circulating in cooling coils. During runs, the cell is pressurized with deuterium to 2 atm. The cell can be evacuated in order to measure background from (d,n) reactions on the nickel window and other metallic components of the target and, especially, from carbon and nitrogen contamination which tends to accumulate on the target. These contaminants provide a source of background which increases steadily with usage of the target. As is the case for all thin-window targets, this type of target cannot be used once the nickel foil is punctured. The targets can be recycled by dissolving the epoxy in an organic solvent, cleaning off carbon residue, and installing new foils.

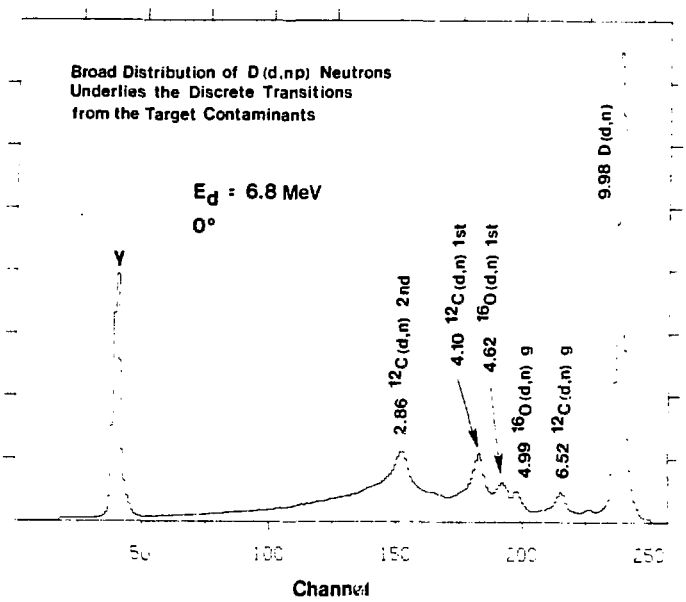

Fig. 11. Raw time-of-flight spectrum from a deuterium-gas target as measured with a scintillator at 7.4 meters. Position of the gamma-ray peak is determined by electronic considerations.

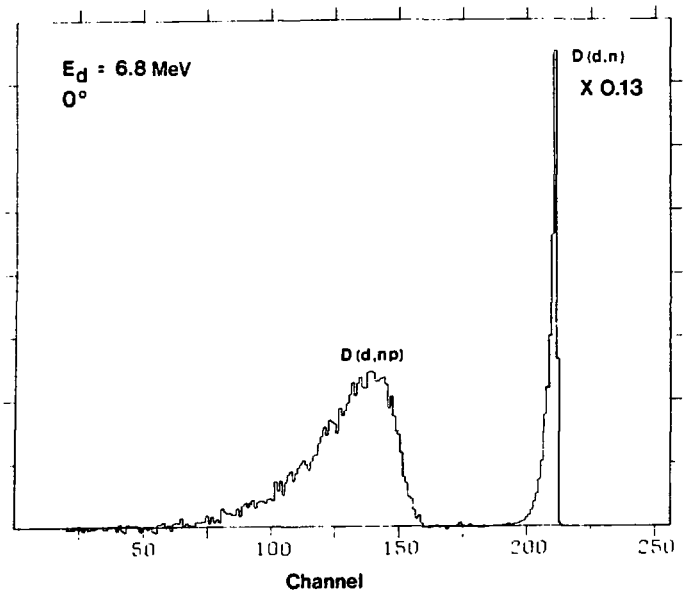

Fig. 12. Time-of-flight spectrum for neutrons from a deuterium-gas cell with all background subtracted.

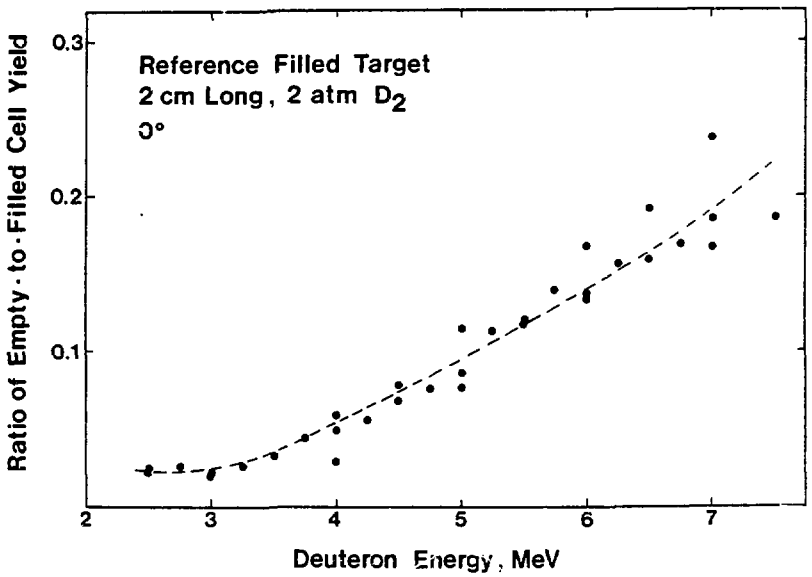

Fig. 13. Measured ratio of background to total neutron yield from a deuterium-gas target. This curve is only an indication of the relative importance of the background. Since the background depends upon the condition of the target, it must be measured periodically during experiments.

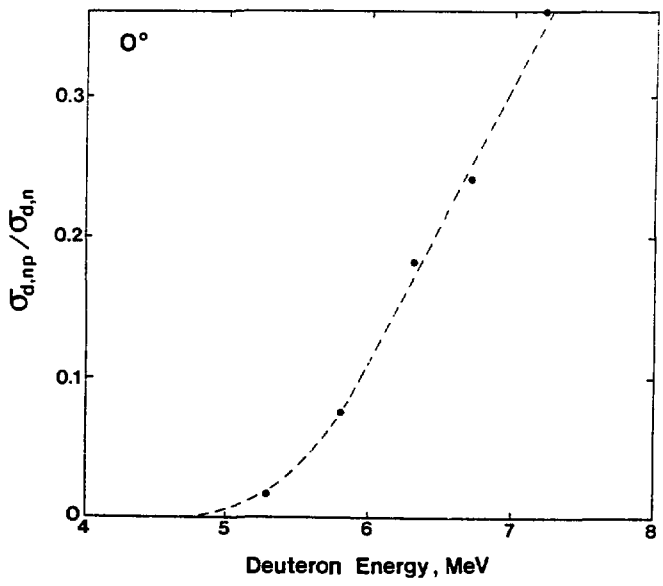

Fig. 14. Measured ratio of (d,np)-to-(d,n) neutron yield from deuteron bombardment of deuterium.

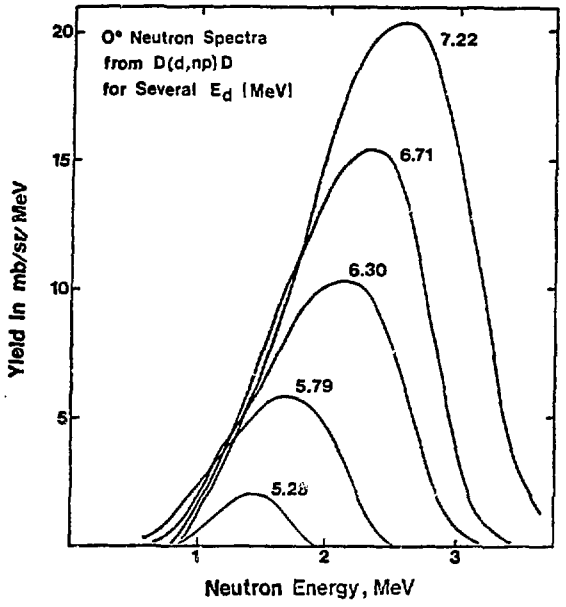

Fig. 15. Continuous neutron distributions from the breakup of deuterium by deuteron bombardment. Data deduced from time-of-flight measurements.

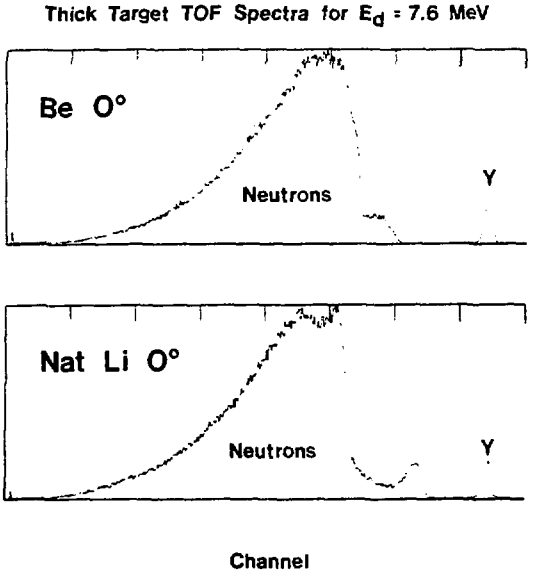

Fig. 16. Raw time-of-flight spectra produced by stopping 7.6-MeV deuterons in thick beryllium lithium targets. Measurements were performed with a scintillator at 5 meters.