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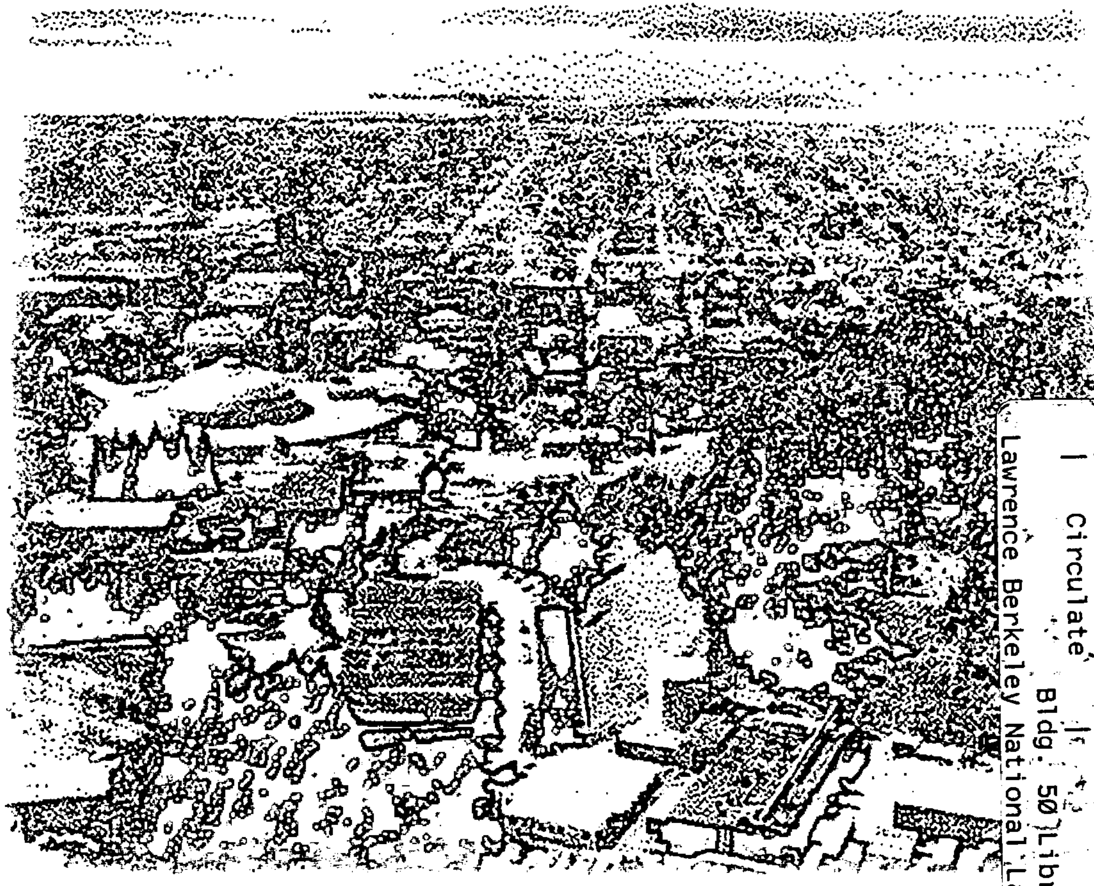

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Measurements of low-energy (d,n) reactions for BNCT

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Abstract

Neutron yields and energy spectra have been measured for various deuteron-induced reactions at low energy. The main features of these reactions are presented and discussed with regards to their potential use as a source of epithermal neutron beams for Boron Neutron Capture Therapy (BNCT). Among the studied reactions, the ^{13}C(d,n)^{14}N presents features potentially interesting for an accelerator-based neutron source for BNCT.

Keywords: Boron Neutron Capture Therapy (BNCT); Accelerator-based neutron source; Neutron-producing reactions.

1. INTRODUCTION

A renewed interest in Boron Neutron Capture Therapy (BNCT)[1] has recently prompted searches for high quality, easily available and cost-effective neutron sources. Although different materials and designs of moderator and filter assemblies have led to improvements in the quality of epithermal neutron beams produced at nuclear reactors, an accelerator-based neutron source may, in the near future, constitute a more attractive solution for the production of neutron beams with the intensity and spectral purity necessary for cancer therapy. In fact, with a suitable choice of the neutron-producing reaction, it may be possible to produce epithermal neutron beams with an energy distribution narrowly centered around the optimal value for the treatment of deep-seated tumors. In particular, relative to reactor-based sources, epithermal neutron beams produced with accelerators may be affected by a smaller contamination of high-energy neutrons ($E_n >\sim 25$ keV). Because of the large dose delivered to the skin and the brain surface by recoil protons, such neutrons pose severe constraints on the dose that can be delivered to the tumor and their presence in the therapeutic beam should therefore be minimized. Finally, fewer safety problems associated with radiation hazards may facilitate the installation of BNCT facilities in metropolitan areas, and the relatively low costs associated with the construction and maintanence of an accelerator-based source may allow for a wider diffusion of such therapy in many medical centers.

An accelerator-based neutron source relies upon a proton- or deuteron-induced reaction on a suitably chosen target. The choice of the primary beam energy and of the target is mainly dictated by the requirement of a high yield of low-energy neutrons, although economic considerations may also play a role in the choice of the most convenient reaction. The size and cost of the accelerator can be minimized by reducing the energy of the primary beam, while targets with good mechanical and thermal properties may result in less complicated and less costly cooling systems. Depending on the melting point of the target, a sophisticated cooling system may in fact be necessary to dissipate the large amount of power associated with the high beam currents required.

Currently, two reactions are being considered for use in the first generation of accelerator-based neutron sources for BNCT: the ^{7}Li(p,n)^{7}Be reaction at an energy around 2.5 MeV[2–4], and the ^{9}Be(p,n)^{9}B at energies approaching 4 MeV[5–7]. Both reactions produce neutrons with adequate energy and yield, but their use is complicated by the low melting point of the Li target and the production of the radioactive ^{7}Be residue for the Li(p,n) reaction, and the high proton energy and poorer neutron beam quality for the Be(p,n) reaction. In order to investigate whether alternative reactions exist for neutron production in BNCT, we have performed measurements of several (d,n) reactions. To restrict the investigation to low-energy deuteron beams, which could be produced with relatively small and inexpensive accelerators, we have concentrated on reactions characterized by a positive or slightly negative Q-value. Furthermore, Coulomb barrier considerations limit the target choice to light elements. Finally, the requirement of stable, mechanically and thermally convenient materials restricts the number of potentially useful targets to essentially ^{6}Li, ^{9}Be, ^{10}B, ^{12}C and ^{13}C. Among the different reactions, the ^{9}Be(d,n)^{10}B and the ^{12}C(d,n)^{13}N have recently been investigated for their potential use in BNCT (see for example ref.[8]), although the energy and angular distributions of the emitted neutrons are not well determined. On the other hand, the ^{13}C(d,n)^{14}N reaction has not yet been considered as a low-energy neutron source for BNCT, although Brune et al.[9] recently reported on the large cross-section that characterizes this reaction, which could make it interesting for several applications that require intense neutron sources.

In this paper we present measurements of energy and angular distributions for the potentially interesting (d,n) reactions. Differential and total yields have been measured for thin and thick targets, with the aim of providing a basis for further evaluation of epithermal neutron production from an accelerator-based source for use in BNCT. In sect. II the experimental method is described. The energy and angular distributions of the neutrons produced in the different reactions are presented and discussed in section III. A summary and conclusions are given in sect. IV.

2. EXPERIMENTAL METHOD

The measurements were performed at the 88-Inch Cyclotron of the Lawrence Berkeley National Laboratory. Deuteron beams of 1.5 MeV, and proton beams of 2.5 MeV were obtained by accelerating D_3^+ and H_3^+ molecular beams from the Electron Cyclotron Resonance source. To minimize in-scattering, the targets were positioned inside a thin-walled scattering chamber, 30 cm in diameter, mounted at the end of a long beam-line far from magnets or other heavy material. A plungable Faraday Cup was mounted inside the chamber to measure the integrated beam current. For thick target measurements, the total charge of the beam stopped in the target was measured. However, since no electron suppression was implemented, the measured charge in the thick target measurements was only used for relative normalization purposes. To estimate the uncertainty in the absolute yield measurements, the ^{7}Li(p,n)^{7}Be reaction at 2.5 MeV of incident energy was also measured with the present setup. The overall uncertainty, associated with the integrated beam charge as well as with the efficiency, was estimated from the Li(p,n) reaction to be of the order of 30 %.

Neutrons were detected by means of five BC501A liquid scintillator cells 12.7 cm in diameter and 5 cm thick, read out by an XP2410 Phillips phototube. The detectors were mounted at polar angles of 0, 30, 60, 90 and 140 degrees, on alternate sides of the beam axis, at a distance of 50 cm from the target. The neutron energy was determined from the time-of-flight (ToF) relative to the Cyclotron radiofrequency (RF) signal. For an RF period of $\sim$ 100 nsec, the choice of a 50 cm distance allowed the measurement of neutrons with energy as low as 100 keV, although it also resulted in a poor energy resolution for high-energy neutrons. For an overall time resolution of $\sim$ 4 nsec, as measured from the prompt gamma-rays peak (see fig. 1 for example), the energy resolution goes from ± 6 keV for 0.1 MeV neutrons, to ± 1 MeV for 5 MeV neutrons.

To suppress environmental gamma-ray background, the scintillator cells were surrounded by a 3 mm thick lead sheet. Background from scattered neutrons was minimized by positioning the detectors at a minimum distance of 1.5 m from the walls or other scattering material (the beam height relative to the floor is 120 cm). Furthermore, the small distance from the target helped to minimize the contamination of scattered neutrons, because of solid angle as well as time-of-flight considerations. Together with the ToF, the light output and a standard pulse shape discrimination time for each event were recorded. More details on the detectors and their efficiency can be found in ref.[10]. An estimated threshold of ~ 10 keV electron equivalent (keVee) was maintained during the measurements, so as to detect neutrons of energy as low as 100 keV. Such a low threshold on the neutron energy is fundamental, when studying neutron production for BNCT, since a significant contribution to the epithermal neutron beam can originate from these low-energy neutrons.

3. RESULTS

A. ^{9}Be(d,n)^{10}B reaction

Guzek et al.[11] have recently investigated the ^{9}Be(d,n)^{10}B reaction at a deuteron energy slightly above 1 MeV as a possible intense neutron source. In particular, although the Q-value of this reaction is quite large, $Q = 4.36$ MeV, the authors reported the presence of a strong neutron peak at an energy well below 1 MeV. The absolute yield of low-energy neutrons, and the amount of contamination of high-energy neutrons in the spectrum are, however, still somewhat unclear.

Fig. 1a shows the background-subtracted ToF spectrum measured at 0^{o} for the ^{9}Be(d,n)^{10}B reaction at E_d=1.5 MeV. The solid and dashed histogram represent the results for the thin (0.8 mg/cm^{2}) and thick targets respectively (the thin target corresponds to an energy loss of the d beam of ~ 200 keV). Together with the gamma peak, two neutron peaks are observed in the spectrum. The intense peak at large ToF corresponds on average to 400 keV neutrons, as evident from the efficiency-corrected energy distribution of fig. 1b. The very low energy and apparently high yield that characterizes this peak, as well as the low energy of the primary beam and the good mechanical and thermal properties of the

Be target, have led Guzek et al. to suggest this reaction as a potential neutron source for BNCT[11]. However, together with the 400 keV peak, a high-energy peak is evident in the ToF spectrum. The energy of this peak is estimated to be centered around 3.6 MeV, which corresponds to the excitation of the 2.15 MeV level in the ^{10}B residue. Although this peak has not been reported in ref.[11], previous works[12,13] have shown the existence of a significant high-energy contamination, which may make it very difficult to produce with this reaction epithermal neutron beams with intensity and spectral characteristics adequate for BNCT.

As is clear from the present measurements, the amount of contamination depends on the target thickness. In particular, after correcting for the efficiency, the high-energy neutrons have been estimated to constitute about 38 % of the total yield for the thin target case. For the thick target, the contamination increases to approximately 50 %, as expected from energy considerations (the excitation of the 5.1 MeV neutron group becomes energetically impossible for deuteron energies below $\sim$ 1 MeV). Although a bombarding energy dependence of the high-energy neutron yield cannot be excluded, it does not seem likely that it will decrease to a value sufficiently low to allow for the use of this reaction in BNCT. Furthermore, both the 0^o and the total yield, as evident from the angular distribution of fig. 1c, do not justify a particular interest in this reaction as a potential neutron source for accelerator-based BNCT. For higher deuteron energies, the rapid increase of the cross-section is offset by a corresponding increase of the average neutron energy, which leads to a further worsening of beam quality after moderation, thus making this reaction interesting only for applications that do not require high-quality epithermal neutron beams.

B. ^{12}C(d,n)^{13}N

The slightly negative Q-value that characterizes this reaction (Q = -0.28 MeV), combined with the good mechanical properties of carbon and, expecially, with its extremely high melting point, make this reaction worth investigating for use as a low-energy neutron source. A deuteron energy of 1.5 MeV may represent a reasonable compromise between the need of minimizing the neutron energy, and that of maximizing the cross-section which is strongly influenced, at low incident energy, by the Coulomb barrier. Fig. 2a shows the energy distribution measured for this reaction at 0°. A clean, low-energy neutron emission is observed, peaked at 500 keV, as expected from energy considerations. The angular distribution, shown in fig. 2b, is characterized by a minimum at 0°, and by an increase for angles greater than 90°, a behavior consistent with previously reported measurements at slightly lower incident energy[14,15]. Although it may be possible to setup a neutron beam line at angles different from the primary beam direction (which might lead to other advantages, such as multiple treatment rooms, or a symmetric dose monitoring room), the yield at all angles for this reaction seems too small for any practical application in BNCT. Preliminary estimates indicate that an epithermal neutron beam of the necessary intensity could only be obtained at this beam energy with a deuteron beam current of several hundred mA. A higher deuteron beam energy may result in a significantly higher neutron yield, even though such an increase does not necessarily result in a higher epithermal neutron flux, since increasing the energy of the primary beam would also lead to an increase of the neutron energy. We have not investigated further this possibility, but it seems unlikely that this reaction will constitute a viable alternative to the higher energy proton reactions, and in particular to the 2.5 MeV Li(p,n), as also indicated in ref.[8].

C. ^{13}C(d,n)^{14}N

Among the deuteron-induced reactions at low incident energy, the ^{13}C(d,n)^{14}N is certainly one of the most interesting because of the previously mentioned advantages of carbon as target material, the stable reaction residue and, especially, the large cross-section that characterizes it. As reported in a recent study[9], a cross-section of 300 mb, slightly smaller than the one for the 2.3 MeV Li(p,n) reaction, is observed even for deuteron energies as low as 1 MeV. However, neither the energy nor the angular distribution of the neutrons have been reported for this reaction. Because of the large, positive Q-value (Q = 5.33 MeV), the neutron energy cannot be easily estimated, since the ^{14}N residue can be left in several excitation energy levels[16]. In order to check the applicability of this reaction to accelerator-based BNCT, we have measured the neutron energy distribution and yield at various angles for this reaction. The ToF spectrum measured at 0^o for a thick target, at a deuteron energy of 1.5 MeV, is shown in fig. 3a. An abundant low-energy neutron production is observed. The main contribution can most probably be associated with the 5.83 MeV level in the ^{14}N residue. Higher energy peaks are also observed in the figure, which can be associated with the 5.32 and 2.31 MeV levels. Fig. 3b shows the efficiency-corrected energy spectrum at 0^o.

For this reaction, a large fraction of the neutrons are emitted with an energy below 1 MeV. The overall fraction of neutrons with $E_n > 1$ MeV is estimated to be ~ 30 %. However, most of those neutrons are concentrated in a peak of energy slightly larger than 1 MeV, with only 6 % of the neutrons emitted with energy greater than 2 MeV (the energy resolution prevents for an accurate determination of the high-energy neutron peaks). Fig. 3c shows the angular distribution for the ^{13}C(d,n)^{14}N reaction at E_d = 1.5 MeV. The solid symbols represent the total yield, while the open symbols refer only to the low-energy component ($E_n < 1$ MeV). The yield of low-energy neutrons decreases as the angle increases, while a sizable yield of the high-energy component survives at all angles. As a consequence, a large fraction of neutrons emitted at angles greater than 60^o have energy in excess of 2 MeV. A complete simulation of the reflection and moderation process has to be performed in order to study the effect of these neutrons on the final dose distribution. In fact, the presence of higher energy neutrons at large angles does not necessarily lead to a worsening of the epithermal beam quality, and may result in more therapeutic neutrons being produced with useful energy for a proper choice of the reflector and moderator assembly.

By interpolating and integrating the angular distribution of fig. 3c, we estimate the total neutron yield for the ^{13}C(d,n) reaction at E_d=1.5 MeV to be $\sim 1.9 \cdot 10^8$ neutrons/μC. This value is ~ 30 % lower than the total yield estimated from the cross-section of ref.[9], which is $\sim 2.4 \cdot 10^8$ neutrons/μC. Although the discrepancy is comparable to the uncertainty in our absolute normalization procedure, the presence of a very low-energy emission

(E_n <0.1 MeV), to which the present setup was insensitive, cannot be excluded. While further measurements are needed in order to estimate the neutron yield with higher accuracy, the present results suggest that the $^{13}C(d,n)^{14}N$ reaction could be potentially interesting as a low-energy neutron source. The optimal neutron energy has not been investigated, and it cannot be excluded that higher incident energy may be more appropriate in terms of neutron yield or energy spectra. Preliminary calculations of the moderator process indicate that for deuteron energy of 1.5 MeV a beam current of less than 100 mA is required to keep the treatment time below one hour, with a beam quality superior to currently available reactor beams (although inferior to neutron beams that can be produced with the Li(p,n) reaction). Such high intensity beams may be within reach of the accelerator technology currently being developed[17–19], with the clear advantage of a low primary beam energy and convenient target material. More accurate experimental data are needed in order to access the applicability of this reaction to the production of clinically useful epithermal neutron beams for BNCT.

4. SUMMARY AND CONCLUSIONS

The results of the present measurement are summarized in table I. The total yield and the contamination from high-energy neutrons ($E_n > 1$ Mev) for the measured (d,n) reactions are reported in the table and compared with the prediction for the Li(p,n) reaction and with experimental data on the Be(p,n) reaction at 2.5 MeV. A comparison between the neutron angular distribution for the different reactions is shown in fig. 4.

Among the different reactions, only the $^{12}C(d,n)^{13}N$ and the $^{9}Be(p,n)^{9}B$ reactions at E_p <3 MeV present a spectral purity of low-energy neutrons comparable to the Li(p,n) reaction, but their yield seems too low for currently available accelerator technology. On the other hand, both the $^{9}Be(d,n)^{10}B$ and the $^{13}C(d,n)^{14}N$ reactions present reasonably high yields, but are affected by a sizable contamination of high-energy neutrons. However, while neutrons of several MeV energy are emitted in the $^{9}Be(d,n)^{10}B$ reaction, most of the contamination for the ^{13}C targets is concentrated at neutron energies slightly above 1 MeV, so that the quality of the epithermal neutron beam may not be significantly worsened by the presence of this higher energy peak.

In conclusion, the low-energy (d,n) reaction investigated here produce neutrons with lower yield and, in most cases, higher average energy than the 2.5 MeV ^{7}Li(p,n) reaction. However, our results suggest that, with a proper choice of the deuteron energy and moderator design, a more cost-effective accelerator facility might result from the use of the ^{13}C(d,n)^{14}N reaction which represents, in our opinion, the only potentially interesting alternative for neutron production for BNCT among the different (d,n) reactions studied so far. More accurate measurements and detailed simulations of the moderation process are however necessary to estimate the intensity of the deuteron beams needed to produce the flux of epithermal neutrons necessary for the therapy.

Because of the limited resolution for high-energy neutrons, and the uncertainty on the absolute yield, the present results are by no means conclusive, and are meant to stimulate further discussion and measurements necessary to identify, if any, proton or deuteron induced reactions that would lead to simple and inexpensive accelerator-based neutron sources while satisfying all of the dose requirements for BNCT.

The authors wish to thank the 88-Inch accelerator crew for their endless effort in the difficult task of providing quality beams of unusually low energy. In particular, thanks are due to Dan Xie for setting up the AECR injector of the D_3^+ and H_3^+ molecular beam, to Aran Guy for tuning the cyclotron, and to Dave Clark for the excellent job in minimizing the time resolution of the beam.

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TABLES

Table 1. Summary of the angle-integrated yield, average energy and level of high-energy contamination, measured for different deuteron-induced reactions at 1.5 MeV. The total yields have been estimated by interpolating and integrating the angular distributions shown in fig. 4. For comparison the yields for the Li(p,n) and Be(p,n) reactions at 2.5 MeV are also reported in the table.

Reaction	E_{in} (MeV)	Tot. Yield (n/μC)	$<E_n>$ at 0^o (MeV)	Frac. of neut. at 0^o with E_n >1 MeV
^{7}Li(p,n)^{7}Be	2.5	$9.8 \cdot 10^8$	0.6	0
^{9}Be(p,n)^{9}B	2.5	$3.9 \cdot 10^7$	0.4	0
^{9}Be(d,n)^{10}B	1.5	$3.3 \cdot 10^8$	1.66	50 %
^{12}C(d,n)^{13}N	1.5	$6.0 \cdot 10^7$	0.55	0
^{13}C(d,n)^{14}N	1.5	$1.9 \cdot 10^8$	1.08	30 %

FIGURES

Fig. 1. (a): Background-subtracted time-of-flight distribution measured at 0° for the ^{9}Be(d,n)^{10}B reaction at E_d=1.5 MeV. The solid and dashed histogram refer respectively to thick and thin targets measurements. (b): Efficiency corrected neutron energy distribution for the thick (solid histogram) and thin (dotted histogram) target measurement at 0°. The broad distribution above 1 MeV is the result of the convolution of different high-energy peaks. (c): Measured angular distribution for the d+^{9}Be reaction. The solid line depicts the total yield as a function of angle, while the dotted line represents the yield of the low-energy (E_n <1 MeV) component only.

Fig. 2. (a): Efficiency corrected neutron energy distribution for the ^{12}C(d,n)^{13}N reaction, measured at 0°. The data refer to thick target measurements only. (b): Measured angular distribution for the same reaction.

Fig. 3. (a): Background-subtracted time-of-flight distribution measured at 0° for the ^{13}C(d,n)^{14}N reaction at E_d=1.5 MeV on a thick target. (b): Efficiency corrected neutron energy distribution for the thick target measurement at 0°. (c): Measured angular distribution for the d+^{13}C reaction. The solid and dotted lines refer respectively to the total yield and that for neutrons with E_n <1 MeV.

Fig. 4. A summary of the yield as a function of laboratory angle for some studied reactions. All results are relative to thick target measurements. For comparison, the predictions of the 2.5 MeV ^{7}Li(p,n)^{7}Be reaction are also shown in the figure. The uncertainty on the absolute yield determination, mainly associated with the normalization procedure and detector efficiency, has been estimated to be $\sim$ 30 %. In all measurement, the minimum neutron energy measured was 100 keV.

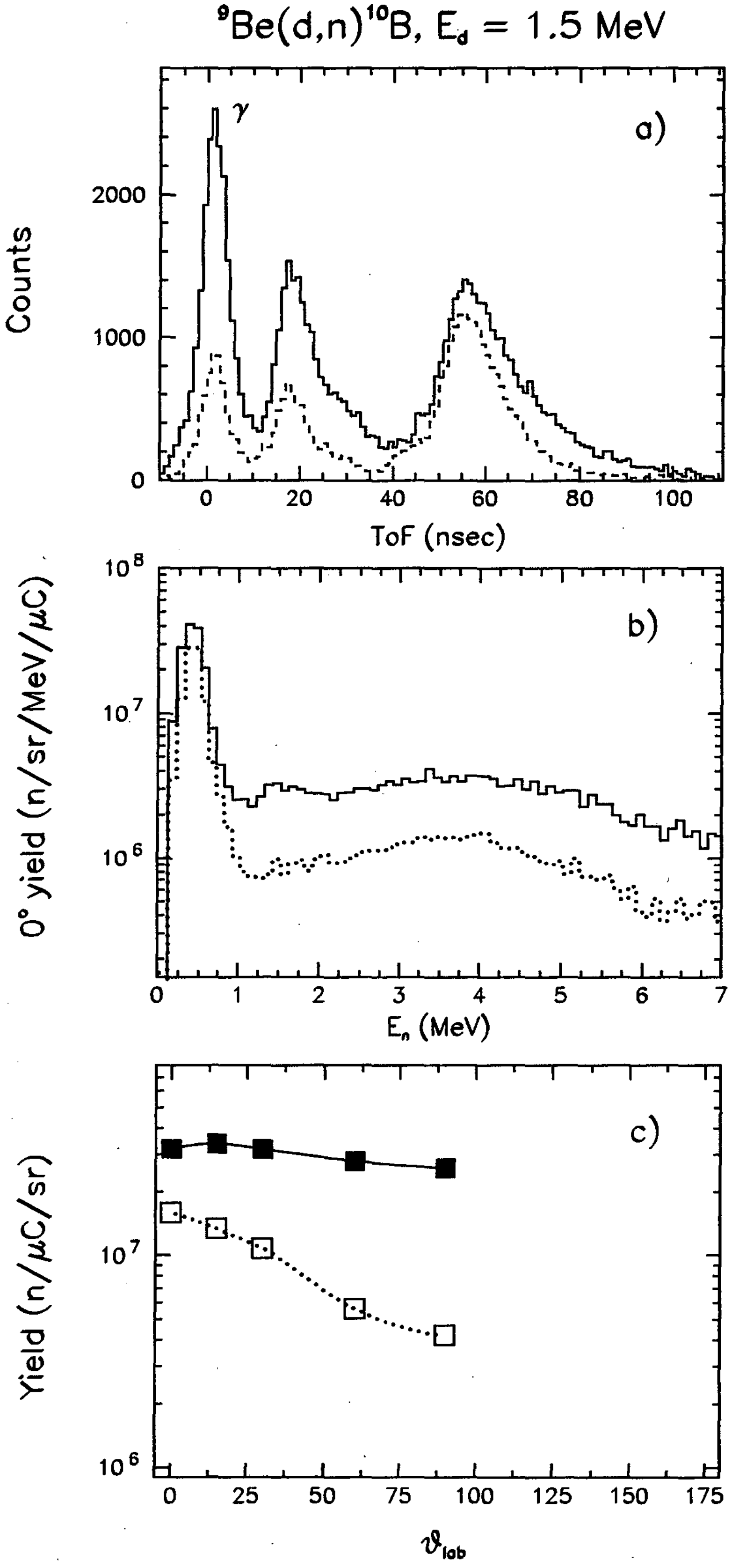
^{9}Be(d,n)^{10}B, E_d = 1.5 MeV
a)
γ
Counts
0
1000
2000
0
20
40
60
80
100
ToF (nsec)
b)
0° yield (n/sr/MeV/μC)
10^6
10^7
10^8
0
1
2
3
4
5
6
7
E_n (MeV)
c)
Yield (n/μC/sr)
10^6
10^7
0
25
50
75
100
125
150
175
ϑ_{lab}

Figure 1

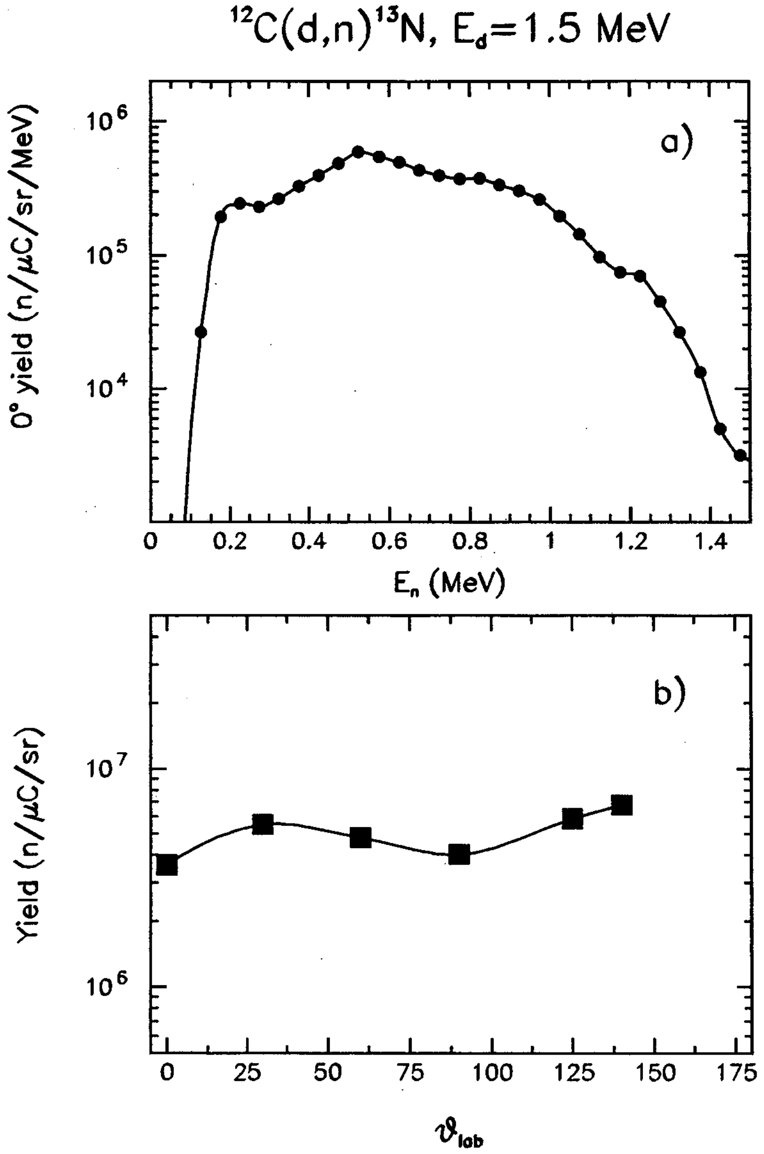
$^{12}C(d,n)^{13}N$, E_d=1.5 MeV
a)
0° yield (n/μC/sr/MeV)
10^6
10^5
10^4
0
0.2
0.4
0.6
0.8
1
1.2
1.4
E_n (MeV)
b)
Yield (n/μC/sr)
10^7
10^6
0
25
50
75
100
125
150
175
ϑ_{lab}

Figure 2

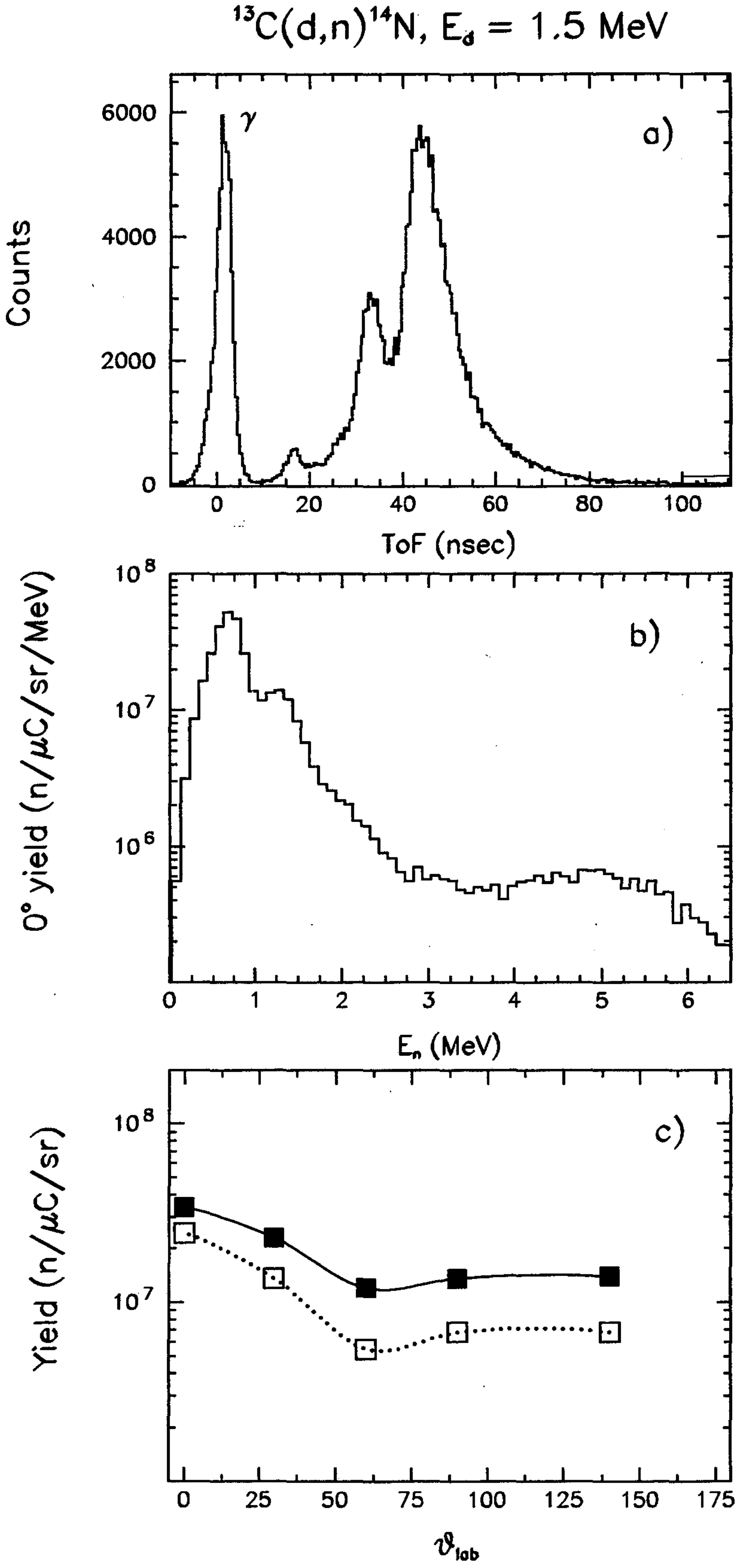
^{13}C(d,n)^{14}N, E_d = 1.5 MeV
a)
γ
Counts
6000
4000
2000
0
0
20
40
60
80
100
ToF (nsec)
b)
0° yield (n/μC/sr/MeV)
10^8
10^7
10^6
0
1
2
3
4
5
6
E_n (MeV)
c)
Yield (n/μC/sr)
10^8
10^7
0
25
50
75
100
125
150
175
ϑ_{lab}

Figure 3

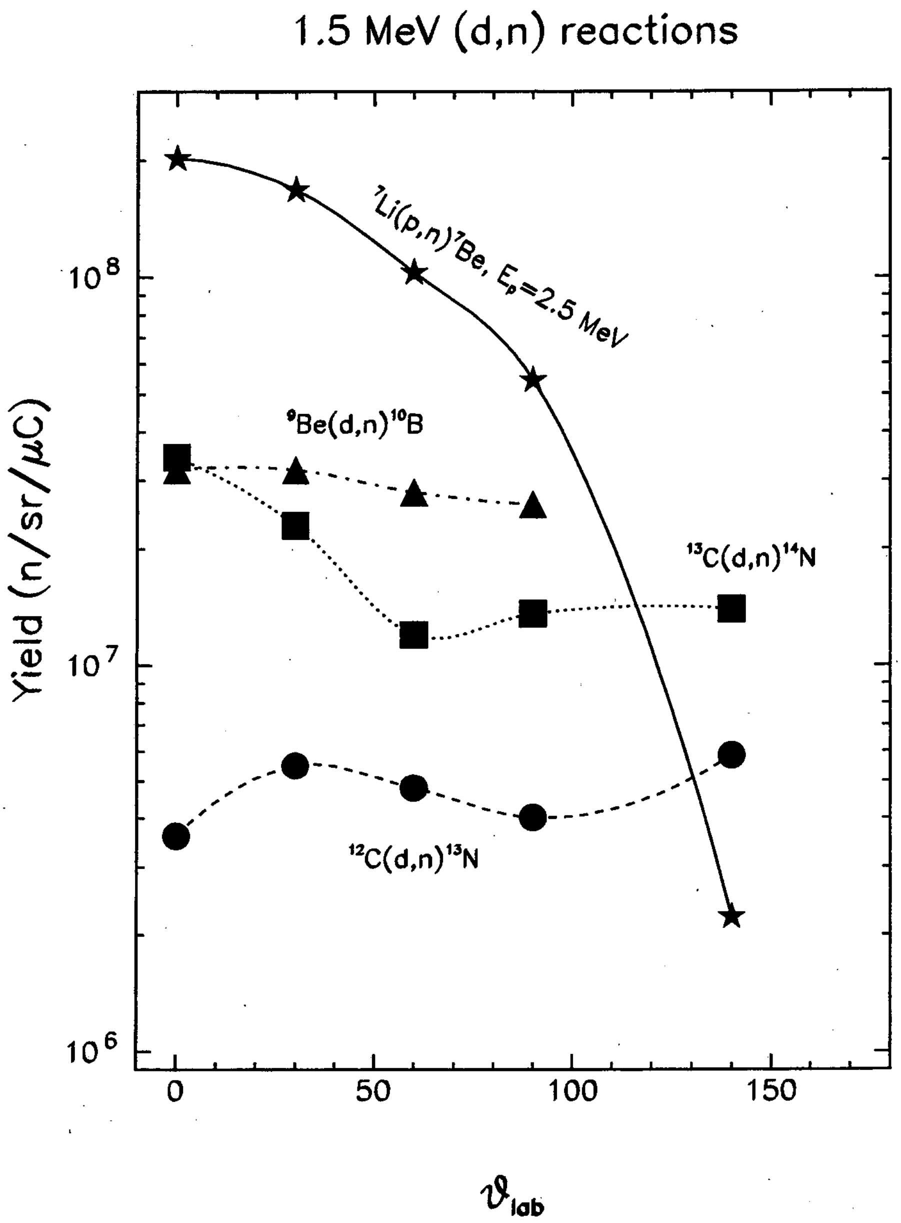
1.5 MeV (d,n) reactions
^{7}Li(p,n)^{7}Be, E_p=2.5 MeV
^{9}Be(d,n)^{10}B
^{13}C(d,n)^{14}N
^{12}C(d,n)^{13}N
Yield (n/sr/μC)
10^8
10^7
10^6
0
50
100
150
ϑ_{lab}

Figure 4

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