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ABSTRACT
The thick target neutron energy spectra of the reaction $^9$Be(p,n) were measured at several angles for proton bombarding energies of 3.0, 3.4, 3.7, 4.0 and 5.0 MeV. Time-of-flight techniques were used to determine the neutron energy spectra and to discriminate against background radiation. By using lithium-loaded glass scintillators and low proton pulse rate frequency, the neutron spectra have been determined at energies as low as 70 keV. The detectors were calibrated for efficiency using the neutron spectrum of the reaction Al(d,n) which was accurately measured using fission chamber detectors.
I. INTRODUCTION

A. Purpose

The thick target neutron energy spectra of the reaction $^9$Be(p,n) have recently been measured using standard time-of-flight (TOF) techniques at the Ohio University Accelerator Laboratory (OUAL). Since the neutron production threshold of this reaction is low, and the thermal properties of beryllium are favorable for heat removal, the reaction is an important high intensity source of neutrons for many applications. Two promising medical applications are accelerator-based Boron Neutron Capture Therapy (BNCT) and Boron Neutron Capture Synovectomy (BNCS). Both procedures utilize the reaction $^6$B(n,$\alpha$)$^7$Li to deliver radiation selectively to cells loaded with a $^{10}$B-containing compound. An international community of researchers is pursuing BNCT as a treatment for various types of cancer including glioblastoma multiforme, a form of brain cancer, and metastatic melanoma in the brain [1]. More recently, the potential for treating rheumatoid arthritis by injection of a $^{10}$B-loaded compound directly into arthritic joints followed by irradiation by a beam of neutrons has been investigated by researchers at MIT in collaboration with the Harvard Medical School [2]. This approach has been called Boron Neutron Capture Synovectomy as it is the inflamed synovial membrane that is the target with this treatment modality. Both BNCT and BNCS require a large flux of thermal neutrons at the therapy site (a tumor several centimeters deep in tissue for BNCT or the inflamed membrane up to approximately 1.5 cm deep in the joint for BNCS). In accelerator-based BNCT and BNCS, a particle reaction, such as $^9$Be(p,n), is used to produce energetic neutrons which are then moderated to form an epithermal therapy beam.
BNCT and BNCS research is being conducted at the Massachusetts Institute of Technology’s Laboratory for Accelerator Beam Applications (LABA). The accelerator at LABA, a uniquely designed tandem electrostatic, high current, multiplier type [3], produces a proton or deuteron beam whose energy and current can be varied independently over a wide range of values. The maximum designed proton energy is 4.1 MeV, and the maximum designed current is 4 mA. The total power rating of the accelerator is 10 kW. The protons will strike one of several neutron producing targets including beryllium. To investigate the use of each of these targets, accurate thick target neutron spectra are needed, especially at neutron energies below 500 keV. These data are needed as input for the development of suitable moderator and filter assemblies for epithermal beam design. The data are also needed for investigating target cooling schemes and facility shielding requirements. These data do not exist for the reaction $^9$Be(p,n) at proton energies below 4.1 MeV. The thick target reaction will provide a range of neutron energies; therefore, it is necessary to use the neutron time-of-flight technique to obtain the energy spectrum.

The measurements of the $^9$Be(p,n) spectra were completed at OUAL using 5.0, 4.0, 3.7, 3.4, and 3.0 MeV protons. This paper describes the experimental methods used to determine the neutron energy spectra. The method of calibrating the neutron detectors for these experiments is discussed. The various neutron spectra are presented, with a discussion of the sources of error.
B. The $^9\text{Be}(p,n)$ Reaction

1. Neutron production channels

The reaction $^9\text{Be}(p,n)$, as it is generally discussed, is actually several distinct reactions. Throughout this paper, the notation $^9\text{Be}(p,n)$ will symbolize, collectively, all reactions in which neutrons are produced from the proton bombardment of beryllium. A list of all known reactions which take place below $E_p = 5.0$ MeV is presented in Table I. The $^9\text{Be}(p,n)^{10}\text{B}$ reaction often proceeds through the formation of a compound nucleus, $^{10}\text{B}$, but could also result from direct charge exchange. Since the threshold for excitation of the first excited state in $^{9}\text{B}$ is 4.64 MeV, only ground state transitions $(p,n_0)$ will contribute to this reaction channel in our study at all energies but 5.0 MeV.

One unique aspect of the isotope $^9\text{Be}$ is that it has the smallest neutron binding energy (1.67 MeV) of any stable isotope. As a result, the three-body breakup of $^9\text{Be}$ can occur for incident proton energies as low as 1.85 MeV [4]. This process is labeled as $^9\text{Be}(p,p'n)^9\text{Be}$. Two reactions producing neutrons proceed through excited states of $^9\text{Be}$ and are shown in Table I. These two reactions are often collectively referred to as $(p,p')(n)$ reactions [4].

2. Previous measurements

Some of the first quantitative measurements of the $^9\text{Be}(p,n)$ reaction were made in the early 1950s using photographic film [5]. These studies used a thin beryllium target, and proton energies near 3.8 MeV. The results clearly showed the existence of a large $n_0$ group, and a significant lower energy neutron
continuum. Since that time, many other measurements have been made at proton energies from threshold to 4.0 MeV. Table II contains a list of these studies, as compiled by Byrd [6]. It is important to note that many of these measurements were undertaken to investigate the properties of a particular nuclear energy level. These experiments often consisted of relative measurements of neutron production at various laboratory angles, and in each case, thin targets were used. In most experiments, the neutron group of interest was the \( n_0 \) group, and the researchers would deliberately detect only neutrons in this group.

In the late 1950’s two important experiments were conducted to generate the neutron excitation function [7,8]. The excitation function is shown in Figure 1. The cross section rises rapidly from threshold to a value of 160 mb, at a resonance at \( E_p = 2.56 \) MeV. A second resonance was identified at 4.6 MeV [4]. Between these two resonances, the neutron production is attributed to a third broad resonance centered at 3.5 MeV [4,8]. This broad resonance is believed to be responsible for the neutron production below the 2.56 MeV peak as well [8].

Since the threshold of the reaction \( ^9\text{Be}(p,p'\eta)^8\text{Be} \) is smaller than that of the \( ^9\text{Be}(p,n_0)^9\text{B} \) reaction, it is possible to evaluate the cross section of the former reaction below \( E_p = 2.059 \) MeV. Measurements of the total neutron production at the resonance energies of the first and second excited states in \( ^9\text{Be} \) via \( ^9\text{Be}(p,p'\eta)^8\text{Be} \) have been done [9,10] and are also shown also in Figure 1. At proton bombarding energies up to approximately 2.5 MeV, neutron production is dominated by the \( n_0 \) group. Above approximately 3.0 MeV, the neutrons generated from \( (p, p'\eta) \) and \( (p, p')(n) \) reactions are more significant, and form a continuum of neutrons which is
noticeable in the energy spectrum [4]. An exact evaluation of the percentage of neutrons in this continuum is difficult since the continuum extends through the \((p,n_0)\) (and at higher energies, \((p,n_1)\)) group [4]. One report states that the continuum neutron cross section at 90°, \(E_p = 4.6\) MeV, is 25 mb/steradian, compared with 11 mb/steradian for the \(n_0\) group. The corresponding figures at \(E_p = 2.7\) MeV are reported as 6 mb/steradian and 5 mb/steradian, respectively [4]. Another estimation is that the \(n_0\) contribution drops from 65% at \(E_p = 2.5\) MeV, to 30% at 5 MeV, and 5% at 18 MeV [6]. At much higher energies (above 15 MeV), the low energy continuum dominates the spectrum [11,12,13]. As shown in Table II, there have been few studies which accurately examine the complete neutron energy spectra in the energy range \(E_p < 4.0\) MeV, which may be provided by thick target neutron spectra.

3. Gamma production

In addition to the production of neutrons, the proton bombardment of beryllium results in a significant production of gamma rays. At energies below 5 MeV, gamma production comes from only two sources: the \(^9\text{Be}(p,\alpha)^{\alpha}\text{Li}*(\gamma)^6\text{Li}\) reaction and proton capture (which is at least a factor of 1000 less than \((p,\alpha\gamma)\) above 3.0 MeV) [7]. The \(^9\text{Be}(p,\alpha)^{\alpha}\text{Li}*(\gamma)^6\text{Li}\) reaction produces a 3.562 MeV gamma. The thick target gamma yield has been previously reported for 55° at energies up to 9 MeV [14, 15] and only the 3.562 MeV 0\(^+\), \(T=1\) state in \(^6\text{Li}\) decays primarily by gamma emission. All other states in \(^6\text{Li}\) are either stable (the ground state) or decay primarily by particle emission. The spin of the 3.562 MeV state in \(^6\text{Li}\) also means that the angular distribution of the outgoing photon must be isotropic in the center of mass. A sample
of these results is presented in Figure 2. These results are especially important for
BNCT, since gamma rays provide a radiation dose which is not well localized in the
tissue containing $^{10}$B. The rapid variation of the gamma yield suggests that proton
energies below 4 MeV may be optimal in enhancing the neutron to gamma ray ratio.
II. EXPERIMENTAL METHOD

A. The Ohio University Accelerator Laboratory

Standard TOF techniques were used in the determination of the neutron energy spectra from the reaction $^9$Be(p,n). OUAL utilizes a tandem Van de Graaff accelerator with a maximum terminal voltage of 4.5 MV to produce klystron bunched proton pulses of width $< 1$ nsec for neutron TOF measurements [16]. For these measurements, the proton pulse frequency was $f = 5/16$ MHz, (or 5/32 MHz in a second experiment) and the neutron flight path was 10 m, allowing neutron energies down to 70 keV to be measured.

The axis of the accelerator is perpendicular to the axis of the flight tunnel, so the beam must be redirected by using the beam swinger [17]. The fixed detectors can be set to record data for laboratory angles from $-4^\circ$ to $+160^\circ$ simply by rotating the swinger. The target chamber is mounted at the end of the beam swinger gantry. During beam current integration, a 300 Volt battery is connected to an electron suppression screen to repel any secondary electrons back onto the target or chamber walls.

The variation of current on the target wheel with changing electron suppression screen voltage was measured. It was determined that above 50 volts, the variation diminished rapidly and then remained steady over the remainder of the measurements, indicating that this voltage is enough to suppress the most energetic secondary electrons. This series of tests confirmed that the accuracy of the charge collection is better than 2 %. The targets used for these measurements were 2.5 cm square, and 0.25 or 0.5 mm thick, and 99.9% pure in their beryllium content.
These targets were thick enough to stop 4 MeV protons. Neutrons produced at the target must pass through a collimator with a diameter of 30 cm in a 1.2 m thick concrete wall before entering the flight tunnel, shielding the flight path from neutrons scattered near the source. A detector array capable of holding seven detectors was located at 10 m flight path inside the tunnel. A diagram of the OUAL TOF facility is shown in Figure 3.

B. Neutron Detectors

The evaluation of neutron spectra at energies below approximately 500 keV, and up to 2.3 MeV, requires the use of detectors which can generate discernible signals from the interaction of low and high energy neutrons. To yield accurate energy information, the detectors must also have the ability of generating fast timing information. Four (or six in the second experiment) lithium-loaded glass scintillators were used based on their fast response time and sensitivity to low, as well as high, energy neutrons [18,19]. The lithium glass detectors contained 6.6% weight fraction of lithium, which was enriched to 95% $^6$Li. The detectors were 1.27 cm thick and 12.7 cm in diameter.

The efficiency of lithium glass detectors is complicated to calculate. At low energies, neutron detection is the result of the $^6$Li(n,α) reaction while at higher energies, (n,n′) reactions on O, Si and Li produce gamma rays which are detected in the scintillator. The problems associated with calculating the efficiency led to a decision to measure the efficiency of the detectors. The measured efficiencies differed by as much as 20% for nominally identical detectors, justifying this decision.
The efficiency of the lithium-glass detectors was determined in a two step process. First, a standard reference neutron spectrum at 120° from the reaction Al(d,n), using 7.44 MeV deuterons, was measured using fission detectors. A stopping aluminum target was used, and the neutrons were detected using TOF techniques, with a fission chamber furnished by Argonne National Laboratory. The fission chamber used ultra-pure methane as a counting gas, and was composed of four $^{235}\text{U}$ foils: U-235-J, U-235-P, U-235-S, and U-235-T, which have been previously described [20,21]. The Al(d,n) spectrum used for the calibration is shown in Figure 4. The lithium-loaded glass scintillators were then used to measure the Al(d,n) spectra. Comparisons between the reference standard and measured spectra of Al(d,n) obtained using the scintillators yielded the intrinsic efficiency data for these detectors. Details of the measurement have been published [22].

This calibration process works well at energies above 220 keV, but is problematic at lower energies because the yield of the calibration reaction, Al(d,n), decreases rapidly below 300 keV. This occurs precisely near the rapid change in lithium glass efficiency caused by the 250 keV peak in the $^6\text{Li}(n,\alpha)$ cross section. For this reason, the shape of the lithium glass efficiency curve below 220 keV was taken from the literature [18, 23]. The absolute magnitude of the curve below 220 keV was then scaled to match the experimentally determined efficiency between 220 keV and 1.0 MeV. This process resulted in a lithium glass efficiency curve shape very similar to those of Neill et al., over the region of interest for these measurements ($E_n < 3$ MeV) [18]. The average lithium glass efficiency used in the analysis of the measurements is shown in Figure 5.
A second experiment was completed in August of 1998 with 1 inch of lead in front of the detector to reduce the background due to gamma-rays from the tunnel. The efficiency above 220 keV was determined from the Al(d,n) calibration reaction at 7.440 MeV and 120°. For the second experiment, the neutron yield from the \textsuperscript{6}Be(p,n) reaction at 0° and \(E_p = 4.00\) MeV determined by the first experiment was used as a neutron standard below 220 keV. This secondary calibration with Be(p,n) was needed as Neil’s efficiencies were all taken with either a bare detector or with a polyethylene disk. The addition of lead in front of the detector increased the detection efficiency for low energy neutrons. This increased efficiency was presumably from the elastic scattering of neutrons from lead. The \textsuperscript{6}Li(n,\alpha) cross section is essentially \(1/\nu\) in this region and any reduction in energy by elastic scattering from lead would lead to a higher cross section but only a negligible time difference for the scattered neutron compared to an unscattered neutron. An additional effect of the elastic scattering from lead could be the increase of the effective thickness of the scintillator by non-normal incidence of the neutrons. The results of the calibration of the second experiment are also shown in Figure 5. A comparison of the results for zero degrees and 3.7 MeV are show in Figure 6. The excellent agreement between these two independent measurements gives us good confidence in our efficiency calibrations.

C. Data Acquisition and Processing

The time-of-flight (TOF), energy (E), pulse shape discrimination (PSD), and routing information (detector number) for each detector event are generated by
the electronic components and circuits depicted in Figure 7. These events are processed through sequential reading of the sample holds for TOF, E and PSD by the ADC and stored as event files. The time calibration and linearity of the time-to-amplitude converter are determined as described by Massey et al. [22].

The analysis of the ADC data to produce the neutron spectrum is accomplished with a suite of FORTRAN programs which execute a number of processing steps. The data were replayed and converted to time spectra by use of the gamma peak from a stilbene detector, the time calibration and combined linearity of the time-to-amplitude converter and the ADC. Any drift in the position of the gamma flash is corrected by use of the stilbene detector. This has been observed to be up to 4-5ns/day. The data are binned in energy regions and the background is subtracted for each detector. The TOF spectra are then converted to energy spectra from the TOF time calibrations. Each neutron energy spectrum is converted to neutron yield by dividing by the intrinsic detector efficiency, which was determined as discussed above. The individual detector yields for the various detectors are added together to form the total neutron yield for each energy bin. The propagation of the total statistical error is accomplished within this series of FORTRAN programs.

D. Systematic Error

In addition to the statistical error, there are several factors which add to the systematic error of the results. The solid angle subtended by the detectors is measured by determining the diameter of the detector surface and the neutron path length. These are determined to within 2 mm relatively and 5 mm absolutely. The
detectors are aligned with the target by using an alignment telescope, and the flight path is measured by a metal ruler embedded in the flight tunnel. Using Al(d,n) for the efficiency calculation results in no uncertainty in the solid angle calculation since the solid angle is the same for both the efficiency determination and the $^9$Be(p,n) measurements. However, there is a 1% uncertainty attributed to the solid angle when measuring Al(d,n) with the fission detectors. The total integrated charge has been discussed above, and an absolute error of 2% is assigned to this value. The solid angle factor and integrated charge are multiplying factors for the data, and would affect the absolute value of the data, but not the shape of the individual spectra.

The uncertainty in the shape of the spectra is affected primarily by two quantities in addition to the counts in the TOF ADC channels. These are: the energy uncertainty and the detector efficiency uncertainty. Since the energy measurement is essentially a velocity measurement, it depends on accurate assignment of path length and TOF. The accuracy of the path length has two contributions: distance from target to detector face, and thickness of the detector. For a 10 m flight path (measured to within 5 mm), and a 1.27 cm thick detector, the $\Delta L/L$ uncertainty is approximately 0.1%. The timing uncertainty is dependent on a few factors. The width of the proton beam pulse, as discussed above, is approximately 750 psec. The accuracy of the timing is optimized by taking the neutron event timing from one of the last dynodes in the photomultiplier tube amplifying chain. The neutron channels are assigned energies based on their time of flight relative to the gamma peak, which results from gamma rays produced at the target. The width of this gamma peak is approximately 1.5 nsec in a stilbene monitor detector. The total gamma time width
for the Li detectors is approximately 4 nsec. Since energy is proportional to \((\text{TOF})^2\), the uncertainty affects the higher energy portion of the neutron spectra more than the lower portion.

Finally, the accuracy of the shape and magnitude of the spectra are both affected by the accuracy of the detector efficiency calibration. The interaction cross section of the fission foils is known to between than 1 and 3% dependent upon the energy; however, the weight, and hence, thickness of the foils has an uncertainty of \(\pm 5\%\) [20,21]. The efficiency calibration is also dependent on the statistical quality of the Al(d,n) measurements, which is approximately 5%, combined with a systematic error of approximately 5% [22]. At energies below 250 keV, the efficiency determination is dependent on the accuracy of the initial calibration data in the literature [18,23]. The error introduced by transcribing this data from the literature is difficult to judge. Considering all of these effects, the detector calibration efficiency is assigned an error of 8% above 250 keV, and 12% below 250 keV. The total error in the absolute magnitude of the neutron spectra data, then, is estimated to be less than 10 % above 250 keV, and 20% below 250 keV.

E. Confirmation of Spectra Using \(^6\text{Be}(d,n)\) Reaction

To confirm the accuracy of the detector calibration procedure, and all other aspects of the measurement process (time calibration, charge collection accuracy, programming, etc.), the measurement of the \(^9\text{Be}(p,n)\) spectra was immediately preceded by thick target measurements of the reaction \(^9\text{Be}(d,n)\). This reaction was measured (on an absolute basis) at 0°, for \(E_d = 2.6\text{-}7.0\) MeV, at 400 keV.
intervals. These results were then compared to data of Meadows’ [21]. One such comparison, using 7.0 MeV deuterons, is shown in Figure 8. The error bars reflect only the total statistical error.

The agreement between the two data sets is very good. The average neutron yield is at most on average 3% lower than Meadows’. This is not an indication of a problem however as Meadows’ absolute normalization is also not known to better than 4%. The maximum disagreement is at low energies below 300 keV. At these energies, the stated statistical uncertainty of the Meadows’ data is 2.5%, in addition to a total systematic error of 4%. The energy region below 250 keV is one which can be subject to errors which are difficult to estimate due to the difficulties in background subtraction. In this energy region, the time-of-flight spectrometer spreads the data over many channels. Thus, random TOF contributions to the background subtraction have more effect on the data in this region than at higher energies, even if the cross sections are comparable. In addition, Meadows’ measurement was done with a fission counter at 2.68 meters with 1 MHz frequency. The background was determined in Meadows’ experiment by measurement at 2 MHz to obtain the shape of the wraparound tail. The energy bins are 20 keV at this energy in the current work, while they are between 40 and 50 keV in Meadows’ work. The narrower energy bins in our work tend to enhance the difference from a rapidly changing spectrum.

The second discrepancy between Meadows’ results and the current work is the region between 4 and 6 MeV. In Figure 8 two wide peaks are present in Meadows’ work that are not present in the current experiments. Since Meadows used fission
chambers, his work required nearly $10^4$ times the beam on target was needed to obtain their spectra. Additionally, the high currents required for the measurement also necessitated water cooling and a fairly elaborate target assembly. Their published experiment [21] showed a large range of energies covered by their work. The best explanation for these features is that they are due to deuterium build up from experiments at a lower deuteron incident energy in the beryllium target of Meadows. Meadows’ results were also lower than previous work in this region. Our current results agree much better with Meadows’ results than any of the previous work.
III. RESULTS AND DISCUSSION

The neutron spectra were measured at the various combinations of energy and angle shown in Table III. The complete data set is provided at the National Nuclear Data Center®. There is a separate table for each proton bombarding energy. The data for each neutron spectrum are given as a series of yields with associated statistical errors. The data points represent the yield of a particular energy bin. The energy at the center of this bin is provided. The bin widths are 20 keV up to a neutron energy of 700 keV and are 50 keV otherwise. The units of the neutron spectra are neutrons/ (MeV- steradian- microcoulomb).

Samples of the neutron energy spectra for the various combinations of angle and energy are shown in Figure 9 and Figure 10. Figure 9 shows the neutron energy spectra at 4 angles for 3.7 MeV incident protons. Figure 10 shows neutron energy spectra at 4 angles using 4.0 MeV protons. Figure 11 depicts the 0 degree spectrum at the various proton bombarding energies.

The explanation of the shape of the various neutron spectra is the topic of ongoing research at OUAL. Several contributing reactions have been previously reported as discussed earlier. The neutrons produced with energies above 1 MeV are believed to be due primarily to the reaction $^9$Be(p,$n_o$)$^9$B which proceeds through the compound nucleus $^{10}$B. It is important to note that the best information on the reaction mechanism would come from thin target measurements, and not thick target measurements. However, a calculation may help to understand the neutron energy spectrum for 0°. The shapes of the spectra at other energies are investigated also. The calculation is based on an excitation curve for neutrons in the $n_o$ group, 0°.
laboratory angle, measured by Marion [4]. Data for this calculation from Figure 5 of Marion’s paper were obtained from the NNDC, the extracted $^9$Be(p,n$_0$) was assigned a maximum absolute uncertainty of 30% based upon the extraction of the n$_0$ group from the continuum. The excitation function at 0° (is generally flat in the region 2.8 MeV < $E_p$ <3.6 MeV. This differs from the excitation function for all angles (also presented in the same paper), which has a large broad resonance centered at 3.5 MeV.

The calculations of the thick target yields were made considering the thick beryllium target as a continuum of atoms. The calculations below used stopping powers from Ziegler [24]. The attenuation of the proton beam is neglected as only 16 in 100,000 protons actually produce a neutron. It is reasonable, then, to consider a beam of uniform intensity passing through the target, slowing down, and eventually stopping. As the proton beam enters the beryllium target, with energy 4 MeV, it produces neutrons in the n$_0$ group corresponding to an energy E(n$_0$) (near 2.2 MeV). The protons lose energy traversing the target and slowing down producing fewer neutrons due to the lower cross section, and these neutrons have a slightly lower energy. This process continues until the protons reach the resonance at 2.56 MeV. Here, although the n$_0$ group has less energy, the production actually increases. This result can be compared with the actual normalized measured result from the present data as shown in Figure 12. It should be emphasized that this comparison is qualitative, and not quantitative in nature due to the large uncertainty of Marion’s data. There are also uncertainties in the stopping power of protons on beryllium, especially at low energies.
The neutron production near 500 keV may also have some contribution from the decay of the second (2.443 MeV) excited state of $^9$Be. The $^9$Be nucleus is excited via the reaction $^9$Be(p,p'n). A peak near this energy has been seen before in experiments using a stilbene crystal for neutron detection [13]. Finally, a group of neutrons with energies centered around 500 keV was previously reported using a proton bombarding energy of 4.5 MeV. This peak existed only at the forward angles [4].

The development of the neutron production from a thick neutron target at zero degrees with energy is show in Figure 12. At low energy (3.0 incident proton energy) the production of neutrons is dominated by sequential and three body break-up. The peak of the distribution is 600 keV. This peak becomes larger and broadens as the energy increases. At higher incident energy (4.0 MeV), the majority of the neutrons come from the $^9$Be(p,n$_0$) reaction. The other channels contribute a substantial fraction of the neutrons at energies below 1.0 MeV which are of interest for BNCT. To show this more clearly we have subtracted the contribution from the $^9$Be(p,n$_0$) reaction from the zero degree spectra at 3.0, 4.0 and 5.0 MeV. The resulting spectra for the sequential and multiparticle decay are shown in Figure 13.

The total neutron yield of the reaction can be determined by integrating the various neutron spectra over the measured energy range and solid angle. All data from both experiments were initially considered for inclusion for fitting of the angular distribution to a third or fourth order polynomial. These data are shown in Figures 14 and 15. Discrepant points were then removed from the data set for the fitting. Finally, only data from our first experiment with a bare detector were used for fitting.
the angular distribution. This was done primarily to remove the influence of known systematic errors on the total error of the fit as there is \( \sim 2\% \) error in the relative measurement and a 5% error between the two sets for absolute normalization. (The discrepant points from above were then renormalized using the angular distribution of the yield of neutrons with energies above 0.220 MeV to correct for known problem of charge normalization. The error in this renormalization is \( \sim 2\% \) or the same as expected using simple charge integration.) Using proton bombarding energies of 3.7 and 4.0 MeV, the absolute total yield was determined by integrating the polynomials (fit to the reduced data set) to be \( 9.61 \pm 0.48 \times 10^{11} \) and \( 1.48 \pm 0.07 \times 10^{12} \) neutrons/mC respectively. The neutron yield estimated from the complete set of neutron spectra now agrees within the error bars of the calculated total neutron yield for a thick target. The calculated total neutron production from a thick target is shown in Figure 16. The major source of error in the calculation is from the stopping powers from Ziegler [24]. The measured stopping powers for beryllium vary widely. In addition, extrapolation based on other elements may not be valid as beryllium is the lightest solid which can be well measured. The errors from all of the nuclear reactions are less than 5-10%.

The statistics of the neutron yield versus angle, also shown in Figures 14 and 15, may suggest a different approach to neutron collimation and moderation for BNCT. The average neutron energy decreases slowly versus angle at both energies where the angular distribution have been measured. The neutron yield is fairly flat up to nearly 90 degrees. A collimation/moderation system may thus be placed at an angle other than 0 degrees to select the spectrum of neutrons presented in the
direction of the patient. This should result in smaller collimation assemblies and ultimately more neutrons delivered for therapy.

There have been two measurements of the total neutron yield for the $^9$Be(p,n) reaction. The first was performed at Argonne National Laboratory covering the energy range of 3 - 7 MeV [25]. The second experiment was performed at Birmingham and covered the energy range from 2.2 to 3 MeV [26].

The Argonne National Laboratory experiment consisted of two different experimental setups. The first setup was a paraffin castle with embedded BF$_3$ counters. The majority of the data was taken with this setup. The other setup was a vanadium bath. In a vanadium bath a thick target is placed at the end of a slender snout. The snout is inserted into a hole in the wall of the bath. The contents of the bath are then recirculated between the bath and a gamma detector. For both of these methods corrections need to be applied for the high energy neutron emission. These corrections have not been applied due to the lack of neutron energy distributions as a function of angle and incident proton energy. The corrections were expected to be less than 10% and should have increased the observed yield. This data is plotted in Figure 16.

The work of Campbell and Scott at Birmingham [26] was also done in a vanadium bath. They had configured their bath in the shape of a sphere to ease the calculation of the neutron correction factors. The correction factors considered were neutron leakage, self absorption of the source, neutron absorption in sulfur and oxygen. Their estimated accuracy was from 1.5 to 3.5%. These thick target results of Campbell and Scott [26] compare very well with the yields calculated from the thin
Following the submission of this paper, we have become aware of the work by Guzek et al. [27]. Their measurements employed a 5×2.5 cm NE102A plastic scintillator at between 1.5 - 2.0 meters from the target. The measurements were done at the 6 MV van de Graaff facility at the National Accelerator Center, Faure, South Africa. They have completed measurements of thick target yields at 2.57, 3.0 and 4.0 MeV with complete angular distributions at these energies.

We have compared our results with theirs when possible. A direct comparison of the 3.0 and 4.0 MeV 0 degree spectra showed a reduced yield for Guzek’s work relative to the current work near 600 keV (the peak of the neutron yield). This could be due either to a problem with efficiency determination or insufficient energy resolution or both. No information was given in Guzek’s paper on the efficiency calibration.

Guzek also report gamma ray yields and these are shown in Figure 2 for comparison to previous work. The gated measurement is a factor of ten higher then previous measurements. This may be due to gamma rays from neighboring structural materials. The gamma yield from Be is less than the neutron yield, reflecting the lack of particle stable excited states in $^9$Be and particle stable states in $^9$B.

A second measurement by Yue et al. [28] has also been recently published. Their measurement was made with a 5.0 cm thick by 10.4 cm diameter ST-451 type scintillator at 2.663 meters. The neutron spectra are in reasonable agreement with our current results at 3.0 and 3.4 MeV when the poorer resolution and 400 keV cutoff energy are taken into account. Yue et al. reported neutron spectra at zero degrees
and the derived zero degree neutron yield. Their zero degree yield and our results for zero degree yield are shown in Figure 17. The results of Yue at 3.0 and 3.5 MeV are in qualitative agreement with our data. At 4.0 MeV their results deviate from the current results by more than what can be accounted for from the differences in the lower energy neutron cutoff. The zero degree yield from the current results has been fit to an exponential form. The values of the coefficients are given in Figure 17.

The number of gamma-rays produced per neutron by the source reaction is important in BNCT. The dose from the gamma-rays produced by the source reaction increases the dose to the surrounding healthy tissue in the treatment of brain tumors. We have used the compiled total neutron yield as shown in Figure 16 and gamma-ray yield to determine the gamma to neutron ratio for the $^9$Be(p,n) reaction. For the sake of comparison we have also calculated the thick target yield for $^7$Li(p,n) from the thin target data of Sekharan [29]and Gibbon [8]. The gamma-ray to neutron ratio for these two candidates source reactions for BNCT are shown in Figure 18. The error in the points is expected to be dominated by the error in the gamma ray yield measurement. These points are expected to have an errors somewhat less than 10%. Additionally, since the gamma yield was taken from a 55° measurement some correction for the angular distribution of the gamma-rays is needed. For protons bombarding beryllium, the largest gamma ray transition is from the decay of the 3.563 state in $^6$Li which is $0^+;T=1$ to the $1^+;T=0$ ground state and thus the transition must be isotropic in the center of mass and the correction for gamma ray angular distribution should be on the order of 10-15%, while for protons bombarding $^7$Li, the main gamma-ray is the transition from the 0.477 MeV $1/2^-$ state to the $3/2^-$ ground
state in $^7$Li. The angular distribution of this 0.477 MeV gamma is expected to be small and result in less than a 25% correction to the gamma-ray yield. This gamma-ray yield data is for natural lithium and would have to be increased by up to 7% if lithium enriched in $^7$Li is used. Regardless, it is clear that for $^9$Be(p,n) from the viewpoint of the number of gamma-rays/neutron the $^9$Be(p,n) is between a factor of 30 less at 3.0 MeV and of 9 less at 4.0 MeV than the $^7$Li(p,n) reaction.
IV. CONCLUSIONS AND SUGGESTIONS FOR FUTURE MEASUREMENTS

A complete data set now exists for the absolute thick target yield of $^9$Be(p,n) for $E_p = 4.0$ and 3.7 MeV. The data have been confirmed indirectly through the measurement of the $^9$Be(d,n) spectra at several energies. A useful addition to the data set would be to continue the measurements at proton bombarding energies below 3.7 MeV to near threshold. The shape of the total reaction spectrum seems to be based on the shape of the expected $^9$Be(p,n$_b$) spectrum, to which the low energy continuum is added. To further refine an understanding of the shape of the spectra, thin target measurements would be required.

The $^9$Be(p,n) reaction is a potentially valuable calibration source for neutron detectors. The spectrum at 0 degrees is generally flat for $E_p = 4.0$ MeV, and the neutron yield is appreciable at least down to 200 keV. These qualities make the reaction an ideal choice for calibrating neutron detectors. This was used for the efficiency determination of the lithium glass detectors with one inch of lead in front of them in this work. The possibility of more accurately determining a single energy spectrum of this reaction for calibration purposes is being investigated at OUAL.
Footnotes

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** E-mail address: massey@ohiou.edu. Full data set described in this paper is available at http://www.phy.ohiou.edu/~massey.

†Present address: King Fahd University of Petroleum and Minerals, Physics Department, Dharan 3126, Saudi Arabia

‡ Present address: NASA Johnson Space Center, Space Stations Operations Planning, Mail Code DO47, Houston, TX 77058

@This data is being archived at the National Nuclear Data Center in the Experimental Nuclear Reaction Data (EXFOR(CSISRS)} data base. This data may be accessed via the NNDC web site http://www.nndc.bnl.gov by choosing the link to the EXFOR data base. The parameters needed to retrieve this work are Target: 9Be, Projectile: p and then choose Retrieve by Reaction. A list of reactions will come up and the (p,X) should be choosen. A list of data available for this reaction will then come up. This data will be listed under thick data data (TTD).
Acknowledgements

This work has been supported by the United States Department Energy under grant DE-FG-02-88ER40387. Of the authors (S.I.A.), acknowledges the support received from the King Fahd University of Petroleum and Minerals, Dhahran, Saudi Arabia. One of the authors (T.N.M.), dedicates his work on this problem to the memory of Oliver G. Lien, Jr.
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26. J. Campbell and M. C. Scott, “Absolute Neutron Yield Measurements on Li, Cu, Co and Be from Threshold to 3.0 MeV” in the Proceedings of the Fourth Conference on the Scientific and Industrial Applications of Small Accelerators,


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<th>Reaction</th>
<th>Q Value</th>
<th>Threshold</th>
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<tr>
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Table II
References for $^9$Be(p,n)

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<th>Group(s)</th>
<th>$E_{p \gamma}$ (MeV)</th>
<th>Angles</th>
<th>Error (%)</th>
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<td>(7-10)</td>
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<td>integral</td>
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<td>Marion &amp; Levin (4)</td>
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<td>2.3-5.6</td>
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<td>(10)</td>
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<td></td>
<td>integral</td>
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<td>90</td>
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<td>Gibbons &amp;</td>
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<td>(20)</td>
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<td>4-14.3</td>
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<td>3.4</td>
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Captions for Tables I-III

Table I: Neutron producing reactions for \( ^9 \text{Be}(p,n), E_p \leq 4.0 \text{ MeV} \).

Table II: Measurements of \( ^9 \text{Be}(p,n) \) for \( E_p \leq 4.0 \text{ MeV} \) [6]. Notes: (a) The number in parenthesis corresponds to the reference number. (b) "none" indicates that the neutron energies were not evaluated. "n\(_0\)" indicates that only the n\(_0\) group was measured. (c) The number in parenthesis is the number of angles measured. (d) Numbers in parenthesis have been assigned by the author of the review article. In the case of the research by Gibbons and Macklin, Byrd claims that this number is stated in the original work, as 3%. This does not seem to be supported by a careful reading of the original work. No clear statement of absolute error is reported in the original paper.

Table III: Combinations of energies and angles for which the neutron spectra of \( ^9 \text{Be}(p,n) \) were measured.
Captions for Figures 1-18

Figure 1  Excitation function of the reaction $^9{\text{Be}}(p,n)$ for $E_p \leq 4.0$ MeV.

Figure 2  Thick target gamma-ray yield on a beryllium target. The data shown is from references [13,14].

Figure 3  Ohio University Accelerator Laboratory (OUAL) facility.

Figure 4  Neutron spectrum of the reaction $\text{Al}(d,n)$ used to calibrate the neutron detectors.

Figure 5  The lithium glass efficiency for the same detector used for both a bare detector and with 1 inch of lead in front of it.

Figure 6  Comparison of results for $E_p = 3.7$ MeV, 0 degrees.

Figure 7  Neutron detection electronic circuit.

Figure 8  Comparison of MIT/OUAL and Meadows’ measurement of neutron spectrum for $^9{\text{Be}}(d,n)$, $E_d = 7.0$ MeV.

Figure 9  $^9{\text{Be}}(p,n)$ neutron spectra at three angles for $E_p = 3.7$ MeV.

Figure 10  $^9{\text{Be}}(p,n)$ neutron spectra at three angles for $E_p = 4.0$ MeV.

Figure 11  $^9{\text{Be}}(p,n)$ neutron spectra at 0 degrees for four energies.

Figure 12  Results of a calculation used to illustrate the possible source of neutrons in the 0 degree, $E_p = 4.0$ MeV spectrum of $^9{\text{Be}}(p,n)$. The symbols are the data from the current work and the dashed and solid lines are the calculated $^9{\text{Be}}(p,n_0)$ contributions at the incident proton energies of 3.0, 3.4, 3.7, 4.0, and 5.0 MeV.
Figure 13 Spectra at zero degrees at 3.0, 4.0, and 5.0 MeV with the calculated $^9$Be(p, $n_0$) contribution subtracted. The error bars shown are from the original spectra only with the contribution to the error from the calculated $^9$Be(p, $n_0$) contribution neglected.

Figure 14 The experimental 3.7 MeV angular distribution for $E_n > 0.070$ MeV is shown in the upper panel. The average neutron energy versus angle is show in the lower panel along with the average energy plus and minus the first moment of the energy distribution and the maximum observed energy of the neutrons at each angle.

Figure 15 The experimental 4.0 MeV angular distribution for $E_n > 0.070$ MeV is shown in the upper panel. The average neutron energy versus angle is show in the lower panel along with the average energy plus and minus the first moment of the energy distribution and the maximum observed energy of the neutrons at each angle.

Figure 16 Calculated and experimental thick target neutron yield. The neutron yield was calculated using the evaluated cross section in Figure 1. The ANL and Birmingham total neutron yields are uncorrected for leakage of high energy neutrons.

Figure 17 Measured zero degree neutron yields in the upper panel. The lower panel shows the energy dependence of the mean neutron energy and the mean neutron energy plus and minus the first moment of the energy distribution. Also shown in the lower panel is the maximum energy of the observed neutron spectrum.
Figure 18  The measured ratio gamma-ray to neutron ratio yield for protons on beryllium and lithium. The gamma ray yield is from 55° measurements and are estimated to have less than 10% error.
$N_{\gamma}/(\mu C \text{sr})$ at $\theta_{\text{lab}} = 55^\circ$

- $^{6,7}\text{Li}(p,x\gamma)$
- $^{9}\text{Be}(p,x\gamma)$
- $^{10,11}\text{B}(p,x\gamma)$

Guzek TOF Gate
Guzek no Gate
Figure 4

$^{27}\text{Al}(d,n)$

$E_d = 7.44 \text{ MeV}$

$\theta = 120^\circ$
$^9$Be($p$,n)

$E_p = 3.7$ MeV

$\theta = 0^\circ$

Figure 6
Beam Pickoff Electronics

Li Glass Electronics

Figure 7
Figure 8

$^9$Be(d,n)

$E_d = 7.0$ MeV

$\theta = 0^\circ$

neutrons/($\mu$C sr MeV)

$E_n$ (MeV)

Li Glass

Meadows

Figure 8
Figure 9
$^{9}\text{Be}(p,n) E_p = 4.0 \text{ MeV}$

neutrons/(MeV sr µC)

$E_n$ (MeV)
Figure 11

The graph shows the neutron yield for the reaction $^9\text{Be}(p,n)$ as a function of neutron energy $E_n$ (in MeV). The data points are labeled for different projectile energies: 3.0 MeV, 3.4 MeV, 3.7 MeV, 4.0 MeV, and 5.0 MeV. The vertical axis represents the number of neutrons per unit volume, normalized to MeV, steradians, and micrometers.
Estimate of Continuum Contribution

Figure 13
$^9\text{Be}(p,n)$

$E_p = 3.7\text{ MeV}$

\begin{figure}
\centering
\includegraphics[width=\textwidth]{figure14.png}
\caption{Figure 14}
\end{figure}
Figure 15
$^9$Be(p,xn) Neutron Yield

Current Results
ANL wax castle BF$_3$
ANL V bath
Birmingham V sphere
Thin Target Calculation
$^9\text{Be}(p,n)$

$\theta = 0^0$

yield = $a_0 \exp(a_1(E_p - 2.05763))$

$a_0 = 8.53789 \times 10^6$

$a_1 = 1.34188$
$N/_{\gamma}(\theta=55^0)/N_n$ vs $E_p$ (MeV)

- Dashed line: Li + p
- Solid line: Be + p

$10^0$ - $10^{-1}$ - $10^{-2}$