DEVELOPMENT OF A NEW METHOD TO MEASURE (n,α) AND (n,p) CROSS SECTIONS USING A LEAD SLOWING-DOWN SPECTROMETER

J. T. Thompson and Y. Danon

Gaerttner LINAC Laboratory, Rensselaer Polytechnic Institute, Troy, NY 12180, USA

The need for nuclear data comes from several sources including astrophysics, stockpile stewardship, and reactor design. Although models can be used to predict some of this data, these predictions are only as good as the experimental data that constrains them. A new method of measuring energy dependent (n,α) cross sections was developed for the energy range of 0.1 eV - 100 keV using a lead slowing-down spectrometer (LSDS) driven by a 60 MeV electron linear accelerator. The LSDS provides a ~10^7 neutron flux increase over time-of-flight (ToF) methods of similar energy-time relation allowing the measurement of small sample masses (µg’s to mg’s) or small cross sections (µb’s to mb’s). The ^147,149Sm(n,α)144,146Nd cross sections were measured as a proof of concept. This measurement of ^147Sm verified and extended a previous measurement while the measurement of ^149Sm is the first to cover this energy range.

I. INTRODUCTION

Accurate nuclear data is of crucial importance when investigating reactor parameters and operation, stockpile safety and reliability, and stellar and nucleosynthesis models. Even since before the famous publication of Burbidge, Burbidge, Fowler, and Hoyle (commonly referred to as B’FH) there has been curiosity about the origin of isotopes. Over 50 years later much progress has been made in the field of nucleosynthesis1-3. However, several problems still exist. Woosely and Howard4 has proposed that an intense γ flux created by medium near 2 x 10^9 degrees is capable of reproducing solar isotopic abundances of p-nuclei, rare proton-rich isotopes produced primarily or exclusively during the p-process. Unfortunately no one temperature or time scale is capable of accurately reproducing all abundances. One problem here may be inaccurate cross sections, especially for reactions involving α particles and intermediate mass nuclei.

Models can be used when cross section have not been experimentally measured. To verify these models experimental results are needed. This method has met with limited success for reactions involving α particles and intermediate to heavy nuclei because the limited numbers of experiments are not directly applicable to the mass or energy range of interest to astrophysics.

Work is currently being performed at the Gaerttner LINAC Laboratory using a lead slowing-down spectrometer (LSDS) to measure (n,α) and later (n,p) cross section measurements, providing the experimental results needed to verify these models.

I.A. GAERTTNER LINAC LABORATORY

The Gaerttner LINAC Laboratory houses a linear electron accelerator and several detector systems which use time-of-flight (ToF) techniques to measure total7, capture8 and scattering9 cross sections. The linear accelerator produces a pulsed beam of up to 60 MeV electrons with a maximum power of about 16 kW10, when connected to the LSDS the maximum power is kept around 1 kW due to target and lead heating concerns. These electrons are incident on a series of tantalum plates where they produce Bremsstrahlung radiation which can interact with additional tantalum plates to produce neutrons in an evaporation spectrum.

I.B. THE LSDS

In addition to several ToF stations, RPI’s Gaerttner LINAC Laboratory has a 1.8 m, 75-ton lead LSDS11, shown in Fig. 1. Neutrons are produced with an evaporation energy spectrum in the tantalum target at the center of the lead cube. Below 1 MeV the lead cross section is dominated by elastic scattering causing neutrons slowly gain lethargy at a predictable rate. The relationship between slowing-down time, t, and average neutron energy, \( E \) [eV], for the LSDS11 at RPI11 is given by

\[
E = \frac{165000 \text{ eV} \cdot \mu s^2}{(t + 0.3 \mu s)^2}
\]
and is plotted in Fig. 2. This relationship is very similar to relationship between time and energy for ToF experiments, and the values correspond closely to those for a ToF path 5 m in length.

Neutrons are born with the broad energy of an evaporation spectrum, however fast neutrons tend to scatter sooner and lose more energy per collision than slow neutrons. This leads to a focusing of neutron energies around the mean neutron energy given by (1). The focusing effect is not perfect and the neutron population reaches its lowest full-width at half-max (FWHM), $\Delta E/E$, of ~30% at $\bar{E} = 1$ keV. Below this energy resolution broadens again due to the thermal motion of the lead nuclei. The FWHM is modeled as

$$\frac{\Delta E}{E} = \sqrt{\frac{0.128}{E} + 0.0835 + 3.05 \times 10^{-5} E}$$  \hspace{1cm} (2)$$

and is shown in Fig. 3. This causes a broadening of cross sections since the effective cross section at a mean energy is the flux weighted average of the true cross section. With the assumption that the neutron population’s velocity is distributed normally about the mean the broadened cross section, $\sigma^b$, can be computed from the true cross section, $\sigma$, as

$$\sigma^b(\bar{E}) = \frac{1}{\sqrt{2\pi}} \int_0^\infty G(E',\bar{E})\sigma(E')\,dE'$$  \hspace{1cm} (3)$$

where $G$ is a Gaussian with a FWHM given by (2).

Because neutrons remain in the LSDS and frequently scatter in new directions, the same neutron may pass through the sample multiple times before being finally being removed through absorption by the lead or leaking from the edge of the LSDS. This leads to flux increase of $\sim 10^4$ over the time/energy equivalent ToF station at 5 meters from an equivalent neutron source. This large flux increase allows for the possibility of measuring samples of only 10’s of nanograms or larger samples with reaction cross sections of 100’s of microbarns.

Romano et. al.\textsuperscript{15} was able to perform a measurement of the $^6$Li(n,α)$^3$H cross section a using only 62 µg of natural lithium with the LSDS at Los Alamos National Laboratory while Rochman et. al.\textsuperscript{16} demonstrated that the $^{239}$Pu(n,f) $^6$Li(n,α)$^3$H cross section could be measured using only 9.87 ng and 760 ng of $^{239}$Pu and $^6$Li, respectively. Later, Romano et. al.\textsuperscript{13} also used the LSDS at RPI to perform simultaneous cross section and fission fragment mass distributions measurements using 23.5 µg of $^{235}$U. These measurements showed that sizable cross

Fig. 1. A photograph of the LSDS installed at the Gaerttner LINAC Laboratory at RPI.

Fig. 2. Average neutron energy as a function of time in the RPI LSDS compared to 3 different ToF stations.

Fig. 3. Full-width at half-maximum as a function of average neutron energy.
section (> 1 barn) could be measured on very low mass samples, however using larger masses to make up for lower cross sections adds an additional problem since sample masses are usually limited by the range of the charged particles. Rochman and Romano were able to detect the fission fragments and α particle directly because the samples were thin enough for the charge particles to escape. The work presented here will focus on the measurement of the $^{147,149}\text{Sm}(n,\alpha)^{144,146}\text{Nd}$ reaction cross sections (~1 mb) on samples with masses ~10 mg and sub-mb cross sections.

II. EXPERIMENT

Measurements of the (n,α) reaction cross section were performed on two isotopes of samarium. Samarium was selected for the initial test of the system due to the high $Q$-values for several of its isotopes, around 10 MeV, and that some measurements were already performed, thermal measurements$^{17,18,19}$, some measurements with monoenergetic neutron beams in the 100’s of eV to 10’s of keV region$^{20,21,22}$, and one high precision measurement over the majority of the useful energy range of the LSDS$^{23}$. Samples of enriched $^{147}\text{Sm}$ and $^{149}\text{Sm}$ (98.3 ± 0.05 % and 97.669 ± 0.03 %, respectively) were prepared by stippling 7.91 ± 0.04 mg and 8.11 ± 0.04 mg of the respective isotopes, (in the form of Sm(NO$_3$)$_3$) onto 99.9% pure aluminum disks.

The detector consisted of two Canberra PIPS model PD-150-30-404PI-40 solid state detectors. Because the detector must be placed inside the LSDS they were within half a meter from the neutron source. Each neutron pulse was preceded by an intense burst of γ-rays and RF interference, termed the "γ-flash." In the LSDS the γ-flash is strong enough to saturate most detectors and the electronics connect to them so the signals from the PIPS detectors were combined after one was inverted, as shown in Fig. 4. Combining the two signals prior to the preamp allowed the γ-flash, which produced simultaneous and roughly equal signals on both detectors, to sum to nearly zero while discrete signals from α particles were persevered. This circuit induced a large overshoot into signals from α particles and causing them to become bipolar.

During slowing-down times corresponding the resonances in samarium γ pile-up becomes a concern. The two PIPS units were placed facing the sample at equal distances from the sample with roughly a 2 mm air gap. The sample backing prevented α particles from reaching one of the detectors but capture γ’s were not significantly attenuated and could interact with either detector. The compensation circuit was able to reduce the signal height during these times and reduced the false event rate.

The amplified signal was sent to an Agilent 12-bit digitizer model U1066A. The board collected waveforms, triggered by a threshold trigger, 800 ns in length (200 ns prior to and 600 ns after the trigger) with a sampling rate of 420 MHz. Pulse shape analysis was performed to filter γ-pile-up, noise and γ-flash using two conditions. The criteria for α particles was that the minimum and maximum sample were between 80 and 120 samples apart (~190 ns and ~285 ns respectively) and the peak-to-peak amplitude ratio be between 0.6 and 1.2 after adjusting for the DC offset. In addition to the pulse shape discrimination, pulse height discrimination was performed to remove low amplitude signals that corresponded in the γ-flash ring and low energy α particles from the boron dopant in the PIPS detectors.

The LINAC was operated at 180 Hz with a beam energy of 55 MeV and an average current of 13 μA. $^{147}\text{Sm}$ was measured for 4.77 hr and $^{148}\text{Sm}$ for 12.74 hr. Experimental results from Glendenov et. al.$^{23}$ were broadened using (3). Data from $^{147}\text{Sm}$ was normalized to the broadened results of Glendenov et. al. between 300 eV and 2500eV. Results from $^{149}\text{Sm}$ were normalized to the $^{147}\text{Sm}$ measurement here by the run time.

III. RESULTS

Fig. 6. shows results from the measurement of the $^{147}\text{Sm}(n,\alpha)^{144}\text{Nd}$ cross section compared to broadened evaluations from ENDF/B-VII$^{24}$ and experimental results from Glendenov et. al.$^{23}$. The shape corresponds closely with results from Glendenov. Fig. 5. Shows this measurement of the $^{149}\text{Sm}(n,\alpha)^{146}\text{Nd}$ cross section compared to broadened evaluations from ENDF/B-VII$^{24}$. This measurement
does not show as much structure as ENDF predicted. It seems that due to the lack of data, the evaluations are simply a scaled down version of the total cross section.

IV. CONCLUSIONS

To improve stellar and nucleosynthesis models more accurate predictions for cross sections are needed. The current limiting factor for the accuracy of these models is knowledge of the α+nucleus potentials. α-scattering and high incident energy (n,α) measurements on heavy targets are not easily applicable to the energy and mass region focused on in astrophysics. The LSDS has proven that it is capable of measuring (n,α) reaction cross section of less than 1 mbarn in the intermediate mass range using sample masses ~10 mg without the need to devote months to a single experiment. This allows for the expansion of this method into short-lived radioactive samples and to other reactions such as (n,p).

ACKNOWLEDGEMENTS

The authors would like to thank the LINAC staff members for the operation and maintenance of the LINAC. This work was funded by the SSAA, part of the NNSA, under grant number DE-FG52-09NA29453.

REFERENCES

7. Y. Danon, R. C. Block, M. J. Rapp, and F. J. Saglime, G. Leinweber, D. P. Barry, N. J. Drindak and J. G. Hoole, Beryllium and Graphite


