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Cross-section measurements for ^{239}Pu(n,f) and ^{6}Li(n,α) with a lead slowing-down spectrometer

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Abstract

We present fission cross-section measurements with ~10 ng of ^{239}Pu performed using the LANSCE Lead Slowing-Down Spectrometer. Results of ^{6}Li(n, α) measurements with a sample size of 760 ng of ^{6}Li are also reported. This technical achievement demonstrates the feasibility of measuring neutron-induced fission cross-section on samples with 10 ng of fissile actinides that are available on ultra-small quantities. Furthermore, results on neutron-induced alpha emission show that measurements for astrophysics purposes are feasible with the LSDS.
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1. Introduction

The development of a method for neutron-induced fission measurements on ultra-small quantities of actinides (less than 10 ng) is motivated by the lack of experimental data on short-lived isotopes and short-lived isomers. For practical reasons such as high decay activity of the sample, only limited quantities of these elements can be handled. Therefore, conventional neutron sources do not have high enough neutron flux to induce acceptable count rates with such a small quantity of actinides.

An interesting example of a short-lived isomer is the metastable state of uranium 235 at 76.8 eV [1,2] with a half-life of 26 min. Different predictions [3,4] above the resonance region and up to several MeV say that the fission cross-section of this isomer is less than that of the ground state of ^{235}U by up to a factor of two. At thermal neutron energies, where the fission cross-section is dominated by nearby resonances, measurements [5–7] show just the opposite, with the cross-section of the isomer being larger by a factor of 1.4–2.5. Because there have been to date no measurements above thermal energy, it is important to validate the predictions [3,4] and to study the cross-section in the resonance region, where the fission cross-section cannot be calculated. Only small quantities can be obtained (by alpha decay of ^{239}Pu) and thus a high neutron flux is needed to measure the fission cross-section.

To answer this need, a Lead Slowing-Down spectrometer (LSDS) has been installed at the 800 MeV proton

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accelerator at the Los Alamos Neutron Science Center (LANSCE). After characterization of this spectrometer [8,9], different tests have been made to prepare the measurements with ^{235m}U targets. One of the most important tests is to demonstrate the feasibility of measuring the fission cross-section on a sample with less than 10 ng. To do so, we chose a fissile material with a fission cross-section comparable to that of ^{235g}U, namely ^{239}Pu. ^{235g}U was not chosen because it does not have a high enough natural decay rate to allow an easy measurement of the mass of a sample of a few nanograms.

Furthermore, a compensated ionization chamber for (n,α) reaction was tested and results are presented in this paper. Neutron-induced reactions that produce alpha particles are important in astrophysics. These reactions occur in nucleosynthesis networks related to the weak *s*-process [10]. For instance, for the calculation of stellar reaction rates, cross-section data for (n,α) reactions are needed for neutron energies from thermal up to a few hundred keV. Neutron-induced alpha reaction cross-sections are also important in nuclear transmutation and in radiation damage due to the accumulated helium gas. With its high neutron flux up to a few hundreds of keV, the LSDS is a good candidate for (n,α) cross-section measurements related to astrophysics and first measurements with a ^{6}Li target are investigated.

The capabilities of the LSDS system need to be presented in the context of beam-target facilities. One example is a study with neutron beams on small targets of neptunium isotopes [11]. In that work the details of the samples, their half lives, the maximum neutron energy reported, and the cross-section at that energy are given in Table 1. The neutron source was an intense, spallation neutron source that is highly moderated to give an approximately $1/E$ spectrum. Because the cross-section is also dropping off with increasing energy of the incident neutron, the measurement had an upper limit of neutron energy.

For the ^{235m}U measurements, we will likely have about the same sample mass as these measurements on neptunium isotopes. The expected cross-section is smaller by a factor of $\simeq 2$ at the energies of E_n^{max} and drops off to about 1 barn at 100 keV. Because of the short half-life of ^{235m}U, the measuring time will be less than in Ref. [11] by a factor of 10 or more. We wish to measure the ^{235m}U fission cross-section up to 100 keV, and if the neutron flux had a $1/E$ spectrum, there would be another factor of 10 decrease in the neutron flux at this energy compared with E_n^{max} in the ^{236}Np measurement. Thus, we would be looking at a factor of at least $6 \times 10 \times 10 = 600$ increase in difficulty for the ^{235m}U measurement compared with the neptunium beam-target experiment.

Table 1
Characteristics of the targets, beam energy and fission cross-sections from Ref. [11]

Isotope	Half-life	$E_n^{max.}$	Cross-section (barns) at $E_n^{max.}$
^{236}Np	1.18×10^5 y	10 keV	8
^{238}Np	2.1 d	100 eV	~20

In this article we describe the LSDS facility followed by a discussion on the fission cross-section measurements of a 9.87 ng sample of ^{239}Pu and of ^{6}Li(n,α) cross-section with a sample size of 760 ng of ^{6}Li.

2. Neutron source

Detailed descriptions of the LSDS at LANSCE can be found elsewhere [8,9]. In order to measure the small amount of ^{235m}U that can be made available (10 ng), a high neutron flux is required. Following previous simulations [9], a time-average proton beam intensity of 1 μA and a repetition rate of at least 20 Hz are needed. Even with the greatly increased flux in the LSDS, many samples (~10) of the isomer must be measured to obtain sufficient statistics. Different challenges arise from the use of a spallation source at such proton beam intensity and special detector developments are needed to work in this environment.

The main advantage of the LSDS over a conventional time-of-flight method is the high flux intensity achieved by the neutron slowing down process in lead. The 800 MeV proton beam produced by the LANSCE accelerator can be directed to the LSDS through the Proton Storage Ring (PSR). The PSR compresses the 750-μs proton pulses from the linac into pulses of typically 250–300 ns. At the center of the high purity lead cube of 1.2 m on a side, the proton beam hits a tungsten target and neutrons are produced by spallation process.

As shown previously in the literature (e.g. Ref. [12]), the neutron energy-time relation can be expressed as

$$\bar{E} = \frac{K}{(t + t_0)^2} \tag{1}$$

where $\bar{E}$ is the mean neutron energy in keV, K is the slowing-down constant in keV μs^2, t is the measured time in μs after the proton pulse and t_0 is a constant also in units of μs. In the case of the LSDS at LANSCE, the two constants K and t_0 are measured to be 161 ± 1 keV μs^2 and 0.37 ± 0.15 μs, respectively [9].

As presented in previous works [9], the production of neutrons in the LSDS is accompanied by high-energy charged particles and gamma rays from the tungsten target and from secondary reactions in the lead (so-called "γ-flash"). The flash appears during the spallation process and its intensity increases with the proton beam intensity. The intense initial gamma and neutron flash interacts with the detector materials and sample and causes saturation that can last for a few hundreds of nanoseconds to several microseconds. This limits the measurements to neutron energies of less than 10 or 100 keV, depending on the proton beam intensity. A common solution is to use a compensated detector [13,14]. The results are presented in the following paragraphs.

The neutron fluence was obtained from a fit of the MCNPX calculations [9] with the following function, obtained from previous measurements [15]:

$$\Phi(E) = E^{-a} \exp\left(-\sqrt{\frac{b}{E}}\right) \tag{2}$$

where E is the neutron energy in eV, Φ is the neutron fluence and a and b are two adjustable parameters. In the case of the LSDS, a and b are equal to 0.506 and 0.22 eV, respectively. This neutron fluence was previously used for measurements with large samples (fission with tens of μg of ^{235}U from 0.1 and 20 keV) and results showed good agreement with the expected cross-section [9].

3. Measurement on nanogram sample for ^{239}Pu(n,f)

3.1. Compensated fission chamber

In order to minimize the effect of the γ-flash on the fission detectors, the well-known method of compensation is used. Two types of detectors are used in our work, a compensated solar cell and a compensated fission chamber. The description of the compensated solar cell is given in Ref. [16] and will not be detailed here. The compensated fission chamber (CFC) consists of two parallel plate ionization chambers with the fissionable sample in one of the chambers. The chambers are arranged so that the difference in signals is recorded. Gamma rays or particles produced from the spallation process travel through the two chambers almost equally and therefore cancel in the difference signal. Fission events, on the other hand, take place only in one chamber and remain in the difference signal as desired events.

The compensated fission chamber was tested in the LSDS with 9.87 ng of ^{239}Pu, 0.8 cm in diameter, deposited onto a titanium foil of 3.4 cm in diameter, 0.51 mm thick. The distance between plates was $\simeq$ 5 mm, with a voltage on each plate of $\pm \simeq 200$ V. The preamplifier used was a Cremat model CR 110, modified to have 1 μs fall time [17]. The P-10 gas was used in the chamber with a constant flow at 1 LANL atm (0.78 atm at sea level). The proton beam intensity was slowly increased and the compensation was adjusted by a variation of the voltage on the parallel plates.

The data acquisition system consisted of waveform digitizers (Acqiris DC265 with a maximum sampling rate of 1 GS/S), triggered by the timing signal of the proton pulse. The time-of-flight signals from the detector were recorded, after passing through a discriminator. The waveforms were analyzed in real time to extract the leading edge with a resolution of 50 ns. The time spectra were recorded using the MIDAS data acquisition system with the ROOT analysis and display package [18,19].

3.2. Fission cross-section of ^{239}Pu

The neutron-induced fission cross-section of ^{239}Pu was measured with the compensated fission chamber as a test for the $^{235\text{m}}$U measurements. The sum of different runs with the 9.87 ng deposit are presented in Fig. 1. The proton beam frequency was 20 Hz, for different time-averaged intensities from 500 nA to 1 μA.

For comparison, a previous measurement performed at a different position in the LSDS with a compensated solar cell and 27 ng of ^{239}Pu and 300 nA of proton beam intensity is presented, together with the evaluation from ENDF/B-VI.8, broadened with the measured LSDS energy resolution [9]. As the measurements are relative, the data were normalized to the ENDF/B-VI.8 broadened cross-section at 200 eV.

In the case of the 9.87 ng sample measurement, the detector and its electronics were submitted to a higher radiation environment than in the previous case at 300 nA. The direct consequence is a degradation of the compensation at short times after the proton pulse and the output signal is saturated. The detector and its electronics recover slower and become operational between 1 and 2 μs later ($E_n \simeq 10$ keV), whereas in the case of the measurement with 300 nA, the saturation disappears at about 1 μs after the proton pulse ($E_n \simeq 100$ keV).

In both cases, structures of the fission cross-section appear, especially at 10, 30 and 60 eV. At low neutron energy, the neutron flux is strongly decreasing, and the cross-section is slowly increasing. The count rate becomes lower and structures slightly appear. One can see that there is a difference between the broadened cross section and the measurements for neutron energies less than $\simeq$ 8 eV. The broadened cross-section (thick line in Fig. 1) is equal to the ENDF/B-VI.8 cross-section at a given neutron energy,

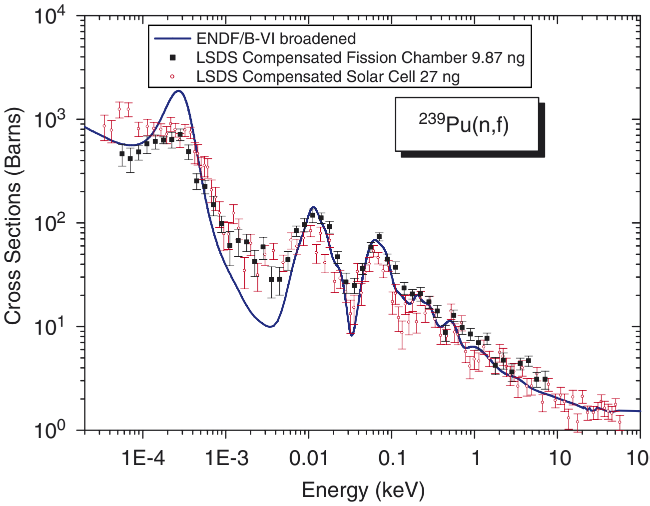

Fig. 1. Measured neutron-induced fission cross section of ^{239}Pu with the compensated fission chamber (9.87 ng deposit) and the compensated solar cell (27 ng deposit), compared to the broadened cross-section from ENDF/B-VI.8.

convoluted with a Gaussian function of area one, and with the ΔE obtained from the measured energy resolution of the LSDS [9]. One of the reasons for this difference is that the neutron flux can be locally modified by the hydrogen in the detector or the cabling. Also a better reproduction of the broadened cross-section, with a different function than a Gaussian, can improve the comparison. Furthermore, at low energy, the neutron flux can vary as a function of the position in the LSDS, which can explain the difference between the two datasets for $E_n < 0.1$ eV.

4. Measurement of ^{6}Li(n, α)

4.1. Compensated ionization chamber

A compensated ionization chamber (CIC) was placed in the LSDS to measure the ^{6}Li(n, α)^{3}H cross section as a feasibility test for further work. The CIC consists of three stainless steel plates, each 3.15 cm in diameter, mounted on ceramic rods and spaced 2 cm apart. The distance between the plates was chosen to collect nearly all the energy of the 2.05 MeV alpha particles emitted from a ^{6}Li(n, α)^{3}H reaction in P-10 at 0.78 atm. The energy deposited by the alpha particle in the chamber is then 0.9–2.05 MeV depending on the angle of emission and its interaction with the Teflon spacers. The triton has a range of 11.7 cm, which means that the maximum energy collected in the gas is about 0.42 MeV which is not enough to be seen above the noise level. Therefore, the effect of the tritons is neglected.

4.2. Cross-section for ^{6}Li(n, α)

A LiF sample of $91 \pm 29\,\mu\text{g/cm}^2$ with an area of $0.7\,\text{cm}^2$ ($1.1 \pm 0.4\,\mu$g of ^{6}Li) deposited on a 20 mil thick titanium backing was used as the cathode in the CIC with the sample facing the top plate. The proton beam current was 25 nA at 20 Hz. In order to actively compensate the chamber, a positive bias of 200 V was placed on the top plate, and a negative bias of 400 V was placed on the bottom plate. The compensation of the gamma-flash was not ideal and a dead time of about 7 µs was observed. These results are shown in the top plot in Fig. 2 [20].

The bias was filtered through a 20 MΩ resistor with a 47 nf capacitor to ground. The signal from the cathode was then filtered through a 10 nf capacitor with a 20 MΩ resistor to ground prior to reaching the preamplifier. The charge sensitive preamplifier was a Cremat CR-110 with a shaping constant of 140 µs and a gain of 1.4 V/pC.

The sample was then replaced with $62 \pm 7\,\mu\text{g/cm}^2$ with an area of $0.7\,\text{cm}^2$ of LiF ($0.76 \pm 0.09\,\mu$g of ^{6}Li) and the proton beam current was increased to 300 nA. This increased the dead time to 20 µs due to the larger gamma-flash. The top plate bias was −400 V and the bottom plate bias was 300 V to provide compensation. These results are shown in the bottom plot of Fig. 2. As seen in figure, the compensation did not work well at short times (highest energies) probably due to the large resistor in the filter prior to the preamp which allowed the cathode to hold a charge. Future experiments will be conducted with a filter with a modified filter arrangement that includes a smaller resistor to ground.

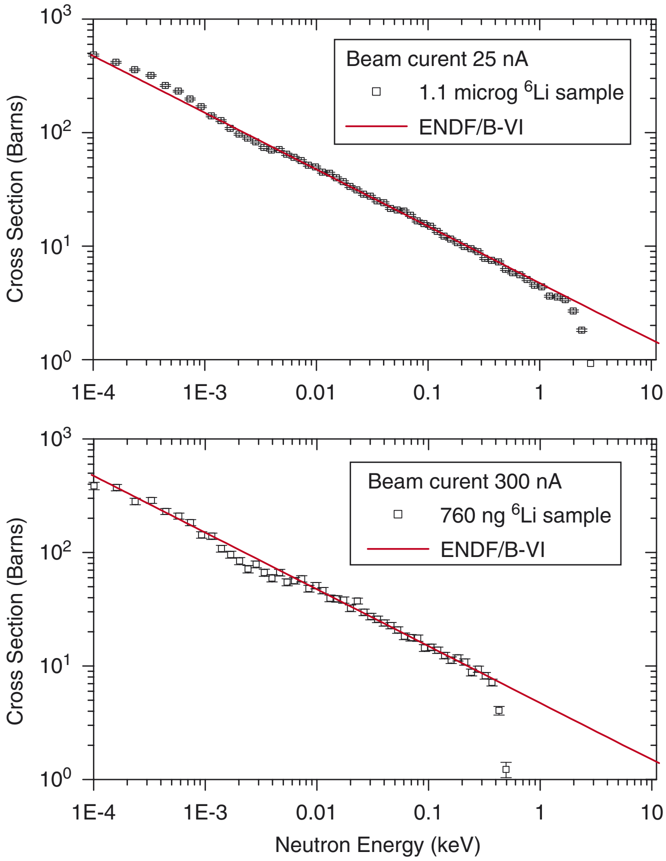

Fig. 2. Experimental results, normalized to the evaluated library, for the 25 nA and 300 nA proton beam for ^{6}Li(n, α).

There are some areas where the cross-section varies from a straight $1/v$ value. The drop in the cross-section between 300 and 1000 eV is due to electronic dead time that was not compensated for in this data. The dip at about 2–4 eV in both plots may be due to neutron interactions in the material of the chamber. This will be investigated further through mathematical models of the flux in the area of the detector (interactions with the material of the detector itself).

5. Conclusion and perspective

Measurements of the neutron-induced fission cross-section on samples of ^{239}Pu as small as 9.87 ng for neutron energies from 0.1 eV to 100 keV have been demonstrated with a Lead Slowing-Down Spectrometer driven by the 800 MeV pulsed proton beam of the Los Alamos Neutron Science Center. The ^{6}Li(n, α) reaction has also been studied in this range with samples smaller than 1 µg. For the ^{239}Pu(n,f) reaction, the experiments were carried out with 1 µA average proton current, and the results demonstrate the feasibility of measuring the neutron-induced fission cross section of ^{235m}U. In both cases, the use of compensated detectors shows that the saturation due to the so-called gamma-flash can be strongly reduced.

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