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Measurements with the high flux lead slowing-down spectrometer at LANL

Y. Danon [a,*], C. Romano [a], J. Thompson [a], T. Watson [a], R.C. Haight [b], S.A. Wender [b], D.J. Vieira [b], E. Bond [b], J.B. Wilhelmy [b], J.M. O'Donnell [b], A. Michaudon [b], T.A. Bredeweg [b], T. Schurman [b], D. Rochman [c], T. Granier [d], T. Ethvignot [d], J. Taieb [d], J.A. Becker [e]

[a] *Rensselaer Polytechnic Intstitute, 110 8th Street, Troy, NY 12180, USA*
[b] *Los Alamos National Laboratory, Los Alamos, NM 87545, USA*
[c] *Brookhaven National Laboratory National Nuclear Data Center (NNDC), Upton, NY 11973-5000, USA*
[d] *CEA-DAM, BP 12, 91680 Bruyères-le-Châtel, France*
[e] *Lawrence Livermore National Laboratory, Livermore, CA 94550, USA*

Abstract

A Lead Slowing-Down Spectrometer (LSDS) was recently installed at LANL [D. Rochman, R.C. Haight, J.M. O'Donnell, A. Michaudon, S.A. Wender, D.J. Vieira, E.M. Bond, T.A. Bredeweg, A. Kronenberg, J.B. Wilhelmy, T. Ethvignot, T. Granier, M. Petit, Y. Danon, Characteristics of a lead slowing-down spectrometer coupled to the LANSCE accelerator, Nucl. Instr. and Meth. A 550 (2005) 397]. The LSDS is comprised of a cube of pure lead 1.2 m on the side, with a spallation pulsed neutron source in its center. The LSDS is driven by 800 MeV protons with a time-averaged current of up to 1 μA, pulse widths of 0.05–0.25 μs and a repetition rate of 20–40 Hz. Spallation neutrons are created by directing the proton beam into an air-cooled tungsten target in the center of the lead cube. The neutrons slow down by scattering interactions with the lead and thus enable measurements of neutron-induced reaction rates as a function of the slowing-down time, which correlates to neutron energy. The advantage of an LSDS as a neutron spectrometer is that the neutron flux is 3–4 orders of magnitude higher than a standard time-of-flight experiment at the equivalent flight path, 5.6 m. The effective energy range is 0.1 eV to 100 keV with a typical energy resolution of 30% from 1 eV to 10 keV. The average neutron flux between 1 and 10 keV is about 1.7×10^9 n/cm^2/s/μA. This high flux makes the LSDS an important tool for neutron-induced cross section measurements of ultra-small samples (nanograms) or of samples with very low cross sections. The LSDS at LANL was initially built in order to measure the fission cross section of the short-lived metastable isotope of U-235, however it can also be used to measure (n, α) and (n, p) reactions. Fission cross section measurements were made with samples of ^{235}U, ^{236}U, ^{238}U and ^{239}Pu. The smallest sample measured was 10 ng of ^{239}Pu. Measurement of (n, α) cross section with 760 ng of Li-6 was also demonstrated. Possible future cross section measurements include fission and (n, p) and (n, α) reaction in radioactive samples.

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1. Introduction

When it is desirable to measure the energy-dependent neutron-induced reaction cross sections of small samples or samples with very low cross sections a high neutron flux

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[*] Corresponding author. Tel.: +1 518 276 4008; fax: +1 518 276 4832.
E-mail address: danony@rpi.edu (Y. Danon).

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is required. An example of such measurement is the fission cross section of the metastable isomer of ^{235}U. The isomer has a short half life of 26 min and can be produced by chemically separating ^{235}U from is precursor ^{239}Pu [1]. Thermal and cold neutron cross section measurements of the ratio of the fission cross sections $R = \sigma^{235\mathrm{m}}/\sigma^{235}$ gave a value of 1.4–2.2 [2–4]. Calculations of R at 100 keV give a value of about 0.7 [5,6]. It is feasible to obtain a sample of only about 20 ng of ^{235}U with an isomer-to-ground state ratio of less than 0.8. Under these conditions it was shown in [1] that an LSDS measurement can be used to obtain the ratio R as a function of the neutron energy.

In the past measurements of short lived isotopes were also done using the time-of-flight (TOF) method with pulsed a neutron source [7]. In the case of fission cross section measurements and the TOF method, when a neutron does not interact with the sample it is lost and will not contribute any information to the experiment. The LSDS slows down neutrons by scattering interactions with the lead. This creates an isotropic flux inside the lead cube, and a single neutron can pass several times through the sample during the slowing-down process which increases its probability to interact with the sample. The relationship between the neutron energy and the slowing-down time was calculated and measured [1] to be:

$$E[eV] = \frac{161000}{(t+0.4)^2}, \tag{1}$$

where t is the slowing-down time in μs. The resolution of the LSDS is less than 40% from 1 eV to 1.5 keV. The resolution function obtained by fitting the data in Fig. 6 of [8] and is given by a variation of the "bath tub" [8] curve

$$\left(\frac{\Delta E}{E}\right)_{\mathrm{FWHM}} = \sqrt{\left(0.041 + \frac{0.12}{E} + 0.003\sqrt{E}\right)}, \tag{2}$$

where E is the neutron energy in eV and is valid in the energy range of $0.1\ \mathrm{eV} < E < 100\ \mathrm{keV}$. The neutron flux was measured and calculated in [1] and using the analytical expression given in [9], the flux can be expressed as

$$\phi(E) = 2.2 \cdot E^{-0.506} \exp\left(-\sqrt{\frac{0.22}{E}}\right), \tag{3}$$

where E is in eV and ϕ is in units of neutron/cm^2/MeV/proton

2. Results

After the initial characterization of the spectrometer [1] it was observed that the flash associated with the proton pulse is high and causes saturation of the detectors and associated electronics. In order to overcome this problem compensated solar cells and ionization detectors were developed [10]. Also the LANSCE pulse repetition rate was increased from 20 Hz to 40 Hz to reduce the number of protons per pulse for a given average proton current. The detectors are connected to fast preamps and shaping amplifiers that were purchased from Cremat [11]. Fast recovery of the detector and electronics is required in order to be able to measure data up to energy of 100 keV which occurs at a slowing-down time of 0.87 μs. The successful use of these compensated detectors was initially tested on the measurements of the fission cross section of ^{235}U [1]. The smallest sample measured so far is 9.78 ng of ^{239}Pu [12]. Recently we also measured the sub threshold fission in a 3 mg (∼1 ppm ^{235}U) sample of ^{238}U shown in Fig. 1. In our recent experiments several attempts to measure the fission cross section of $^{235\mathrm{m}}$U were made; however the sample was not pure and contained ^{233}U. It is estimated that the sample mass included about 5 ng of uranium including both the ground and exited state. Improvements in the chemistry are still required, and difficulties arise due to a compromise between the sample size and the chemical process speed.

We also explored the possibility of using the LSDS for measurements of the (n, α) cross section with small samples or with larger samples of isotopes with small cross sections [14]. Our initial measurements were done on ^{6}Li using a compensated ion chamber and LiF samples. The smallest sample measured contained 760 ng of ^{6}Li and the 1/v (n, α) cross section was measured up to a neutron energy of 2 keV at low proton beam current (25 nA). At high beam power (0.5 μA) the upper energy limit was reduced to 0.6 keV due to overload from the intense flash.

Recently we repeated this measurement with a compensated solar cell detector that was configured to measure alpha particles. In order to improve the energy sensitivity of the solar cell a smaller cell was used and was coupled to a fast Cremat amplifier with an integration constant that matches the fast pulses of the solar cells. The initial results

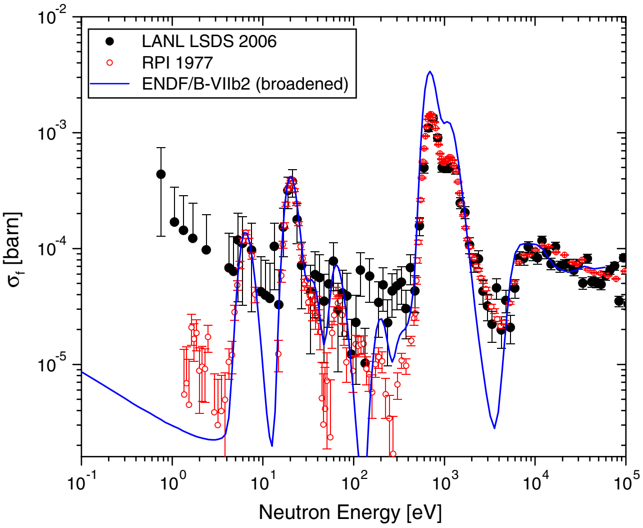

Fig. 1. Sub threshold fission in U-238 compared to the broadened ENDF/B-VIIb2 cross section and an LSDS measurement at RPI [13]. We believe that the higher values of the present measurement in the cross section minima are due to an incorrect discriminator setting that did not entirely eliminate background pulses.

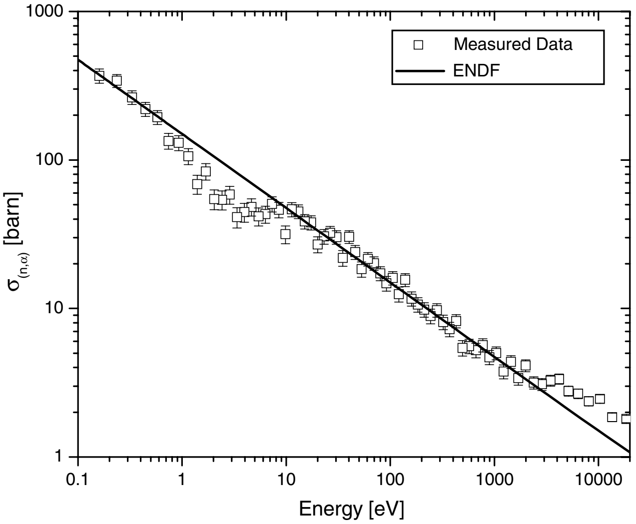

Fig. 2. The (n,α) cross section of ^{6}Li in a LiF sample measured with a compensated solar cell detector.

from this detector are shown in Fig. 2. The data were taken with a proton current of 30 nA and a pulse repetition rate of 8 Hz. The LiF mass was about 21.6 μg (270 ng Li-6). The upper neutron energy limit of the detector was extended to 20 keV. The structure in the cross section between 1 and 10 eV indicates a problem and requires further development.

3. Summary

A 1.2 m cube LSDS coupled to an 800 MeV spallation neutron source at LANSCE is demonstrated to be a powerful high neutron flux tool for energy-dependent cross section measurements. The LSDS was fully characterized; the slowing-down-time energy relation, the energy resolution and the flux shape are well known from both measurements and calculations. Compensated detectors based on solar cells and ionization chambers and the associated fast electronics were developed and tested during measurements of the fission cross section of ^{235}U, ^{238}U and ^{239}Pu and the (n,α) cross section of ^{6}Li. The high flux of the spectrometer enables measurements of ultra-small samples as was demonstrated in a measurement of the fission cross section of a 9.8 ng sample of ^{239}Pu.

Further developments of the detectors and chemical separation techniques are in progress to enable the measurement of the 26 min half life isomer ^{235m}U. Further development of (n,α) and (n,p) detectors is also in progress which will enable cross section measurements of a variety of samples that will help the understanding of astrophysical processes.

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