# First Measurements with a Lead Slowing-Down Spectrometer at LANSCE

D. Rochman<sup>1</sup>, R. C. Haight<sup>1</sup>, S. A. Wender<sup>1</sup>, J. M. O'Donnell<sup>1</sup>, A. Michaudon<sup>1</sup>, K. Huff<sup>1</sup>, D. J. Vieira<sup>2</sup>, E. Bond<sup>2</sup>, R. S. Rundberg<sup>2</sup>, A. Kronenberg<sup>2</sup>, J. Wilhelmy<sup>2</sup>, T. A. Bredeweg<sup>2</sup>, J. Schwantes<sup>2</sup>, T. Ethvignot<sup>3</sup>, T. Granier<sup>3</sup>, M. Petit<sup>3</sup>, and Y. Danon<sup>4</sup>

<sup>1</sup>LANSCE-3, Los Alamos National Laboratory, Los Alamos, NM 87545, USA
<sup>2</sup>C-INC, Los Alamos National Laboratory, Los Alamos, NM 87545, USA
<sup>3</sup>CEA-DAM, BP 12, 91680 Bruyères-le-Châtel, France
<sup>4</sup>Rensselaer Polytechnic Institute, 110 8th St., Troy, NY 12180, USA

**Abstract.** The characteristics of a Lead Slowing-Down Spectrometer (LSDS) installed at the Los Alamos Neutron Science Center (LANSCE) are presented in this paper. This instrument is designed to study neutron-induced fission on ultra small quantities of actinides, on the order of tens of nanograms or less. The measurements of the energy-time relation, energy resolution and neutron flux are compared to simulations performed with MCNPX. Results on neutron-induced fission of <sup>235</sup>U and <sup>239</sup>Pu with tens of micrograms and tens of nanograms, respectively, are presented. Finally, a digital filter designed to improve the detection of fission events at short time after the proton pulses is described.

#### **INTRODUCTION**

Knowledge of cross sections for neutron-induced fission of actinides is important for understanding processes that occur in reactors, accelerator-driven systems and nuclear explosions. To compensate for low count rates for measurements with conventional time-of-flight techniques on small quantities of actinides with short half-lives, a very high neutron flux is needed. In this goal, a Lead Slowing-Down Spectrometer is being developed at LANSCE [1]. An interesting example of a short-lived isomer is the first isomer of uranium 235 with a half-life of 26 min. Until now, only the ratio of the isomer fission cross section to the ground-state fission cross section for thermal and cold neutron energies has been measured [2,3]. In the laboratory, this isomer can be produced from the alpha-decay of  $^{239}$ Pu with an equilibrium ratio being  $2x10^{-9}$ . It is therefore a good candidate for measurements with the LSDS. The characteristics of the LSDS and first measurements on microgram and sub-microgram samples are presented here.

#### **CHARACTERISTICS OF THE LSDS**

The basic features of the LSDS are as follows. It is a 1.2 m cube and consists of 36 blocks, each 40x40x30 cm<sup>3</sup> with masses of 542 kg, made from high-purity lead. A chemical analysis shows that impurities are present in the lead: silver, antimony and cadmium. In each block a channel allows samples and detectors to be inserted for measurements. At the center of the LSDS, two channels facing each other serve as the entrance channel for the proton beam.

#### **Neutron Source**

The LSDS is located at the Los Alamos Neutron Science Center (LANSCE). The linear accelerator provides a beam of 800 MeV protons. To obtain short proton pulses, a 30 m diameter proton storage ring compresses the 750  $\mu$ s proton pulses from the linac into pulses of typically 250-300 ns. The neutron production is realized by spallation in a tungsten target. The tungsten target is a cylinder 25 cm long and

7 cm in diameter. The neutron spectrum from the tungsten is a typical spallation neutron flux, with a maximum at  $\approx 2$  MeV.

## **Energy-Time Correlation**

As the LSDS is mainly made of high-purity lead, neutrons lose a small quantity of energy at each elastic collision with a Pb nucleus; see Fig. 1.



**FIGURE 1.** Simulation with MCNPX [4] of the relative neutron density as a function of time (t=0 is the origin of the spallation process) at a given location in the LSDS. Note that for times greater than 1  $\mu$ s, there is a "focusing" of the neutrons to an approximate time-energy relationship.

A correlation exists between the slowing-down time and the neutron energy inside the lead block [5]:

$$\overline{E_n} = \frac{K}{\left(t + t_0\right)^2} \tag{1}$$

where  $\overline{E_n}$  is the mean neutron energy in keV, K is the slowing-down constant in keV.µsec<sup>2</sup>, t is the measured time in µsec after the source pulse and  $t_0$  is a constant also in units of µsec. The two constants K and  $t_0$  can be determined by simulations or experimentally. Simulations are performed with MCNPX, taking into account the lead impurities and the influence of the surrounding environment of the LSDS.

The results are  $K_{simu} = 161 \pm 5 \text{ keV.} \mu \text{sec}^2$  and  $t_{0,simu} = 0.4 \pm 0.1 \mu \text{sec}$ . Experimentally, the values of K

and  $t_0$  were measured by neutron capture on different samples. Gamma emission from the neutron capture was detected with a small cylindrical cerium fluoride scintillator. Each sample (In, Ta, Au, Ag, Cd, Co, Cu, Mn and Fe) has a large neutron capture resonances at characteristics neutron energies. Values for *K* and  $t_0$ are 161 ± 1 keV.µsec<sup>2</sup> and 0.4 ± 0.1 µsec, respectively. These values are in very good agreement with the simulations and will be used in the following.

## **Energy Dispersion**

The energy dispersion around the mean neutron energy is an important characteristic of the spectrometer. It has been shown in many references (for instance in [6]) that in the ideal case, the energy resolution  $\Delta E/\overline{E_n}$ , with  $\Delta E$  full width at halfmaximum can be as small as 30%. The energy resolution was experimentally evaluated by  $(n,\gamma)$ measurements on well-defined resonances with a cerium fluoride scintillator. These isotopes were the same as those used to determine K and  $t_0$ . Results are shown in Fig. 2, together with MCNPX simulations.

As seen in the figure, the measurements and simulations agree well over a wide energy range. The influence of the impurities results in a small but significant increase of the energy resolution. The simulation with impurities is in fact closer to the measurements, which reflects the importance played by the impurities even at the low concentrations in the present spectrometer.



**FIGURE 2.** Comparison of the measured  $\Delta E/\overline{E_n}$  by  $(n,\gamma)$  reactions and MCNPX simulations.

## **Neutron Flux**

The neutron flux is an important parameter of the spectrometer that is necessary to determine a cross section. It can be simulated with MCNPX at different positions in the lead volume. The results in  $n/cm^2/MeV/proton$  can then be compared to measurements. Two sorts of measurements were performed to evaluate the absolute neutron flux and its shape: by activation and with a <sup>235</sup>U fission chamber. Three materials were chosen for the activation measurements: Au, In and W. We used a <sup>235</sup>U fission chamber with a large amount of <sup>235</sup>U:  $803\mu g \pm 2\%$ , known with precision and usually used for absolute measurements in reactor environment. The results are presented in Fig. 3, for a given position in the LSDS.

The shape and absolute values of the measured neutron flux are in good agreement with the MCNPX simulations. This neutron flux is then used to extract fission cross sections as shown in the next section.

# FISSION MEASUREMENTS ON SMALL SAMPLES

One of the main goals of the LSDS at LANSCE is to measure the fission cross section on very small quantities of the <sup>235m</sup>U isomer (10 ng). First, however, we chose larger samples of long-lived actinides to characterize the LSDS further and to develop measurement techniques. Two types of detectors were used: compensated fission chamber and compensated solar cell.



**FIGURE 3.** Example of a measured and simulated flux at a given position in the LSDS.

The goal of the compensation is to minimize the saturation of the detectors because of gamma rays reaching the detector at short times. Three results are presented in Fig. 4, (1) with 70  $\mu$ g of <sup>235</sup>U in the compensated fission chamber, measurements were taken over 8 hours at 40 nA of proton beam intensity at 20 Hz; (2) with 35 ng of <sup>239</sup>Pu in the compensated fission chamber, over 3 hours of measurements, at 300 nA and at 20 Hz; (3) with 27 ng of <sup>239</sup>Pu in the compensated solar cell detector, over 3 hours of measurements, at 300 nA and at 20 Hz; (3) nA and at 20 Hz.

For the <sup>235</sup>U data, the agreement between ENDF/B-VI broadened with the LSDS energy resolution and the measurement is good, except between 2 and 4 eV due to a higher energy resolution than expected. For the <sup>239</sup>Pu measurements, even with the statistics obtained in three hours of measurements, structures are observed at 10 and 70 eV. The beginning of the resonance at 0.3 eV is not clearly seen, because of poor statistics, and poor energy resolution. No background subtraction was possible because of the low count rate.

#### **DIGITAL FILTERING**

One of the main difficulties in using a spallation neutron source at the center of the LSDS is to overcome the saturation of the detector at short times after the proton pulse. The fission chamber as well as the solar cell used to measured fission events are saturated for 1 to 1.5 µs after the proton beam pulse. This limits the measurements to about 100 keV in neutron energy, following Eq. (1). To answer this problem, compensated detectors were designed, but the improvement of the data by an online analysis with digital filtering is also being explored. Preliminary results were obtained by analyzing offline 5000 waveforms. The digital filter consists of a double derivative plus a smoothing function. Comparison between the fission cross section of <sup>235</sup>U obtained with and without the digital filter is presented in Fig. 5. The advantage of the digital filter is evident. The fission cross section of <sup>235</sup>U can therefore be obtained up to 200 keV, compared to the 10-20 keV limit obtained without filter.



**FIGURE 4.** Fission cross section for <sup>235</sup>U (top) and <sup>239</sup>Pu (bottom). The data for the compensated solar cell are divided by 10 to avoid overlap.



**FIGURE 5.** Example of cross section obtained with and without digital filter.

## CONCLUSION

The characteristics of the Lead Slowing-Down Spectrometer now located at LANSCE were measured with different methods and simulated with MCNPX. Measurement and simulations are in good agreement. Fission cross section from measurements on submicrogram samples were obtained. Finally, preliminary studies with a digital filter show improvements for the measurements at high neutron energy.

## REFERENCES

- Granier, T., Pangault, L., Ethvignot, T., Haight, R. C., Leroux, X., Méot, V., Patin, Y., Pras, P., Szmigiel, M., Rundberg, R. S. and Wilhelmy, J. B., *Nucl. Instrum. Methods Phys. Res. A* 506, 149 (2003).
- D'Eer, A., Wagemans, C., Nève de Mévergnies, M., Gönnenwein, F., Geltenborg, P., Moore, M. S. and Pauwels, J., *Phys. Rev. C* 38, 1270 (1998).
- Talbert, W. L., Jr., Starner, J. W., Estep, R. J., Balestrini, S. J., Attrep, M. Jr., Efurt, D. W. and Roensch, F. R., *Phys. Rev. C* 36, 1896 (1987).
- MCNPX version 2.5d, Los Alamos National Laboratory Reports LA-UR-03-5916 and LA-UR-02-2607, <u>http://mcnpx.lanl.gov</u>.
- Bergman, A. A., Isakov, A. I., Murii, I. D., Shapiro, F. L., Shtranikh, V. I. and Kazarnovsky, M., Proceedings of the First International Conference on Peaceful Uses of Atomic Energy, Geneva, Vol. 4, 135 (1955).
- Abándades. A et al., Nucl. Instrum. Methods Phys. Res. A 478, 577 (2002).