LA-UR-10- 10-05428

Approved for public release; distribution is unlimited.

Title: Actinide Neutron-Induced Fission Cross Section Measurements at LANSCE

Author(s): F. Tovesson, A.B. Laptev, T.S. Hill

Intended for: Manuscript for the 21st International Conference on the Application of Accelerators in Research and Industry (CAARI-2010)
August 8-13, 2010
Fort Worth, Texas

Los Alamos
NATIONAL LABORATORY
EST. 1943

Actinide Neutron-Induced Fission Cross Section Measurements at LANSCE

F. Tovesson[a], A.B. Laptev[a] and T.S. Hill[b]

[a]*Los Alamos National Laboratory, Los Alamos NM 87545, United States*

[b]*Idaho National Laboratory, Idaho Falls ID 83415, United States*

Abstract. Fission cross sections of a range of actinides have been measured at the Los Alamos Neutron Science Center (LANSCE) in support of nuclear energy applications in a wide energy range from sub-thermal energies up to 200 MeV. A parallel-plate ionization chamber are used to measure fission cross sections ratios relative to the ^{235}U standard while incident neutron energies are determined using the time-of-flight method. Recent measurements include the ^{233,238}U, $^{239\text{-}242}$Pu and ^{243}Am neutron-induced fission cross sections. Obtained data are presented in comparison with existing evaluations and previous data.

Keywords: Fission, cross section, actinides, fast neutrons, time-of-flight.

PACS: 25.85.Ec, 27.90.+b, 29.30.Hs, 29.40.Cs

INTRODUCTION

To fulfill present and future world energy demand and decrease world economy dependence on fossil fuel the use of nuclear energy should be widely expanded because it is a reliable source of "carbon-free" electricity production. The DOE Fuel Cycle R&D program is supporting research in the technologies foreseen for the next generation of nuclear reactors.

Sensitivity studies carried out as part of the FC R&D program have demonstrated the need to improve the accuracy of some of the nuclear data used for predicting fuel properties and behavior. A program to provide fission cross sections with the required accuracies has been ongoing at the Los Alamos Neutron Science Center (LANSCE) [1] for the last few years. Below we present a general overview of the experimental program and the technical approach used, as well as new result for the ^{233}U and ^{238}U fission cross sections and preliminary result for recent measurements of the ^{243}Am.

EXPERIMENTAL METHOD

The fission cross section measurements at LANSCE are done at a white neutron source, and the time-of-flight (TOF) technique is used to determine the incident neutron energies. The cross sections are measured using ionization chambers relative to $^{235}U(n,f)$, which is a standard at thermal energies and in the range from 0.15 to 200 MeV.

The LANSCE Neutron Source

An 800-MeV linear proton accelerator drives the spallation targets at LANSCE [1], Fig. 1. The Weapons Neutron Research (WNR) facility gets proton pulses from the accelerator delivered to target 4, which is a bare tungsten target. The pulse repetition rate is 40 Hz, and each macro-pulse has micro-pulse structure of typically 1.8 μs spaced, 150 ps wide proton pulses. The 90L flight path at WNR is used for fission cross section measurement, and is the closest flight path to the spallation target. The fission chambers are at a nominal distance of 10 meters from the spallation target. When running 1.8 μs spacing the lowest accessible neutron energy is 0.15 MeV, and the highest accessible neutron energy is around 200 MeV.

The Lujan Center uses LANSCE target 2, which is surrounded by different moderators. The proton beam repetition rate is 20 Hz, and each pulse is about 250 ns wide. Flight path 5 at the Lujan Center is used for the fission cross section measurements, and the fission chambers are located about 8 meters from target 2 on this flight path. A water moderator shapes the neutron spectrum on this flight path, and the usable neutron energy range is from sub thermal energies, around 1 meV, to 200 keV. The lower limit is set by limited statistics, and the higher limit is set by the gamma shower from spallation and the ability to resolve neutron energies. More information on the neutron spectrum for flight path 5 is found in Ref. [2].

Fission Chambers

The same type of parallel plate ionization chambers commonly used for neutron flux monitoring at WNR is employed for the fission cross section measurements, and the detailed description of the detectors are found in Ref.

[3]. The chambers can hold up to four samples, and each sample is mounted in identical geometry in order to perform relative cross section measurements. The chambers typically provide about 1.2 ns (FWHM) timing for fission fragments, and measured the energy loss of particles in the active volume. The gas gap between electrodes is about 14 mm, so only part of total kinetic energy of the fission fragments and alpha-particles is measured for most tracks. The measured energy loss is used for identify fission and reject alpha-decay and other backgrounds as illustrated in Fig. 2.

Samples

The samples used for fission cross sections are by necessity very thin, typically below 200 μg/cm^2, so that the fission fragment emitted can escape the material an generate ionization in the active volume of the detector. The efficiency for fission detection depends on the thickness of the target, and at the nominal thickness the detection efficiency is about 97-98%.

The ^{243}Am sample was produced at Idaho Nation Laboratory using electro deposition, and has a total mass of approximately 200 μg. The sample still needs to be characterized using alpha-counting. The sample preparation procedure is described in Ref. [4]. The ^{233}U and ^{235}U samples were also prepared from electro deposition and had masses of 18.7 and 15.8 mg, respectively.

Data Acquisition System

The chambers are read out with fast preamplifiers (from RIS Corp.), and NIM electronics is used for signal treatment. A CAMAC system with FERA bus readout is used to digitize the data which is then transferred to a computer. The system has a relatively low dead time, typically on the order of 0.5-5%.

ANALYSIS

The online data is stored by the MIDAS software package, and is converted to ROOT trees in the post-analysis.

Fission identification

Fission is identified using software threshold on the fission chamber pulse height. The fission fragments are generally well separated from other events at seen inFig. 2. The pulse height spectrum from decay radiation is typically seen as a low energy peak in the ADC spectra, and is easy to measure in long beam-off counting. The correction for this type of background is therefore straight forward and associated with relatively small uncertainties. Another, similar type of background is neutron-induced charged particle emission form the backing foils. The response to these events is investigated using data from empty backings, and has been shown to no extend significantly into the ADC peaks produced by fission.

Dead Time

The data acquisition system (DAQ) has a fixed dead time of about 17 μs after each event, which is mainly driven by the conversion time of the analogue–to-digital (ADC) conversion. Each foil in the fission chambers is treated by a separate electronic chain, such that the dead times for the different foils are un-coupled.

The dead time correction needed in each measurement is determined by using hardware scalers in the DAQ system. The total number of events is scaled, as well as the number of events accepted by the DAQ, which is used to determine the integral dead time. A time-independent correction is applied to the data collected at flight path 90L, and a time-dependent correction is applied to the data collected at flight path 5, as described in Ref. [5].

Neutron Background

In addition to the neutrons that reach the fission chambers with a one-to-one correlation between time-of-flight (TOC) and kinetic energy, there are background neutrons with unambiguous relation between TOF and energy. This gives rise to background events in the observed fission spectrum, which needs to be corrected for.

One source of neutron background is "frame overlap", due to the short spacing between proton pulses at WNR. With only 1.8 μs between pulses, some fast neutrons will arrive at the same time as slow neutron from a preceding pulse which leads to ambiguity in the TOF-energy relation. At Lujan Center the pulse spacing is 50 ms, so this problem is eliminated.

Another type of background that only affect WNR experiments is "dark current", which is neutrons produced by proton leakage between pulses. This produces a background of white neutrons that can in some situations make up 1% of the total neutrons.

Neutron scattering around the flight path cave, or "room scattering", affects measurement both at WNR and Lujan Center.

The events from neutron background are carefully corrected for. The "dark current" events are observed in non-fission targets below the fission threshold, and can thus be quantified and subtracted. The "frame overlap" events are modeled and fitted after last micro-pulse in each macro-pulse, as described in Ref. [6]. The room background was investigated by measuring event rates with the fission chambers outside of the neutron beam.

Cross Section Calculation

The normalized ratio of events is used to calculate the measured fission cross section as a ratio to ^{235}U(n,f). The ratio is then converted to a cross section using the standard evaluation of ^{235}U(*n,f*) [7], which extends up to 200 MeV.

RESULTS AND CONCLUSIONS

The ^{233}U fission cross section measured at WNR is shown in Fig. 3. The same target was also measured at Lujan Center, so the complete data sets extend down to thermal energies. The low energy data is still being analyzed. The ENDF/B-VII and JENDL-3.3 evaluations are fairly consistent for this reaction, as expected from the large volume of experimental data available. The more interesting part of the current dataset is above 30 MeV, where only two previous data sets are available in EXFOR; the measurement by Lisovski et al. [8] and Shcherbakov et al. [9]. The current results are in very close agreement with those of Shcherbakov et al. at the high energy end, while the Lisowski results are slightly higher.

The ^{238}U results above threshold are shown in Fig. 4. The current results are generally higher than the ENDF/B-VII evaluation by about 2%, which is within the normalization uncertainty of the measurement. Above 20 MeV the evaluation is based on the data from Lisowski et al., and the current results are in good agreement with the evaluation. However, the cross sections measured by Shcherbakov et al. [9] are higher by about 8% in this energy region.

The preliminary result of the ^{243}Am fission cross section measurement is shown in Fig. 5. This is only a shape measurement at this point, since the sample needs to be better characterized. The cross section has been arbitrarily normalized to the average ENDF/B-VII value for first chance fission. For this reaction the absolute value of the cross section for first-chance fission is actually of interest, since values reported in literature have large discrepancies. Recently several measurements of the ^{243}Am fission cross section were done, for example, Laptev et al. [10] and Aiche et al. [11], but absolute normalization still needs to be proved. The current measurement will hopefully be useful in resolving existing discrepancy in worldwide data once the sample mass have been determined.

The results presented here are important a variety of nuclear applications and uncertainties are generally sufficiently low to meet the requirements for those applications. However, in some cases uncertainties below what is attainable with the present techniques are required. An ongoing activity is therefore to develop better detectors for fission measurements that would help reduce systematic uncertainties. A Time Projection Chamber is currently being developed in collaboration between 3 national laboratories and 6 universities [12] for fission measurements, and is planned for use in cross section measurements at LANSCE.

ACKNOWLEDGMENTS

This work has benefited from the use of the Los Alamos Neutron Science Center at the Los Alamos National Laboratory. This facility is funded by the US Department of Energy and operated by Los Alamos National Security, LLC under contract DE-AC52-06NA25396.

REFERENCES

1. P. W. Lisowski and K. F. Schoenberg, *Nucl. Instr. and Meth. A* **562**, 910-914 (2006).
2. M. Mocko, G. Muhrer, and F. Tovesson, *Nucl. Instr. and Meth. A* **589,** 455-464 (2008).
3. S. A. Wender et al., *Nucl. Instr. And Meth. A* **336**, 226-231 (1993).
4. J. D. Baker, C. A. McGrath, T. S. Hill, R. Reifarth, and F. Tovesson, *J. Radioanalytical Nucl. Chem.* **276**, 555-560 (2008).
5. F. Tovesson and T. S. Hill, Phys. Rev. C **75**, 034610 (2007).
6. F. Tovesson and T. S. Hill, *Nucl. Sci. and Eng.* **159**, 94-101 (2008).
7. A. D. Carlson et al., *Nucl. Data Sheets* **110**, 3215-3324 (2009).
8. P. W. Lisowski, A. Gavron, W. E. Parker, S. J. Balestrini, A. D. Carlson, O. A. Wasson and N. W. Hill, "Fission cross sections ratios for $^{233,\ 234,236}$U relative to ^{235}U from 0.5 to 400 MeV," *Conf. Nucl. Data for Sci. and Technol.*, Juelich, Germany, 1991.

9. O. A. Shcherbakov, A. Donets, A. Evdokimov et al., *J. Nucl. Sci. Technol.*, **Supplement 2**, 230-233 (2002).

10. A. B. Laptev, O. A. Shcherbakov, A. S. Vorobyev, R. C. Haight and A. D. Carlson, "Fast neutron-induced fission of some actinides and sub-actinides" in *Fission and Properties of Neutron-Rich Nuclei*, edited by J. H. Hamilton, A. V. Ramayya, and H. K. Carter, Proc. Fourth Int. Conf., Nov. 11-17, 2007, Sanibel Island, Florida, USA. World Scientific, New Jersey, 2008, pp. 462-468.

11. M. Aiche, G. Kessedjian, G.Barreau et al., "Quasi-absolute neutron-induced fission cross section of ^{243}Am" in *ND2007*, edited by O. Bersillon, F. Gunsing, E. Bauge, R. Jacqmin, and S. Leray, Proc. Int. Conf. on Nucl. Data for Sci. and Technol., April 22-27, 2007, Nice, France. EDP Sciences, 2008, pp. 483-485.

12. M. Heffner, "The Fission Time Projection Chamber Project" in *ND2010*, edited by Hyeong Il Kim and Young-Ouk Lee, Abstract Book of the International Conference on Nuclear Data for Science and Technology, April 26-30, 2010, Jeju Island, Korea. Korea Atomic Energy Research Institute, 2010, p. 143.

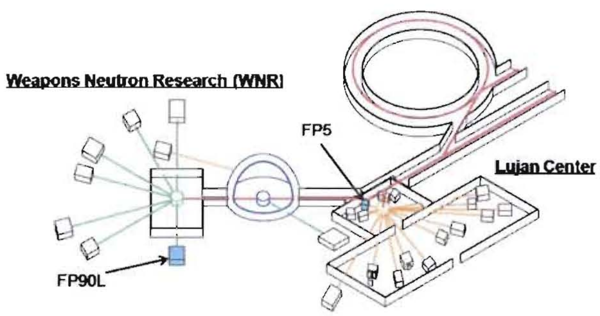

FIGURE 1. Drawing of the Lujan Center and WNR at LANSCE. The two flight paths used in this work are indicated with arrows.

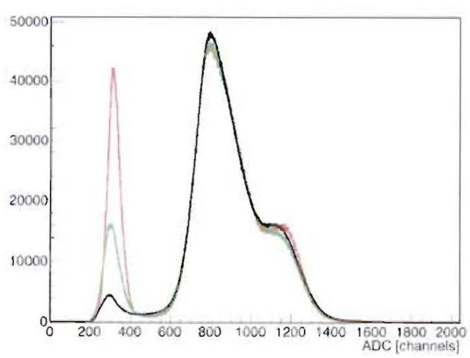

FIGURE 2. Pulse height spectra of signal from fission ionization chamber for fissile targets with different levels of α-activity.

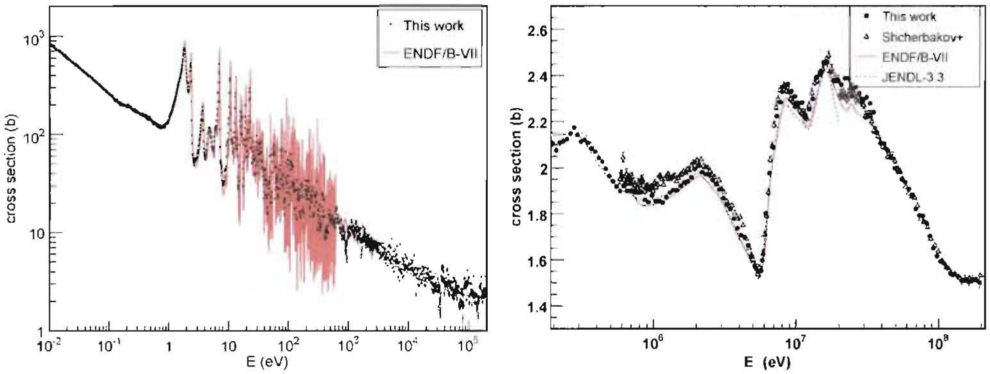

FIGURE 3. Measured fission cross section of ^{233}U from 10 meV to 200 keV (left plot) and from 200 keV to 200 MeV (right plot).

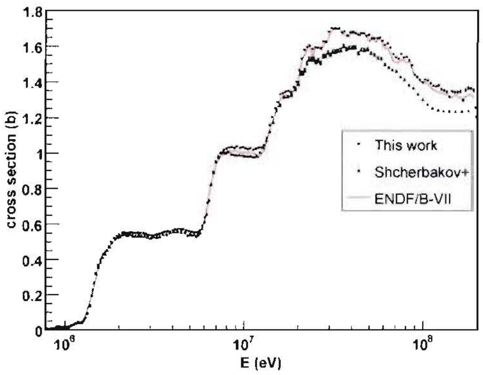

FIGURE 4. Measured fission cross section of ^{238}U from threshold to 200 MeV.

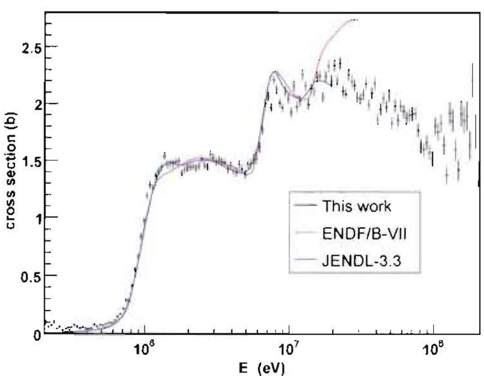

FIGURE 5. Preliminary result for fission cross section of ^{243}Am from 0.2 to 200 MeV.