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Fission Neutron Spectra Measurements at LANSCE – Status and Plans

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Abstract. A program to measure fission neutron spectra from neutron-induced fission of actinides is underway at the Los Alamos Neutron Science Center (LANSCE) in a collaboration among the CEA laboratory at Bruyères-le-Châtel, Lawrence Livermore National Laboratory and Los Alamos National Laboratory. The spallation source of fast neutrons at LANSCE is used to provide incident neutron energies from less than 1 MeV to 100 MeV or higher. The fission events take place in a gas-ionization fission chamber, and the time of flight from the neutron source to that chamber gives the energy of the incident neutron. Outgoing neutrons are detected by an array of organic liquid scintillator neutron detectors, and their energies are deduced from the time of flight from the fission chamber to the neutron detector. Measurements have been made of the fission neutrons from fission of ^{235}U , ^{238}U , ^{237}Np and ^{239}Pu . The range of outgoing energies measured so far is from 1 MeV to approximately 8 MeV. These partial spectra and average fission neutron energies are compared with evaluated data and with models of fission neutron emission. Results to date will be presented and a discussion of uncertainties will be given in this presentation. Future plans are to make significant improvements in the fission chambers, neutron detectors, signal processing, data acquisition and the experimental environment to provide high fidelity data including measurements of fission neutrons below 1 MeV and improvements in the data above 8 MeV.

Keywords: Neutron-induced fission, Fission neutrons, experiment, time of flight, LANSCE, white neutron source, ^{235}U , ^{238}U , ^{237}Np , ^{239}Pu , ^{232}Th

PACS: 25.85.Ec, 24.75.+i, 28.41.-i

INTRODUCTION

The spectrum of neutrons from neutron-induced fission is important for applications and for the understanding of the basic physics of a unique process. The spectrum is an essential ingredient in calculating nuclear neutron multiplication including nuclear criticality. Every reactor design and criticality safety calculation requires accurate data on the fission neutrons. Recently there has been much interest in

these data also for homeland security applications, because of the possibility of detecting fissile nuclear material by interrogation probes. If the objects to be detected multiply neutrons, then the number and spectrum of neutrons emitted can give a signature of illicit material.

Data on fission neutron spectra are recognized to be incomplete and often with large uncertainties. The present development of fission theory is not sufficient to provide accurate data for applications. A recent IAEA Consultants' Meeting summarized the status of experimental data for major actinides [1] and pointed out the need for more measurements. Precise measurements at selected incident neutron energies are needed as well as comprehensive measurements over a wide range of incident energies. The present experiments are designed to address the latter requirement.

The present evaluated data libraries describe the fission neutron spectrum as being similar to a Maxwellian distribution, $\sqrt{E} \exp(-E/T)$ with a temperature parameter that depends on the fissile nucleus as well as the energy of the incident neutron inducing the fission. The model of Madland and Nix [2] gives a more refined description, which is used, with some variations, in most of the evaluated libraries. For the ENDF/B-VII evaluation [3], the fission neutron spectrum relative to the spectrum induced by thermal neutrons is given in Fig. 1 where one sees that the spectrum generally becomes more energetic with increasing incident neutron energy. For incident energies above 6-7 MeV, the (n,n'f) channel known as "second-chance" fission opens and the spectrum softens with the addition of the pre-fission neutron emission. Well above this threshold, the spectrum hardens again.

One of the goals of the experiments described here has been to confirm this behavior as a function of incident energy. Previous work by Lovchikova [4] for selected neutron energies showed some of the features of the opening of the second-chance fission channel. Our more comprehensive experiments have covered a much wider range of incident neutron energies [5-8].

EXPERIMENT

The fission neutron spectrum was measured with a double time-of-flight experiment using the continuous-in-energy ("white") neutron source at the Weapons Neutron Research facility at the Los Alamos Neutron Science Center [9, 10]. The fission samples, used for many years at the CEA [11] are in fission ionization counters and neutrons were detected by the FIGARO array of neutron detectors [12].

At WNR, the bunched 800-MeV proton beam strikes a tungsten target and neutrons with a continuum of energies is produced from 0.1 MeV to several hundred MeV. This experiment was carried out on the 30-degree-right flight path at 22.74 meters from the source. The time of flight of neutrons from the source to the fission chamber identified their energies. The source pulses were separated by 1.8 microseconds with a macrostructure of 625 microseconds at a repetition rate of 40Hz. To reduce the number of low energy neutrons, attenuators of lead and polyethylene (CH₂) were placed in the beam. The neutron beam was collimated to 2.8 cm diameter by a series of CH₂, Cu, and Pb collimators.

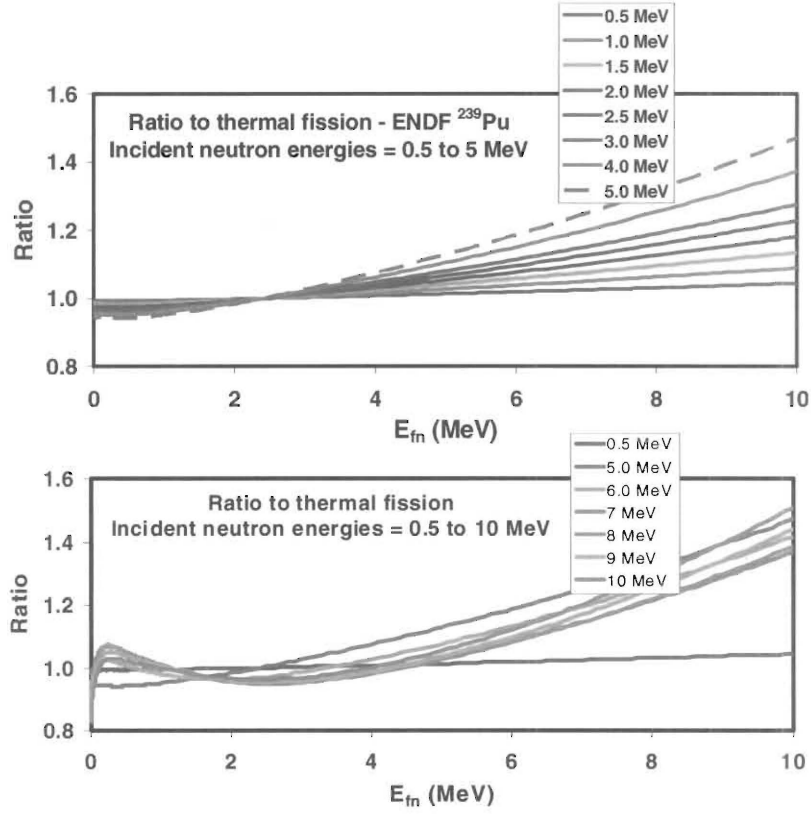


FIGURE 1. ENDF/B-VII evaluated data of the ratio of the fission neutron spectrum for various neutron energies relative to that at thermal. Top panel is for incident neutron energies up to 5 MeV, namely for the region of “first-chance” fission whereas the bottom panel is for higher energies that cover the transition to “second chance” fission in the neighborhood of 6-7 MeV.

Neutrons in this beam induced fission in a multiplate fission chambers [Ref .xx]. Thin layers of actinide materials were deposited on 0.125 mm thick platinum backings. The fission chamber electrodes were connected in parallel for the foils containing a given actinide so that one preamplifier could be used for full set of foils for each isotope.

The FIGARO array of 20 neutron detectors was used to detect the fission neutrons. These detectors are liquid EJ301 scintillators, 12.5 cm in diameter and 5 cm thick, with each being viewed by a Hamamatsu R1250A 5-inch photomultiplier tube. We used pulse-shape discrimination (PSD) techniques to distinguish the detection of neutrons from gamma rays. The array is pictured in Fig. 2 together with the fission chamber.

Standard electronics were used to process the electronic signals. An event was triggered by a pulse in the fission chamber. Pulse height and time relative to the beam pulse were recorded for the fission chamber and for the neutron detectors. For pulse-shape discrimination, two time gates were set on the neutron detector signals, one to integrate over the short component and the other for the full signal. Typical two-dimensional spectra to show the discrimination between neutron and gamma rays are shown in Fig. 3 where the threshold for clean separation can be made for proton recoil energies (minimum neutron energy) of slightly less than 1 MeV.

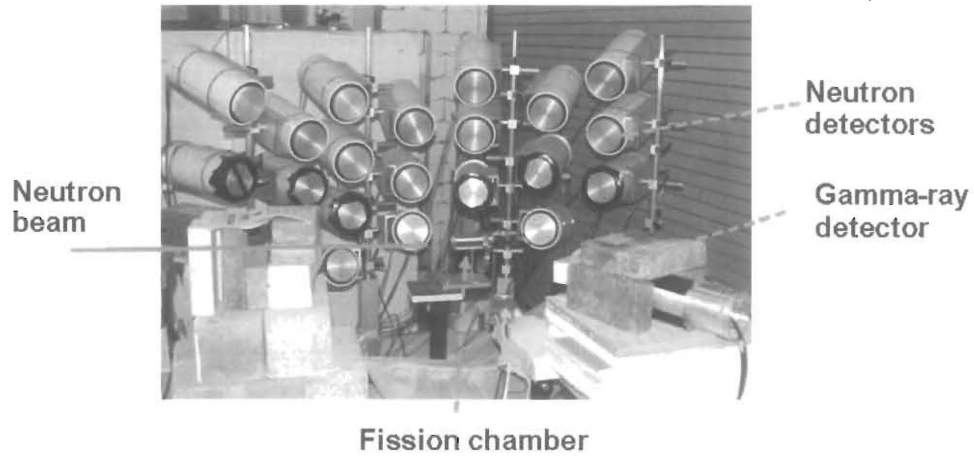


FIGURE 2. Experimental setup showing one of the fission chambers and the FIGARO array of neutron detectors. Not discussed in this report are the fission gamma rays detected by a variety of gamma-ray detectors.

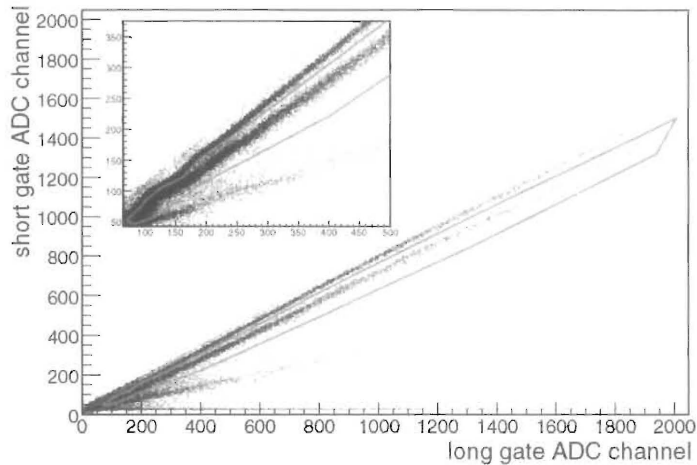


FIGURE 3. Pulse-shape discrimination achieved with the EJ301 liquid scintillators displayed as the short versus long gates of the integrates photomultiplier signal. The lower group of events is for proton recoils from neutron-proton scattering.

The efficiency of the neutron detectors was determined by a combination of measurements with a ^{252}Cf spontaneous fission neutron source and a computer simulation code, SCINFUL [13] to extend the efficiency curve to higher neutron energies. The ^{252}Cf source was placed at the position of the fission chamber and fission events were detected by a fast gamma-ray detector placed typically 15 cm from the source. A 4 cm-thick piece of polyethylene (CH_2) helped reduce the number of neutrons incident on this gamma-ray detector. Time-of-flight data were then taken between the gamma-ray trigger and events in the liquid scintillators. By comparing the energy spectra of the detected neutrons with the standard spectrum of neutrons from

the ^{252}Cf source, the efficiency of the neutron detector was obtained. The statistical accuracy of the resulting efficiency curve was not sufficient for a good determination above about 6 MeV. We therefore used the SCINFUL calculation, matched to the detector threshold, to smooth the efficiency curve and to extend it to neutron energies of 12 MeV. The results for one of the detectors are shown in Fig. 4.

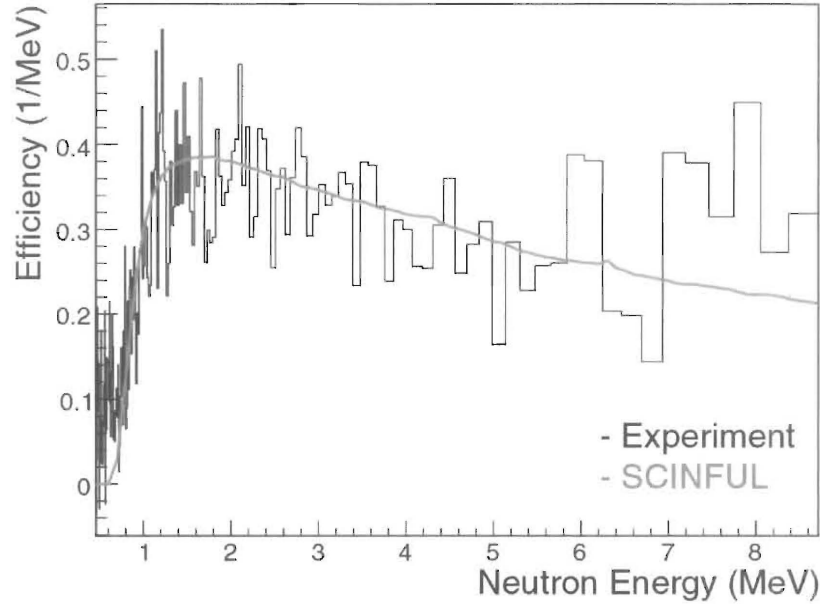


FIGURE 4. Measured and calculated efficiency of one of the liquid scintillators. The measured values come from data taken with a ^{252}Cf spontaneous fission neutron source, which is a standard. The calculated values are from the SCINFUL code [Ref. xx] with a detector threshold as determined with the measure data.

RESULTS

The results from this work have been published in the refereed literature and in a Los Alamos report [5-8]. Average fission neutron energies have been extracted from the data and show the general trends of slowly rising average energies up to the (n,n')f threshold where the average energy decreases in the next MeV or so before resuming the slow increase again.

In our more recent studies, we documented the experiment, reported the measured fission neutron spectra, and compared them with the ENDF/B-VII.0 evaluation [3]. Comparisons are shown in Fig. 5, for fission of ^{235}U and for ^{239}Pu induced by neutrons from 2 to 3 MeV from the white source. The data have been normalized to show the shapes of the fission neutron spectra. From these data, one can conclude that there is no significant deviation from the ENDF/B-VII.0 evaluation over this energy range of fission neutrons. Furthermore, the ratios of the shapes of the fission neutron spectra for these two isotopes, data for which were taken simultaneously, also are in agreement with the evaluations. For considerably higher incident neutron energies, there is some disagreement between experiment and evaluation for the higher end of the neutron

spectrum, where one expects contributions from pre-equilibrium emission of neutrons before fission [Fig. 6]

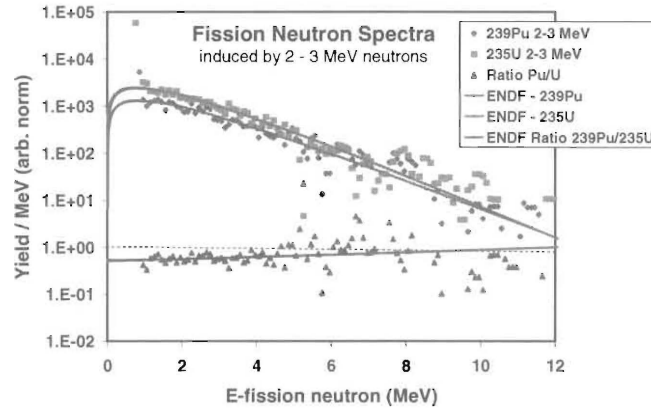


FIGURE 5. Measured fission neutron spectra for the 2-3 MeV bin of incident neutron energies. Results for ^{239}Pu are denoted by the black points and those for ^{235}U by the purple points. The ratios are given by the green triangles. The data are compared with the ENDF/B-VII.0 evaluations. Both the data and the evaluations have been arbitrarily normalized. The dashed horizontal line is a reference to show what a ratio of 1 would be in the relative fission neutron spectral shapes.

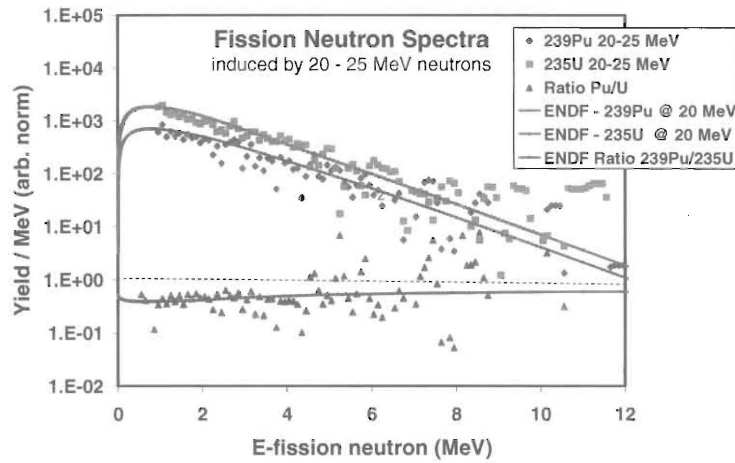


FIGURE 6. Same as Fig. 5 but for a higher incident neutron energy range of 20 – 25 MeV to show the component of pre-equilibrium leads to more neutrons in the range above 8 MeV.

FUTURE PLANS

Our program to improve the knowledge of the fission neutron spectrum is continuing. The results to date have been limited at both the low end of the spectrum, below 1 MeV, and at the high end of the spectrum, above approximately 8 MeV. At the low end, the present measurement technique is limited by pulse-shape discrimination (see Fig. 3), which is a fundamental limitation of liquid scintillators and other scintillators such as stilbene. While some improvement can be made to lower the threshold for good PSD, our goal is to measure the spectrum down to 100 keV, an energy where PSD has not been achieved, to our knowledge. We are therefore investigating other approaches to neutron detection at these lower energies.

For the higher end of the fission spectrum, above 8 MeV, our data are limited by statistics and by background. As shown in Figs. 5 and 6, the number of neutrons per MeV drops off by two orders of magnitude or more from its maximum value around 1 MeV. The solution to the counting statistics is simply to increase the solid-angle coverage of the neutron detector array. At present, we subtend about 2% of 4- π solid angle and this coverage can be increased significantly with more or larger detectors. With more detectors, it will also be possible in principle to investigate correlations of neutrons when two or more are emitted in the fission process. There should be a correlation both in angle and in energy according to the present view that most of the neutrons are emitted from fully accelerated fragments and therefore have the kinematic boost to their velocity in the center of mass of the fragment. .

To reduce the background, improvements are being made for the fission chamber. A parallel-plate avalanche counter (PPAC) is being developed for much improved timing. This counter will also have much thinned backing foils for the actinide deposits so that the gamma-ray production by beam neutrons on the foils will be significantly decreased.

SUMMARY

Neutron emission spectra from fission induced by neutrons from below 1 MeV to 200 MeV are being studied at LANSCE. The neutron emission energy range measured is 1 MeV to about 8 MeV. Average neutron energies have been reported for $^{235,238}\text{U}$ and ^{237}Np . Results of the spectra have been obtained for ^{235}U and ^{239}Pu . This program is continuing with the goal of extending the measured spectra to 100 keV on the low energy side and up to 12 MeV on the high energy side.

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