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NEUTRON INDUCED FISSION CROSS SECTION RATIOS FOR ^{232}Th, ^{235,238}U, ^{237}Np AND ^{239}Pu FROM 1 TO 400 MeV

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Abstract: Time-of-flight measurements of neutron induced fission cross section ratios for ^{232}Th, ^{235,238}U, ^{237}Np, and ^{239}Pu, were performed using the WNR high intensity spallation neutron source located at Los Alamos National Laboratory. A multiple-plate gas ionization chamber located at a 20-m flight path was used to simultaneously measure the fission rate for all samples over the energy range from 1 to 400 MeV.

Because the measurements were made with nearly identical neutron fluxes, we were able to cancel many systematic uncertainties present in previous measurements. This allows us to resolve discrepancies among different data sets. In addition, these are the first neutron-induced fission cross section values for most of the nuclei at energies above 30 MeV.

Introduction

The need for accurate fast-neutron-induced fission cross section data for long-lived actinides has long been recognized. Although measurements on many nuclei date back more than 30 years[1], significant inconsistencies exist and are now being uncovered. As an example, a recent measurement[2] of the ratio ^{239}Pu(n,f)/^{235}U(n,f) shows an 8% difference from the ENDF/B-V evaluation at 14.7 MeV. The origin of discrepancies in neutron-induced fission cross section measurements is at least partly due to the fact that suitable neutron sources have not existed, especially in the MeV energy range. Many measurements made with monoenergetic or quasi-monoenergetic sources have had systematic errors as a function of energy because different source reactions had to be used to cover a broad energy range. The development of 'white' neutron sources driven by electron accelerators and their subsequent application to fission cross section measurements[3] represented a significant advance; but, these sources have adequate intensity for fission measurements only below about 30 MeV. In addition, most measurements have been made with only a few different isotopes simultaneously, therefore increasing the possibility of cross-normalization error.

The results reported in this paper are preliminary values of cross section ratios obtained using the Weapons Neutron Research (WNR) high-intensity spallation neutron source at Los Alamos. These data will be supplemented by new results for the ^{235}U(n,f) cross section from 3 to 30 MeV, reported in another contribution to this conference[4] and by additional measurements[5] of Rapaport *et al.* up to 750 MeV.

Experimental Procedure

Los Alamos National Laboratory has recently commissioned a high-intensity white neutron source[6] which will be used for basic and applied research. The facility uses 800 MeV pulsed proton beam from the Los Alamos Meson Physics Facility (LAMPF) incident on a 7.5 cm long, 3 cm diameter tungsten target. This source differs significantly from that used in earlier measurements[7] because the neutron flux can be used at forward angles, providing as much as a three-fold increase in the neutron energy range. The results presented here were obtained using a 20 m flight path which viewed the neutron source at a production angle of 60°. The proton beam consisted of 250 ps wide pulses separated by ≈ 4 μs with 3×10^8 protons in each pulse. The macroscopic duty factor of LAMPF gave a rate of about 8000 of proton pulses/second. The neutron beam was transported in an evacuated flight tube and passed through a 2.54-cm thick polyethylene filter to reduce frame overlap; a permanent magnet to sweep out charged particles; and a system of three collimators as shown in Ref. 4, giving a beam diameter of 12.7 cm at the fission sample location. The technique used in both the neutron flux and the fission cross section ratio measurements is contained in Ref. 4, and will not be further described here. After passing through the fission chamber, the neutron beam passed through air to an annular proton recoil telescope (APT) which was used to provide a measurement of the neutron flux shape up to 30 MeV using the H(n,p) reaction, and then to a shielded beam dump 5-m downstream. A measurement of the neutron fluence obtained using the ^{235}U(n,f) yield rate and the fission cross section data of Ref. 5 is shown in Fig. 1, where the solid line is from an intra-nuclear cascade calculation. Above about 20 MeV, the calculated values substantially under-predict the data. These measurements agree with the trend of the fluence data up to 30 MeV obtained using the APT and described in our companion paper. The fission chamber held multiple foils of oxide material 10.2-cm in diameter vacuum evaporated onto 127-μm thick stainless steel backings. The fission foil deposits were typically 200 μg/cm^2 thick as determined by weighing samples produced during the vacuum evaporation process. We plan to verify uniformity at a later time by alpha counting. In addition to the fission foils, a ^{252}Cf deposit and a blank steel foil were included in the chamber. The ^{252}Cf was used to gain match pulse height spectra and for diagnostic purposes, and the blank foil was used to measure the background contribution from neutron-induced reactions. That background was negligible below 30 MeV and only about three percent at 400 MeV. For these results it was ignored. Flight paths from the neutron production target to the fission foil location were obtained using ^{12}C neutron transmission resonances.

Results and Conclusions

Preliminary values of our fission cross section ratios in 3% energy bins are shown in Fig. 1 for ^{232}Th, ^{237}Np, ^{238}U, and ^{239}Pu relative to ^{235}U. These data were obtained by computing a single normalization factor for each sample using the results of Meadows^{8} over the 1 to 10 MeV energy range. They are only a small part of the total data available for final analysis. The solid lines show the ENDF/B-V ratios, which have significant differences from our new values in the cases of ^{237}Np and ^{239}Pu. These data represent the only fission results above about 30 MeV for most of the nuclei, and are the most comprehensive ever obtained over the entire energy range under such nearly identical experimental conditions. Comparison of these data with similar results taken over a year earlier show agreement within the statistical uncertainties of about 2%. Over the energy range from 1 to 20 MeV, Fig. 3 shows fission cross sections obtained by multiplying our ratio data by the ENDF/B-V evaluation for the ^{235}U fission cross section. In general, ^{232}Th and ^{238}U show good agreement with ENDF/B-V whereas ^{237}Np and ^{239}Pu have significant discrepancies. Our results are not in agreement with the simple argument that fission cross sections are geometrical at high energies. Although the ratios of ^{237}Np and ^{239}Pu to ^{235}U are nearly unity as would be expected from such a picture, ^{232}Th and ^{238}U ratios approach 0.5 and 0.9 respectively. More sophisticated theoretical analyses now in progress at Los Alamos indicate that our higher energy data is difficult to reproduce with parameters normally used in traditional models.

During the summer of 1988, further measurements and data analysis are planned which should significantly reduce the statistical uncertainties from those presented here. New fission foils are now being fabricated for ^{233,234,236}U, and measurements using those samples are planned for later this year.

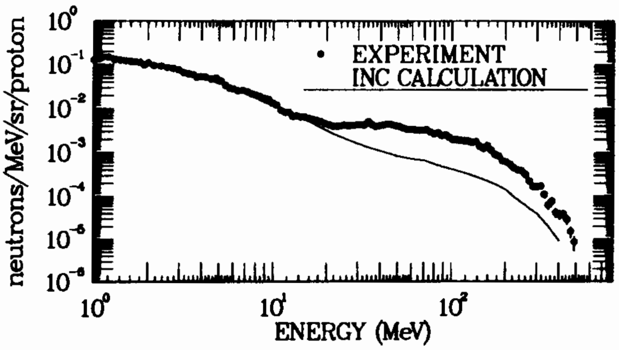

Fig. 1. Comparison of the present measurements of neutron fluence shape with Intra-Nuclear Cascade Model predictions.

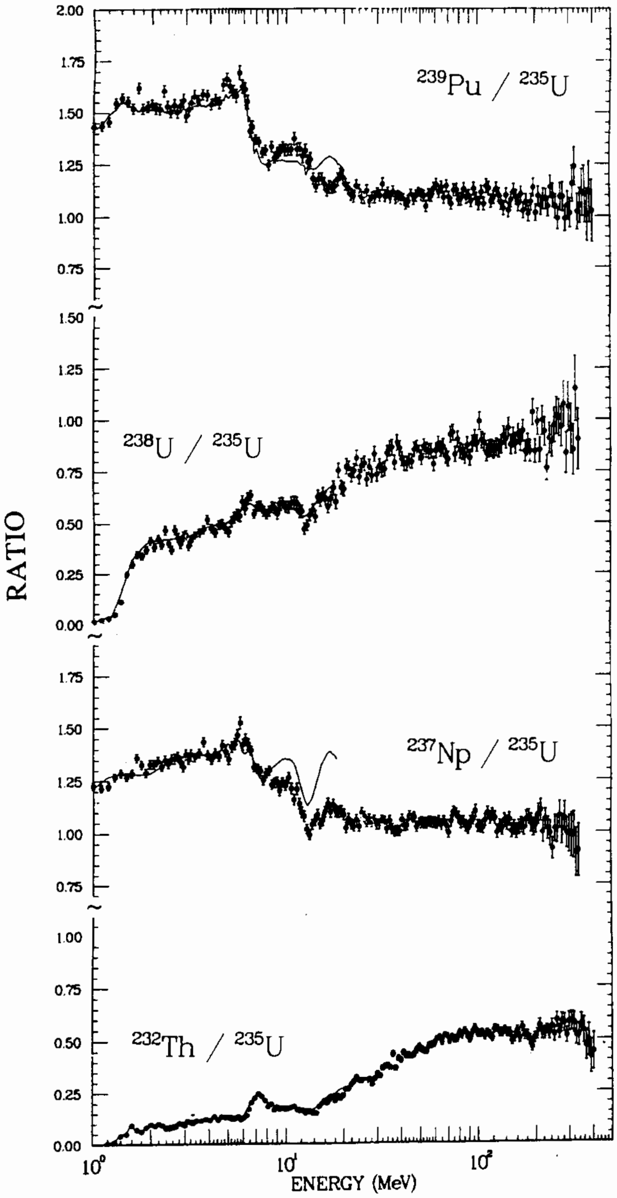

Fig. 2 Preliminary results of fission cross section ratios from 1 to 400 MeV. The solid line extending to 20 MeV is from ENDF/B-V.

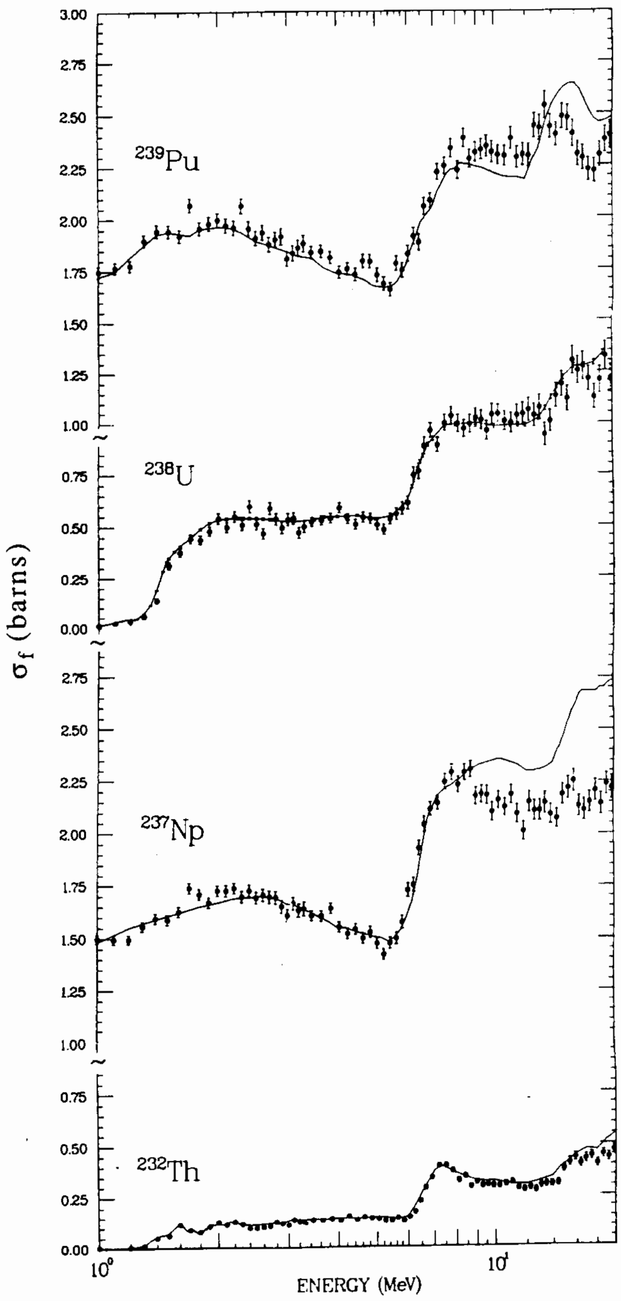

Fig. 3 Preliminary fission cross section results from the present measurement from 1 to 20 MeV. The solid line is from ENDF/B-V.

References

1. G. A. Jarvis, "Fission Comparison of ^{238}U and ^{235}U for 2.5 MeV Neutrons," LA-1571, Los Alamos Scientific Laboratory (1953).
2. J. W. Meadows, "The Fission Cross Sections of ^{230}Th, ^{232}Th, ^{233}U, ^{234}U, ^{236}U, ^{236}U, ^{237}Np, ^{239}Pu, and ^{242}Pu Relative ^{235}U at 14.74 MeV Neutron Energy," ANL/NDM-97, Argonne National Laboratory, (1986).
3. F. C. Difilippo, R. B. Perez, G. de Saussure, D. K. Olsen, and R. W. Ingle: Nucl. Sci. Eng. 68, 43 (1978).
4. A. D. Carlson, O. A. Wasson, P. W. Lisowski, J. L. Ullmann, and N. W. Hill: this conference.
5. J. Rapaport, J. Ullmann, R. O. Nelson, S. Seestrom-Morris, S. A. Wender, and R. C. Haight, "Preliminary Measurement of the ^{235}U(n,f) Cross Section up to 750 MeV," LA-11078-MS, Los Alamos National Laboratory (1987).
6. S. A. Wender, P. W. Lisowski, S. Seestrom-Morris, R. O. Nelson, J. L. Ullmann, T. Burritt, and J. Rapaport: Proc. of an International Conference on Neutron Physics, Kiev USSR, 1987 (to be published).
7. R. L. Schitt, R. E. Shamu, P. W. Lisowski, M. S. Moore, and G. L. Morgan: Phys. Lett. B203, 22 (1988).
8. J. W. Meadows, "The Fission Cross Sections of Some Thorium, Uranium, Neptunium and Plutonium Isotopes Relative to ^{235}U," Argonne National Laboratory, ANL/NDM-33, (1983).