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NEUTRON-EMISSION MEASUREMENTS AT A WHITE NEUTRON SOURCE

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Data on the spectrum of neutrons emitted from neutron-induced reactions are important in basic nuclear physics and in applications. Our program studies neutron emission from inelastic scattering as well as fission neutron spectra. A "white" neutron source (continuous in energy) allows measurements over a wide range of neutron energies all in one experiment. We use the fast neutron source at the Los Alamos Neutron Science Center for incident neutron energies from 0.5 MeV to 200 MeV. These experiments are based on double time-of-flight techniques to determine the energies of the incident and emitted neutrons. For the fission neutron measurements, parallel-plate ionization or avalanche detectors identify fission in actinide samples and give the required fast timing pulse. For inelastic scattering, gamma-ray detectors provide the timing and energy spectroscopy. A large neutron-detector array detects the emitted neutrons. Time-of-flight techniques are used to measure the energies of both the incident and emitted neutrons. Design considerations for the array include neutron-gamma discrimination, neutron energy resolution, angular coverage, segmentation, detector efficiency calibration and data acquisition. We have made preliminary measurements of the fission neutron spectra from ^{235}U , ^{238}U , ^{237}Np and ^{239}Pu . Neutron emission spectra from inelastic scattering on iron and nickel have also been investigated. The results obtained will be compared with evaluated data.

KEYWORDS : Neutrons, Fission, Fission Neutron Spectra, Inelastic Scattering, Spallation Neutron Source

1. INTRODUCTION

Accurate data on neutron emission from fast-neutron-induced fission and other fast-neutron-induced reactions are crucial for many applications including the development of fast reactors, criticality safety, shielding at reactor and accelerators, medical therapy, and radiation effects on electronics. The data are also of great interest in advancing nuclear reaction theory, and activity in new models of neutron-induced fission [1–5] and in inelastic scattering is very active at present.

For fission induced by fast neutrons, the data base is far from adequate for applications or for testing nuclear reaction models. For the important isotope, ^{239}Pu , for example, there are only two data sets for the fission neutron spectra that are useful and these data are in some sense inconsistent. Staples *et al.* [6] measured the part of the fission neutron spectra above the incident neutron energy for incident energies of 0.5, 1.5, 2.5 and 3.5 MeV. Knitter *et al.* [7], did the same for one incident neutron energy, 0.215 MeV. The emission spectra resemble evaporation spectra, with one example being given in Fig. 1, where the data are compared with a Maxwellian form with a temperature parameter of 1.30 MeV and with the ENDF/B-VII evaluated spectrum. The figure points to two of the challenges in measuring these spectra. First, with a multi-gram sample

of ^{239}Pu , only that part of the fission neutron spectrum above the energy of the incident neutron can be measured. The reason is that, below the incident neutron energy, the emission spectrum is a combination of fission neutrons and inelastically scattered neutrons. The second challenge is that the evaporation spectrum decreases 4 orders of magnitude from emitted energies near 1 MeV to 10 MeV.

To view the data in more detail, we divide the measured data by the Maxwellian with the temperature of 1.30 MeV, and we do the same for the ENDF/B-VII.0 spectra. The results are shown in Figs. 2 and 3. These figures show the inconsistency in the measured data, since the fission neutron spectrum is expected to change slowly with the incident neutron energy. The ENDF/B-VII.0 evaluations seem to be a compromise between these two data sets. In fact there are no other useful data sets of fission neutron spectra from fission induced by fast neutrons on ^{239}Pu to constrain the Los Alamos model [8] that was used in the evaluations for ENDF/B-VII.0.

The data situation for some other actinides, such as ^{235}U and ^{238}U , is somewhat better, but for minor actinides, the situation is much worse. There is therefore a need for new measurements of the fission neutron spectra at many incident neutron energies on a wide range of fissionable nuclides to aid the development of improved theory and to provide data for evaluations used in applications.

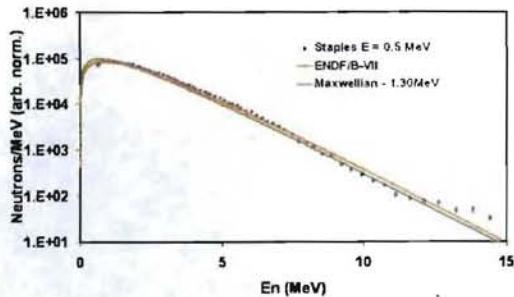


Fig. 1. Neutron-emission spectrum from fission of ^{239}Pu induced by neutrons of 0.5 MeV measured by Staples *et al.* [6]. The ENDF/B-VII.0 evaluation and a Maxwellian with $T=1.30\text{ MeV}$ are also shown.

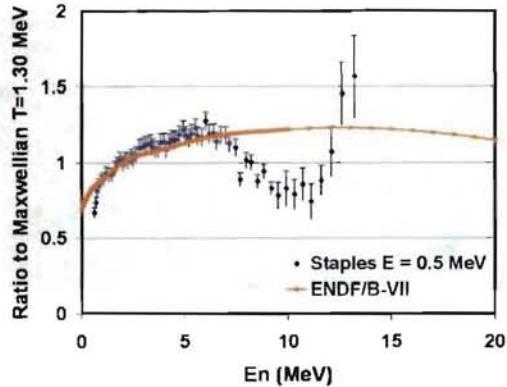


Fig. 2. Data for 1.5 MeV fission neutron emission data from Staples *et al.* cite[Staples95] and the ENDF/B-VII.0 evaluation, both divided by the Maxwellian with $T=1.30\text{ MeV}$.

At the Los Alamos Neutron Science Center (LANSCE), we have been investigating for several years, the neutron emission spectra from fission induced by neutrons from a spallation neutron source. This is a new approach as all of the other measurements have been made with monoenergetic neutron sources. In addition, we are able to measure fission neutrons emitted below as well as above the incident neutron energy. Our aim is to quantify, as well as possible, the full fission neutron spectra as a function of incident neutron energies of below 1 MeV to well above 20 MeV.

For neutron inelastic scattering, such as (n, n') reactions, our interests are in the continuum emission of neutrons. These emitted neutrons give information on the level density of excited states in the target nucleus, as their spectrum is determined by the neutron transmission probability and the phase space that the residual state can reach. The transmission probability can be calculated with some confidence from the optical model parameters, but the phase space, the level density of the residual nucleus, is not so well known. Nuclear level densities are a subject of much interest at present and are approached by a range of nuclear models

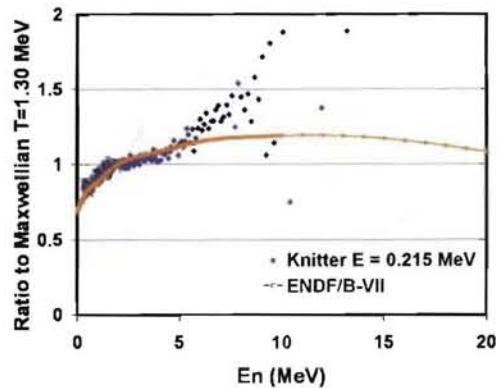


Fig. 3. Data for 1.5 MeV fission neutron emission data from Knitter *et al.* [7] and the ENDF/B-VII.0 evaluation, both divided by the Maxwellian with $T=1.30\text{ MeV}$.

including combinatorial methods, Monte Carlo shell model calculations, back-shifted Fermi gas models with superfluid corrections and so forth.

The data for inelastic scattering including neutron emission spectra are based mostly on experiments with monoenergetic neutron sources. Again the data base is sparse. For ^{56}Fe , for example, there are some excellent data for 14-MeV neutrons incident, e.g. [9], but even here measurements of the emission spectrum for emitted neutrons below 1 MeV is almost non-existent. For other incident neutron energies, there are some measurements, but they do not span a large range of incident energies continuously. Of course there are measurements of production of discrete gamma-rays [e.g. ORELA and LANSCE data] but they do not give information on how the residual states in the nucleus are reached, either by direct neutron emission or by emission of a lower energy neutron followed by a gamma-ray cascade.

2. EXPERIMENT

The approach we use is described in several publications. Briefly, we use the spallation source of fast neutrons at LANSCE [10, 11, 13]. The pulsed and bunched 800-MeV proton beam of the accelerator is directed to a tungsten target with a micropulse width of less than 1 ns and spacings typically of 1.8 microseconds. The continuous, spallation neutron spectrum is collimated to a beam diameter of approximately 3 cm and impinges on a target at the center of an array of neutron and gamma-ray detectors, called the FIGARO array [12]. The energy of the neutron incident on the sample is determined by time of flight from the source to the sample. A beam pickoff determines when the proton beam hits the tungsten target and the stop signal is provided by the fission chamber (either an ion chamber or a parallel-plate avalanche counter (PPAC) or a gamma-ray detector when the sample is not a fissionable isotope).

The energy of the emitted neutron is determined also by time of flight, but this flight path is between the sample and the neutron detector. An array of 20 neutron detectors has been used so far, and a larger array is being planned. The neutron detectors are either liquid organic scintillators (EJ301 from the Eljen Corp.) or ^6Li -glass detectors, available from several suppliers. The former have pulse-shape discrimination so that neutrons interacting in the detector can be separated from gamma rays. For our setup, this separation is good for neutron energies above about 700 keV. Lower-energy neutrons can be detected and identified as neutrons by the $^6\text{Li}(\text{n},\text{alpha})^3\text{H}$ reaction with its positive Q-value of 4.8 MeV.

To make measurements of high quality, details are essential. Backgrounds must be characterized and quantified. The efficiency of the neutron detectors must be known accurately, in our case by a combination of measurement and calculation. Multiple scattering corrections are required. Biases of many possible origins must be eliminated as much as possible. For example, one bias not considered by many researchers in emission spectra from neutron-induced fission is the correlation between the fission fragment distribution and the angular distribution of the emitted neutron. Because the fission-fragment angular distribution varies with the energy of the incident neutrons [14], a measurement of the fission neutrons spectrum can be biased if only a part of the fission fragment distribution is sampled.

A typical example of the time-of-flight spectrum for fission neutrons is shown in Fig. 4. Fission gamma rays are clearly seen and they serve as a time marker. When pulse-shape discrimination is added as a condition, the fission gamma rays nearly disappear from the time spectrum and leave events that are primarily neutrons. The figure also shows background neutrons, those that arrive before the fission gamma rays and those that arrive after neutrons that are at the threshold of detection. The shape of the time spectrum of these background neutrons is determined by substituting a random pulse for the fission chamber pulse, and so we call these “fake fissions”. One goal of our future work is to reduce the background neutrons as much as possible.

3. RESULTS

A few sample results are presented in this section. Extensive measurements have been made of the fission neutron spectra from neutron induced fission of ^{235}U , ^{238}U , ^{237}Np , and ^{239}Pu [18, 19, 15, 17]. Examples of the neutron emission from ^{235}U and ^{239}Pu for a few bins of incoming neutron energies are shown in Fig. 5. The ratios of the shapes of these spectra are shown and they suggest that the fission neutron spectra from these two actinides are quite similar in shape. In the ratio data, several systematic uncertainties cancel and so these data should be quite reliable.

For emission spectra from neutron inelastic scattering, we have at present only a few final results. Shown in Fig. 6 are the emission spectra from inelastic scattering on natural

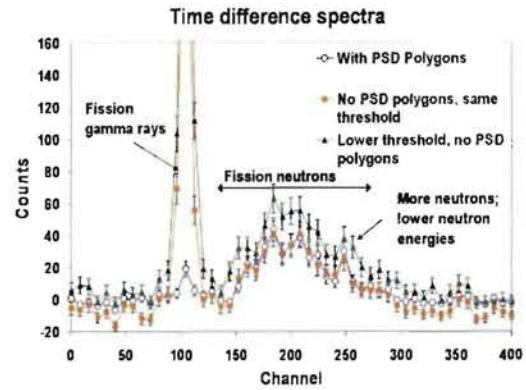


Fig. 4. Time difference spectrum which shows the time of flight from the fission chamber to a neutron detector. Spectra for two different thresholds are shown, with no pulse shape discrimination. Adding PSD removes most of the gamma rays.

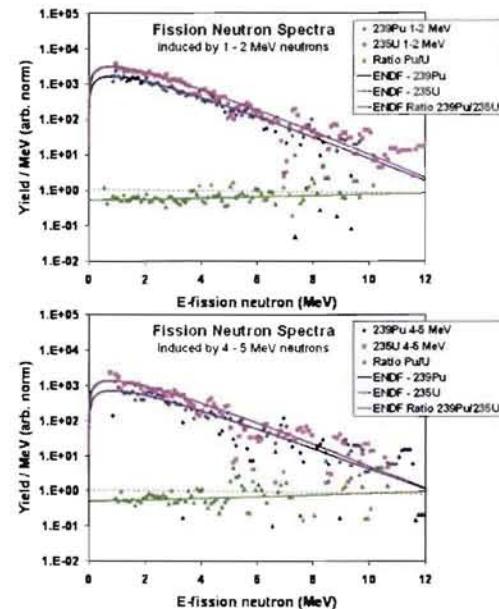


Fig. 5. Neutron emission spectra from fission of ^{239}Pu induced by neutrons of 1-2 MeV (top panel) and 4-5 MeV (bottom panel).

nickel. Calculations were made with the EMPIRE nuclear reaction model code for the two principal isotopes, ^{58}Ni and ^{60}Ni and the results added according the isotopic abundances. An optimization of the calculations was done by modifying the nuclear level density. The results are shown in the figure and they agree nicely with the experimental data.

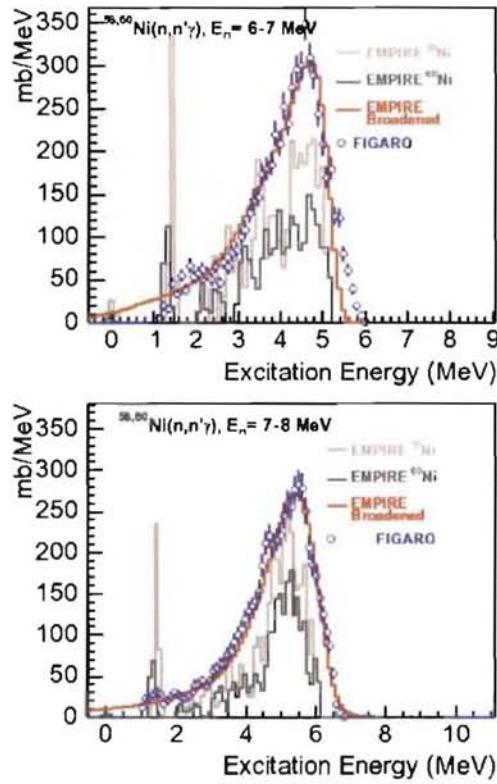


Fig. 6. Neutron emission spectra from inelastic scattering on natural nickel for two different incident energies. The spectra are gated on the first 2^+ -to-ground state transition. The calculations are for all inelastic scattering and combine results for the two major nickel isotopes.

4. DISCUSSION

Average fission neutron energies can be calculated from the spectra where the unmeasured part of the distributions, namely for $E_{fn} \leq 1$ MeV, is approximated by a functional form fit to the measured part of the spectrum. We find that the average fission neutron energy for fission of ^{235}U and ^{239}Pu does not change much as a function of the energy of the incident neutron in the range 1-8 MeV [17-19]. For ^{238}U , however, there is a marked decrease in the average fission neutron energy as the second chance fission ($n, n'\gamma$) threshold is crossed [19].

Fission neutron spectra are calculated in the Los Alamos model with very few parameters, one of which is the total kinetic energy of the fission fragments. This parameter can be adjusted to fit our data, and the results show that the values giving the best fit are very similar to those determined by measuring the fission fragments directly [17]. For neutron inelastic scattering, a very recent development of a Monte Carlo Hauser Feshbach code [16] will allow comparison of the model calculation with our data. In the example given in Fig. 6, the neutron emission spectrum was calculated without regard to the subsequent gamma-ray cascade. In our

experiment, however, we gated the events on a particular transition, namely the first 2^+ -to-ground state transition, which we could identify by the energy of the emitted gamma ray measured with the gamma-ray trigger detector. The new code will allow selection of specific gamma rays in the de-excitation cascade in the residual nucleus (see Fig. 7). Some sensitivity to the spin dependence of the nuclear level density should emerge from these calculations as they fit the corresponding experimental data.

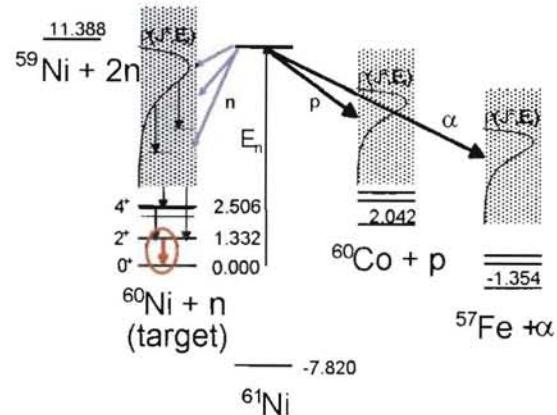


Fig. 7. Schematic level diagram showing how the code of Kawano *et al.* [16] will be used to gate on specific gamma rays in the residual nucleus.

5. FUTURE WORK

The FIGARO array is being renamed “Chi-Nu” as it will be used heavily for measuring the fission neutron spectra from neutron-induced fission. The Chi-matrix relates the incident neutron energy to the distribution of fission neutrons in energy. This neutron detector array will be enlarged in the number of detectors and it will include ^6Li -glass detectors for emitted neutrons below 1 MeV. Significant changes in the data acquisition are planned. It has been shown by several researchers that the use of waveform (flash) digitizers opens up the possibility of extracting more information from the pulse shape of the signals from organic scintillators. For example, several different time intervals can be used to characterize the pulse shape, and these intervals can be changed depending on the pulse height to optimize the neutron-gamma-ray discrimination. We plan to use such techniques in the near future.

Finally, the Monte Carlo Hauser Feshbach code has only very recently been written, and it will be thoroughly exercised to find where the greatest sensitivity exists to exclusive measurements triggered on specific gamma rays. We expect sensitivity to the angular momentum distribution of the nuclear level density and also to specific nuclear structure.

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REFERENCES

- [1] H. Goutte, J.-P. Delaroche, M. Girod, J. and Libert, "Microscopic description of fission properties," AIP Conf. Proc. **891**, 324 (2007).
- [2] W. Younes, D. Gogny, and J. Esher, "Microscopic theory of fission," AIP Conf. Proc. **1005** 194 (2008).
- [3] J. Randrup and R. Vogt, "Calculation of Fission Observables through Event-by-Event Simulation," Phys. Rev. C **80**, 024601 (2009).
- [4] S. Lemaire, P. Talou, T. Kawano, M. B. Chadwick, and D. G. Madland, "Monte Carlo Approach to Sequential Neutron Emission from fission fragments," Phys. Rev. C **72**, 024601 (2005).
- [5] P. Talou, T. Kawano, L. Bonneau, and J. Escher, "Prompt fission neutrons as probes to nuclear configurations at scission," AIP Conf. Proc. **1005** 198 (2008).
- [6] P. Staples, J. J. Egan, G. H. R. Kegel, A. Mittler, and M. L. Woodring, "Prompt Fission Neutron Energy Spectra Induced by Fast Neutrons," Nucl. Phys. **A591**, 41 (1995).
- [7] H. -H. Knitter, "Measurement of the Energy Spectrum of Prompt Neutrons from the Fission of Pu239 by 0.215 MeV Neutrons", Atomkernnergie **26**, 76 (1975).
- [8] D. G. Madland and J. R. Nix, "New Calculation of Prompt Fission Neutron Spectra and Average Neutron Multiplicities," Nucl. Sci. Eng. **81**, 213 (1982).
- [9] A. Takahashi, E. Ichimura, Y. Sasaki, *et al.*, Journal of Nuclear Science and Technology **25**(3), 215 (1988).
- [10] P. W. Lisowski, C. D. Bowman, G. J. Russell, and S. A. Wender, "The Los Alamos National Laboratory Spallation Neutron Sources," Nucl. Sci. Eng. **106**, 208 (1990).
- [11] P. W. Lisowski and K. F. Schoenberg, "The Los Alamos Neutron Science Center," Nucl. Instr. Methods A **562**, 910 (2006).
- [12] D. Rochman, R. C. Haight, J. M. O'Donnell, M. Devlin, T. Ethvignot, and T. Granier, "Neutron-induced Reaction Studies at FIGARO using a Spallation Neutron Source," Nucl. Instr. Methods A **523**, 102 (2004).
- [13] R. O. Nelson, "LANSCE Nuclear Science Facilities and Activities," (this conference).
- [14] J. E. Simmons and R. L. Henkel, "Angular Distribution of Fragments in Fission Induced by MeV Neutrons," Phys. Rev. **120**, 198 (1960).
- [15] R.C. Haight, S. Noda and J.M. O'Donnell, "Los Alamos Analysis of an Experiment to Measure Fission Neutron Output Spectra from Neutron-Induced Fission of ^{235}U and ^{239}Pu ," LANSCE-NS Progress Report, LA-UR-08-2585, (2008).
- [16] T. Kawano, P. Talou, M. B. Chadwick and T. Watanabe, "Monte Carlo Simulation for Particle and Gamma-Ray Emissions in Statistical Hauser-Feshbach Model," J. Nucl. Sci. Tech. (in press).
- [17] S. Noda, R. C. Haight, R. O. Nelson, M. Devlin, J. M. O'Donnell, A. Chatillon, T. Granier, G. Bélier, J. Taieb, T. Kawano and P. Talou, "Prompt fission neutron spectra from fission induced by 1 to 8 MeV neutrons on ^{235}U and ^{239}Pu using the double time-of-flight technique," Phys. Rev. C (submitted) (2010).
- [18] T. Ethvignot, M. Devlin, R. Drosd, T. Granier, R. C. Haight, B. Morillon, R. O. Nelson, J. M. O'Donnell, and D. Rochman, Phys. Lett. B **575**, 221 (2003).
- [19] T. Ethvignot, M. Devlin, T. Granier, R. C. Haight, B. Morillon, R. O. Nelson, J. M. O'Donnell, and D. Rochman, Phys. Rev. Lett. **94**, 052701 (2005).