Title: Nuclear Data Uncertainties in Intermediate Neutron Spectrum Problems and the Intermediate Neutron Spectrum Experiment

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Nuclear Data Uncertainties in Intermediate Neutron Spectrum Problems and the Intermediate Neutron Spectrum Experiment

by

P. J. Jaegers, D. Hayes, R. G. Sanchez, R. H. Kimplaud

Introduction

During the investigation of various surplus fissile material disposition proposals, uncertainties in the data of SiO₂/Pu mixtures were discovered. These uncertainties were detected for Si/Pu ratios ranging from approximately 20:1 to 200:1. Additionally, it has been reported that metal/U²³⁵ mixtures also have data uncertainties (Ref. 1 and 2). These problems with large data uncertainties are typically intermediate spectrum problems, i.e. problems which are dominated by scattering and fissions occurring in the energy range of 10 eV to 1 MeV.

ANSI/ANS-8.1-1983 states that the "bias shall be established by correlating the results of criticality experiments with the results obtained for these same systems by the method being validated". There is however, a definite lack of critical experiments with an intermediate neutron spectrum. We have identified two potential areas from which these uncertainties may arise. The first potential source of uncertainty in the nuclear data is due to fact that the cross-sections of the fissile material have not been tested in an intermediate energy spectrum. The second potential source of uncertainty is that the non-fissile material in the mixture may not have been appropriately tested in a critical experiment. To provide this benchmark data a critical experiment is necessary.

Plutonium/Silicon-Dioxide Calculations

In the course of calculating the critical mass of plutonium mixed with silicon-dioxide, it was noticed that the various nuclear data sets at our disposal predicted nearly identical critical masses for both fast and thermal systems. It was notice however, that for SiO₂/Pu ratios ranging from 20:1 to 200:1, significant disagreements in k_{eff} (and correspondingly large disagreements in the critical masses) were predicted. In addition smaller disagreements in the predicted k_{eff} values persisted over an even wider range in SiO₂/Pu ratios. Upon further examination, it was determined that the neutron energy spectrum was intermediate for this portion of the critical mass curve.
More detailed calculations were performed using DANTSYS (ref. 3), MCNP (ref. 4), and SCALE (ref. 5) on a simple silicon-dioxide/plutonium-239 system. The system was composed of 100 kg of plutonium-239 uniformly loaded into a sphere of SiO₂. The radius of this SiO₂/Pu sphere was then varied from 40 cm to 100 cm in order to vary the SiO₂/Pu ratio from 23.5:1 to 366.6:1. This sphere was then reflected with a 100 cm thick SiO₂ reflector. The SiO₂ used throughout the problem had a density of 2.2 g/cc.

The cross-section sets used consisted of: 1) Hansen-Roach (ref. 6) and MENDF5 (ref. 7) for DANTSYS, 2) ENDF-V continuous and multigroup for MCNP, 3) and SCALE 27 group for SCALE. Figure 1 shows the k_{eff} of the system as a function of the plutonium/silicon-dioxide ball radius. As one can see, there is a spread of approximately 7.5 percent in the predicted k_{eff} of the system. This spread in k_{eff} roughly corresponds to a 30 kg change in the plutonium mass and represents a 30 percent change in the critical mass. If one were to just compare the predictions obtained using the SCALE 27-group and MCNP continuous data the difference is approximately 3.3 percent in k_{eff}.

Figure 2 shows the neutron production spectrum, which is a normalized plot of νΣ_{eff} for a Si/Pu ratio of 65:1. As one can see, nearly 55 percent of all fissions are caused by neutrons between 10 eV and 1 MeV, and thus is an intermediate energy spectrum problem.
Figure 1: Predicted $k_{\text{eff}}$ as a Function of SiO$_2$/Pu Core Radius
Figure 2 Normalized Neutron Production Spectrum for SiO$_2$/Pu Ratio of 65:1

**Fe/Uranium Calculations**

Similar calculations to the ones described above were performed for a system that consisted of homogeneous iron/uranium mixtures in a spherical geometry containing approximately 170 kg of 93% enriched uranium and different amounts of iron. The density of the iron was assumed to be 7.86 g/cc. In all cases the Fe/U spheres were reflected by a 100 cm iron reflector. Again, the radius of the Fe/U sphere was varied between 20 and 100 cm keeping the amount of uranium constant as well as the reflector thickness. All the calculations were performed using DANTSYS, and MCNP and different sets of cross section data such as: 1) Hansen-Roach and MENDF5 for DANTSYS, and 2) ENDF-V and ENDF-VI
continuous, and multi-group for MCNP. The results obtained using the variety of cross section sets and codes are shown in Fig. 3. It is important to note that there is a maximum spread of approximately 50% in the predicted k-eff between MENDF5 and ENDF-V continuous energy cross section data as seen in Fig. 3. A smaller uncertainty (approximately 25% in the predicted k-eff), but also significant, was obtained in the fast neutron energy spectrum, which corresponds to an Fe/U-235 ratio ranging from 7 to 100. Based on these results, it is certain that there are large uncertainties in the predicted k-eff for other systems containing mixtures of Fe/Pu-239 and Fe/U-233, thus the need for critical experiments.

Figure 3 Predicted k\textsubscript{eff} as a Function of Fe/U Core Radius

The Experiment

To provide the necessary benchmark data to resolve such discrepancies as discussed for the SiO\textsubscript{2}/Pu mixtures and the metal/U\textsuperscript{235} mixtures of references 1 and 2, we are currently constructing at the Los Alamos Critical Experiments Facility (LACEF) a benchmark experiment to determine the critical mass of various fissile/non-fissile material mixtures which can be tuned to produce a neutron energy spectrum that will range from fast through intermediate to thermal.
The assembly will initially consist of a core containing a number of 93% enriched uranium plates, 26.67 cm in radius and 3 mm thick, stacked with an interstitial non-fissile material plates and surrounded by a reflector. Several reflector materials are to be used, of which the first sets will consist of copper, iron, and graphite. Candidate non-fissile interstitial materials include copper, iron, graphite, and silicon-dioxide to name a few. Upon completion of the uranium-235 experiments subsequent experiments will be conducted using plutonium and uranium-233 as the fuel.

The neutron spectrum is tuned via several methods. Both copper and iron are predicted to be good fast reflectors, and hence suitable for the fast and intermediate spectrum experiments. On the other hand graphite is a good thermal reflector, and is to be used for the thermal experiments. Thus, some spectral control is obtained from the choice of reflector. Another method that may be used to readily control the neutron spectrum is to vary the thickness of the non-fissile interstitial materials to be tested. This method is predicted to work well for the lower Z materials such as graphite and SiO2, while for the moderate Z materials such as iron and copper, it may be necessary to introduce some hydrogen into the system in order to produce an intermediate spectrum and still achieve a critical state. A nominal iron/hydrogen ratio is expected to be 20:1 in these experiments.

References

1. C. V. Parks, W. C. Jordan, L. M. Petrie, R. Q. Wright Use of Metal/Uranium Mixtures to Explore Data Uncertainties

2. k\textsubscript{\infty} for Certain Metals Mixed with 235U, Crit. Safety Q. (Winter 1993)


Los Alamos National Laboratory only 30 group cross-section set derived from END-F-V/B