NUCLEAR DATA NEEDS FOR APPLICATION IN NUCLEAR CRITICALITY SAFETY PROGRAMS

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INTRODUCTION

In nuclear criticality safety applications, a number of important uncertainties have to be addressed to establish the required criticality safety margin of a nuclear system. One source of these uncertainties is the basic nuclear data used to calculate the effective multiplication factor of the system. Before criticality safety calculations are performed, the bias and uncertainties of the codes and cross sections that are used must be determined. The most common sources of uncertainties, in general, are the calculational methodologies and the uncertainties related to the microscopic cross sections used in the calculational procedure.

Cross-section data are measured, evaluated, and tested prior to their inclusion in nuclear data libraries. Traditionally, nuclear data evaluations are performed to support the analysis and design of thermal and fast reactors. The neutron spectra characteristic of the thermal and fast systems used for data testing are predominantly in the low- and high-energy ranges, with a relatively minor influence from the intermediate-energy range. In the area of nuclear criticality safety, nuclear systems involving spent fuel elements from reactors can lead to situations very different from those most commonly found in reactor analysis and design. These systems are not limited to thermal or fast neutron spectra and may have their most significant influence from the intermediate-energy range. This requires extending the range of applicability of the nuclear data evaluation beyond thermal and fast systems. The aim here is to focus on the evaluated nuclear data pertaining to applications in nuclear criticality safety.

CROSS-SECTION REPRESENTATION IN THE BASIC NUCLEAR DATA FILES

The energy range in which the spacing between two consecutive resonance peaks is greater than the width of the resonance is referred to as the resolved energy region. Without loss of generality, it can be said that the resolved energy range for nuclei of mass numbers \( A > 80 \), such as \(^{235}\text{U}, {238}\text{U}, {239}\text{Pu} \), the resonance structures begin at energies as low as a few eV and extend up to the keV range. For intermediate mass number nuclei, where \( 15 < A < 80 \), the resolved energy range generally starts at \( 10^{-4} \) eV and extends into the MeV range. In the basic nuclear data files such as the ENDF/B libraries for nuclei with \( A > 80 \), the cross-section representation in the resolved resonance region is obtained through evaluated resonance parameters, together with a nuclear cross-section model. In contrast to the parametric representation, nuclei where \( 15 < A < 80 \), the cross sections in the basic nuclear data libraries are given as energy pointwise data values with the cross sections at infinite dilution, except for important structural nuclides such as iron, nickel, etc. An assessment of the pointwise cross-section representation and its adequacy for criticality safety calculations is discussed.
To illustrate the advantage of a parametric representation of the cross sections as opposed to the pointwise representation, infinite multiplication factor ($k_\infty$) calculations with the two representations are presented. The results correspond to calculations performed with a mixture of $^{27}$Al and $^{235}$U. The calculations presented follow the response of the Nuclear Engineering Application Section (NEAS) at the Oak Ridge National Laboratory (ORNL) to an article published in Criticality Safety Quarterly. In the winter of 1993, an article [1] in the Criticality Safety Quarterly pointed out differences in calculated infinite multiplication factors obtained from the SCALE code system [2] and the MCNP code [3]. The systems considered were fictitious mixtures of several metals individually mixed with $^{235}$U, including $^{27}$Al and $^{235}$U mixtures. Since the MCNP calculational methodology is based on the continuous energy approach, it was assumed that the MCNP calculations were more correct. In the absence of experimental data for these metal/$^{235}$U systems, it was decided to calculate the $k_\infty$ of the metal/$^{235}$U systems using a variety of computer codes and cross-section libraries. It was found out that none of the $k_\infty$ from these calculations agreed with MCNP. Since several sources of uncertainties contribute to the overall uncertainty in the integral results, the lack of agreement among the different calculational methodologies led us to investigate the methods and data used in the calculations. First, a detailed examination of the MCNP results suggested that the pointwise cross-section data in its library were not adequate for the systems under consideration. Particularly, it was found out that the aluminum cross sections in the ENDF library were given as smooth averaged cross sections, which cannot be properly energy self-shielded even though aluminum has resonance structure. The aluminum cross sections as given in the ENDF/B-V library are shown in Fig. 1.

They are, respectively, from top to bottom, total, elastic, and capture gamma cross sections. It is noted that the average capture gamma cross sections between 1 keV and 10 keV are poorly represented with not enough energy points. Had a parametric representation been given, the lack of detail in the cross section would not be present, and the resonance self-shielding effects on the cross sections would be properly accounted for. To verify this, a parametric evaluation of the $^{27}$Al cross sections was done [4]. The $k_\infty$ result with the pointwise cross-section representation is 0.9802, whereas the parametric representation yields 1.1176. Both calculations were performed with the SCALE code system with the 238-group library processed from the ENDF/B-V library. The neutron spectrum characterizing the Al/$^{235}$U system studied was in the intermediate energy range where the resonance self-shielding effects of the aluminum cross sections play an important role and, therefore, strongly indicates the pointwise cross-section representation of $^{27}$Al is inadequate.

In a separate study, a criticality safety assessment of the fuel cycle facility [5] at Argonne National Laboratory, Idaho Falls, also indicates a similar problem related to the pointwise-averaged cross-section representation. In this study, it was verified that the cross-section representation of chlorine (Cl)
[6], a material with resonance structure, should not be presented as smooth cross sections.

These issues highlight areas where there appears to be a deficiency in the ENDF evaluations, specifically data for materials used in criticality safety which have no resonance representation.

AN ASSESSMENT OF THE $^{233}$U CROSS-SECTION DATA FOR CRITICALITY SAFETY APPLICATIONS

An effort has recently been conducted to validate $^{233}$U cross-section data for applications in nuclear criticality safety problems [7,8]. The purpose of this work was to investigate the adequacy of the $^{233}$U cross-section data in ENDF/B versions IV and V with regard to criticality safety analyses of the fuel drain tank (FDT) and fuel flush tank (FFT) of the Molten Salt Reactor Experiment (MSRE) at ORNL. The first step in the study consisted of calculating the multiplication factors ($k_{\text{eff}}$) of 51 selected critical benchmark problems using the Monte Carlo criticality safety code KENO V.a of the SCALE system. Most of these benchmarks included mixtures of $^{233}$U and hydrogen with $^{233}$U in aqueous uranyl nitrate and uranyl fluoride. The calculations were carried out with multigroup libraries from the SCALE system which were processed using ENDF/B-IV and ENDF/B-V data. The overall results suggest an improvement of the calculated $k_{\text{eff}}$ using ENDF/B-V data versus ENDF/B-IV data. The validation procedure included results of calculations done with fine multigroup cross-section libraries based on ENDF/B-IV using a 218-group structure, and a 238-group structure based on data from ENDF/B-V. The Monte Carlo eigenvalues computed with the KENO code for each of the 51 benchmarks problems have been plotted against the average energy of the neutrons causing fission. Of the 51 critical benchmarks, 9 correspond to fast systems, and the remaining 42 are thermal systems. The results based on the 218-group structure are shown in Fig. 2, whereas the results of calculations performed with the 238-group structure are shown in Fig. 3.

Figure 2. Benchmark Results of 51 $^{233}$U Critical Experiments for ENDF/B-IV Library.

Figure 3. Benchmark Results of 51 $^{233}$U Critical Experiments for ENDF/B-V Library.
The ENDF/B-IV calculations consistently overpredict the eigenvalues in the thermal energy range and underpredict them in the fast range. The results obtained with the 238-group structure processed from ENDF/B-V also follow this behavior, but, in this case, the bias has been greatly reduced. The overall integral results shown in Fig. 3 clearly demonstrate an improvement in the calculated $k_{eff}$ obtained with ENDF/B-V in contrast to ENDF/B-IV, and the global improvement on the $k_{eff}$ of the 51 critical benchmark experiments can be definitely attributed to the better representation of the $^{233}$U cross section in ENDF/B-V.

In spite of these improvements, problems still remain. A further investigation of the bias associated with the calculated $k_{eff}$ of the 42 thermal critical benchmarks was performed. Figure 4 is a plot of $k_{eff}$ as a function of the average energy of neutrons causing fission obtained with calculations using ENDF/B-V.

It should be noted that the ENDF/B-V results are not good for critical systems in the intermediate energy range. The intermediate energy range is very important in nuclear criticality safety for systems such as the FDT and FFT of the MSRE. To illustrate the need for better $^{235}$U cross-section data for criticality safety issues concerning the MSRE, the percentage of neutrons per energy that cause fission for the FDT is shown in Fig. 5.

This picture clearly points out the importance of the intermediate energy range of the FDT system. Both ENDF/B-V and ENDF/B-IV data for $^{235}$U are based on a 1978 evaluation which employs the Adler-Adler formalism to represent the resolved energy range cross sections. Here the resolved energy range extends to 60 eV. This evaluation is clearly more physically correct than the earlier ENDF/B-IV evaluation which used the single-
level Breit-Wigner formalism that is known to be inappropriate for fissile isotopes, such as $^{233}$U. However, in order to address the MSRE criticality safety concerns, the $^{233}$U cross-section data should be re-evaluated with the Reich-Moore formalism, and the resolved energy range should be extended to 500 eV to improve the calculation of level-level interference resulting in cross-section energy self-shielding. The extension of the resolved energy range can be accomplished by taking advantage of the high resolution feature of the Oak Ridge Linear Accelerator (ORLA). The computer code SAMMY \[9\] can be utilized to provide a set of Reich-Moore resonance parameters.

$^{235}$U CROSS SECTION FOR CRITICALITY SAFETY CALCULATIONS

The present $^{235}$U cross-section evaluation adopted in the ENDF/B-VI covers an energy range of $10^{-5}$ eV to 2250 eV. This evaluation is based on the Reich-Moore formalism and represents a substantial improvement over the previous ENDF/B-V evaluations based on the single-level Breit-Wigner formalism. Despite the improvements, the $^{235}$U evaluation does not reproduce the measured capture-to-fission ratio (alpha), and consequently impacts criticality safety analyses of highly enriched intermediate spectrum systems. To address the $^{235}$U alpha problem, the ENDF/B-VI resonances were adjusted \[10\] to obtain the measured capture-to-fission ratio. To do this, the resonance widths $\Gamma_\gamma$, $\Gamma_\nu$, and $\Gamma_r$ were adjusted in the energy range from 0 to 900 eV. This procedure gives a set of resonance parameters where calculated alpha agrees with the experimental results.

Two critical experimental benchmarks, UH3-UR\[11\] and HISS(HUG) \[12\], were calculated with the SCALE system using $^{235}$U cross sections from the original ENDF/B-VI resonance parameters and with the modified parameters. These calculations were performed using the VITAMIN-B6 199-group cross-section library. The UH3-UR is a uranium hydride system with a uranium reflector. The HISS(HUG) is a homogeneous uranium-graphite system. The calculated effective multiplication factors, $k_{\text{eff}}$, of these systems are shown in Table I.

<table>
<thead>
<tr>
<th>Benchmark</th>
<th>ENDF/B-VI</th>
<th>Lubitz</th>
</tr>
</thead>
<tbody>
<tr>
<td>UH3-UR</td>
<td>1.0222</td>
<td>1.0093</td>
</tr>
<tr>
<td>HISS(HUG)</td>
<td>1.0293</td>
<td>1.0133</td>
</tr>
</tbody>
</table>

The calculations based on the ENDF/B-VI resonance parameters indicate 2–3 % discrepancy in $k_{\text{eff}}$ values. The discrepancies decrease with calculations using the adjusted set of resonance parameters. However, a 1.3 % bias is still observed for the HISS(HUG) system. The systems under consideration are very sensitive to $^{235}$U cross sections, and errors may be due to inaccurate values of these data. Note that these are epithermal systems with intermediate neutron energy spectra in the $^{235}$U resolved energy range.

Another drawback of the ENDF/B-VI $^{235}$U cross-section evaluation is that the energy range ($10^{-5}$ to 2250 eV) is represented by eleven consecutive disjoint sets of resonance parameters that cause nonphysical discontinuities in the cross sections at the energy boundary of consecutive resonance sets. This mismatch is illustrated in Table II for the total cross section calculated at a temperature of 300 K using consecutive sets. They are indicated as the set below and above the boundary energy under consideration.
NUCLEAR DATA NEEDS FOR CRITICALITY SAFETY ANALYSES

Nuclear systems commonly found in criticality safety applications use a large number of fissionable, moderator, and absorber materials. The characteristic neutron spectrum of these systems spans all the way from thermal to very high energies. Of particular interest for many current applications are systems with a neutron spectrum which peaks in the intermediate energy range. In general, the intermediate energy range is between 3 eV and 3 keV. A system is considered an intermediate system when the energy corresponding to the average lethargy of the neutron causing fission (AEF) falls in this range. This energy range, from the viewpoint of differential data, is extremely important since resonance effects in the cross sections are dominant. The status of nuclear data for the majority of isotopes of interest to criticality safety applications in the basic nuclear data files has to be examined. As an example, Table 3 lists eight materials of interest in criticality safety identified as having questionable ENDF/B-VI evaluations. There are a variety of other isotopes that probably should be added to the list in Table 3.

From the point of view of integral data, there is a need for high quality critical experiments with intermediate energy spectra (3 eV to 3 keV) to validate methods and data. Integral results generally are not suitable for determining whether specific reaction types, such as fission or capture cross sections, are adequately represented. However, they can be used to judge the overall performance of the basic data. An assessment of existing experimental data relevant to criticality safety applications has to be performed. Table IV shows experiments of interest for data assessment for criticality safety applications.

Table II. Total Cross Section at the Energy Boundary

<table>
<thead>
<tr>
<th>Energy Boundaries (eV)</th>
<th>$\sigma_t$ (barns) Set Below</th>
<th>$\sigma_t$ (barns) Set Above</th>
</tr>
</thead>
<tbody>
<tr>
<td>4</td>
<td>10.72</td>
<td>10.73</td>
</tr>
<tr>
<td>110</td>
<td>72.75</td>
<td>82.1</td>
</tr>
<tr>
<td>300</td>
<td>18.94</td>
<td>19.31</td>
</tr>
<tr>
<td>500</td>
<td>47.58</td>
<td>46.51</td>
</tr>
<tr>
<td>750</td>
<td>32.96</td>
<td>33.01</td>
</tr>
<tr>
<td>1000</td>
<td>20.48</td>
<td>20.12</td>
</tr>
<tr>
<td>1250</td>
<td>22.07</td>
<td>22.05</td>
</tr>
<tr>
<td>1500</td>
<td>19.62</td>
<td>17.49</td>
</tr>
<tr>
<td>1750</td>
<td>41.4</td>
<td>27.22</td>
</tr>
<tr>
<td>2000</td>
<td>25.87</td>
<td>31.28</td>
</tr>
</tbody>
</table>

Table III. Isotopes For Which Measurements and Evaluation Are Needed

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Cross-Section Representation</th>
<th>Last evaluation</th>
<th>Resonance data</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{20}\text{Ca}$</td>
<td>Aug71</td>
<td>No*</td>
<td></td>
</tr>
<tr>
<td>$^{27}\text{Al}$</td>
<td>Dec73</td>
<td>No</td>
<td></td>
</tr>
<tr>
<td>$^{17}\text{Cl}$</td>
<td>Feb67</td>
<td>No*</td>
<td></td>
</tr>
<tr>
<td>$^{19}\text{K}$</td>
<td>Feb67</td>
<td>No*</td>
<td></td>
</tr>
<tr>
<td>$^{31}\text{P}$</td>
<td>Oct77</td>
<td>No</td>
<td></td>
</tr>
<tr>
<td>$^{14}\text{Si}$</td>
<td>Feb74</td>
<td>No*</td>
<td></td>
</tr>
<tr>
<td>$^{12}\text{Mg}$</td>
<td>Feb78</td>
<td>No*</td>
<td></td>
</tr>
<tr>
<td>$^{233}\text{U}$</td>
<td>Dec78</td>
<td>Yes</td>
<td></td>
</tr>
</tbody>
</table>

*Isotopic evaluations needed.

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Table IV. Critical Experiments With Intermediate Energy Spectra.

<table>
<thead>
<tr>
<th>Critical Experiments</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>HISS(HUG)</td>
<td>Homogeneous Uranium-Graphite</td>
</tr>
<tr>
<td>HISS(HPG)</td>
<td>Homogeneous Plutonium-Graphite</td>
</tr>
<tr>
<td>UH₃-UR</td>
<td>High Enriched Uranium with Low Enriched Uranium Reflector</td>
</tr>
<tr>
<td>PCTR</td>
<td>Low Enriched UO₂/H₂O with a Range of H/U</td>
</tr>
</tbody>
</table>

ACKNOWLEDGMENT

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REFERENCES


4. ORNL Internal Correspondence from R. Q. Wright to C. V. Parks, March 29, 1994.


CONCLUDING REMARKS

In this paper we have discussed the adequacy of the basic data in existing nuclear data libraries for application in nuclear criticality safety evaluations. It has been pointed out that the pointwise representation of the cross sections of nuclei where 15<A<80 have to be avoided. A parametric representation of the cross sections is needed for the evaluation of the energy self-shielding effects. For these nuclei, the resonance structure in the cross sections is very important since the resolved energy range is in the intermediate energy range so important in criticality safety problems.

The MSRE problem suggests that the extension of the resolved energy range of fissile isotopes such as ²³³U is highly recommended. Also, a cross section model using the Reich-Moore formalism should be used for fissile isotopes.


