DOE SPENT NUCLEAR FUEL — NUCLEAR CRITICALITY SAFETY CHALLENGES AND SAFEGUARDS INITIATIVES

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ABSTRACT

The field of nuclear criticality safety is confronted with growing technical challenges and the need for forward-thinking initiatives to address and resolve issues surrounding economic, safe and secure packaging, transport, interim storage, and long-term disposal of spent nuclear fuel. These challenges are reflected in multiparameter problems involving optimization of packaging designs for maximizing the density of material per package while ensuring subcriticality and safety under variable normal and hypothetical transport and storage conditions and for minimizing costs. Historic and recently revealed uncertainties in basic data used for performing nuclear subcriticality evaluations and safety analyses highlight the need to be vigilant in assessing the validity and range of applicability of calculational evaluations that represent extrapolations from "benchmark" data. Examples of these uncertainties are provided. Additionally, uncertainties resulting from the safeguarding of various forms of fissionable materials in transit and storage are discussed.

I. INTRODUCTION

Changes in world politics, social emphases for "greening-the-earth," governmental and political drives toward disarmament and weapon dismantlement, disposal of excess weapons materials, long-term placement of transuranic wastes and discard of spent nuclear fuels is propelling the nuclear criticality safety community at ever-increasing speeds toward simultaneously accommodating political, physical, and economic requirements while satisfying safeguards and safety issues. These requirements are being imposed with a political urgency comparable to the wartime urgency of the "Manhattan Project" period. To the uninitiated, there is the notion that there is little or no more technical knowledge or data needed for nuclear criticality safety than that already obtained from the "Project" days and the power reactor design "hay-days" supplemented with a few new or reevaluated differential neutron cross-section measurements data and data processing. The notion is that today's fissionable material problems are readily solvable with the tools and data at hand. Such a notion is not false in all instances. However, research and development of computational techniques, measured neutron cross-section data and its processing, and integral critical experiments are needed to competently evaluate the safety of the various forms of fissionable materials being encountered with new and evolving safeguarded environments (e.g., interim storage, shipping, and burial). The uncertainties in the adequacy of the data and the bias for a particular application have led to the use of "conservative" assumptions that are used to rationalize safety.

Examples of uncertainties in the knowledge and data bases frequently used for safety evaluations are provided. Also, some alternative approaches to addressing these uncertainties are offered. The examples of uncertainties include

- Studied discrepancies between calculations and experiments involving reflected and unreflected arrays of fissile solution units, resulting in as much as a 3% discrepancy in $k_{\text{eff}}$.
- A series of benchmark integral fission reaction rate measurements of neutron slowing down in and leakage from water spheres, resulting in as much as an 11% discrepancy in calculated to measured fission detector responses from leakage neutrons.
- Recent computational experiments with neutron cross-section data demonstrating as much as 43% differences by using various neutron cross-section libraries with various computational tools.
- Computational evaluations of historic $^{235}$U Molten Salt Reactor Experiment fuel in-reactor and in-storage environments resulting in as much as a 5% difference by using various neutron cross-section libraries with a single computational tool.

Understanding and addressing the relevancy of these uncertainties is necessary to ensure nuclear criticality safety for the evolving issues surrounding safeguarded transport, storage, and long-term disposal of fissionable materials.
II. DISCREPANCIES BETWEEN REFLECTED AND UNREFLECTED FISSILE SOLUTION UNIT ARRAYS

Computational studies, reported in reference 1, were undertaken to resolve long-standing discrepancies between calculations and experiments involving arrays of fissile solution units. The study demonstrated that "room return" of scattered neutrons was sufficient to account for calculated discrepancies of some bare arrays, but calculational results for reflected arrays were observed to remain in disagreement. Additionally, the magnitude of the neutron "room return" raises other unresolved issues.

The calculational study concentrated on a selected series of critical bare and reflected (plexiglas and paraffin) arrays of 5-liter units filled with uranyl nitrate enriched to 93 weight percent $^{235}$U. Results of the computational study for the critical systems showed that for bare, cubic three-dimensional (3-D) arrays the calculated $k_{ef}$ decreased about 2% to 3% as the bare-array size increased from a two-cubed array to a five-cubed array. However, the reflected arrays calculated 2 to 3 percent higher than the bare arrays. An attempt was made to determine the feature of the experiments that might be responsible for the discrepancies by calculating a large number of other experiments.

Two other independent series of array experiments exhibited similar behavior—one involving arrays of solution slabs, and a set of experiments involving plutonium metal units. These experiments also had large unit surface-to-surface separations in the arrays. As in the cases of the cylindrical unit arrays, as the unit separation increased, the calculated $k_{ef}$ values decreased.

When the arrays were modeled to include room boundaries (i.e., concrete walls, floor, and ceiling), the bare array discrepancies reduced to little more than the basic bias of the cross sections used in the calculations. However, the reflected array discrepancies persisted. Further calculational studies were performed on the additional series of experiments to examine the effects of "room return," and they resulted in very similar results.

Other attempts to localize the causes of the discrepancies involved the computational evaluations of numerous hydrogenously moderated homogeneous single units, bare and reflected. Also, submerged lattices of low-enriched uranium rods were studied. Results of these studies did not reveal similar discrepancies.

Further computational studies were conducted to examine the density scaling rule for a bare fissile material system, that is, the critical mass of a bare fissile material system will vary inversely with regard to the square of system material density. This relationship is frequently stated as

$$M = M_0 \left( \frac{\rho}{\rho_0} \right)^{-2}$$

where $M$ = critical mass of bare system at material density, $\rho$, and $M_0$ = critical mass of bare system at material density, $\rho_0$.

In this computational study, this relationship was applied to bare arrays of multiple fissile units to examine consistency with the density scaling rule. This was done by equating the number of units within two arrays, $N$ and $N_0$, with the mass of material in two systems, $M$ and $M_0$. Likewise, the ratio of the densities, $\rho/\rho_0$, is taken to be the ratio of density reduction for the array such that:

$$N = N_0 \left( \frac{\rho}{\rho_0} \right)^{-2}$$

The negative exponent deviated from about -2.00 for very large arrays ($N = 1000$) to less than about -2.05 for small arrays ($N \leq 64$). Figure 1 provides a plot of this variation that is judged to be atypical of the density scaling rule and is difficult to resolve.

The conclusion drawn from the study was that "... our understanding of the fission process in aqueous systems of uranium is incomplete."

III. POINT NEUTRON SOURCE SCATTERING, SLOWING DOWN, AND LEAKAGE BENCHMARKS WITHIN WATER SPHERES

The National Institute of Standards and Technology (NIST) conducted a series of highly controlled measurements between the years 1989 and 1994 (reported in references 2, 3, and 4). The purpose of the experiments was to compare "state-of-the-art" computational results with standardized measurements using highly characterized $^{235}$U, $^{239}$Pu, $^{238}$U, and $^{237}$Np fission chambers to evaluate the neutron leakage of a near-point central $^{252}$Cf neutron source in water spheres of three different diameters: 3, 4, and 5 in. (38.1, 50.8, and 63.5 mm). Figure 2 provides a schematic of a typical setup of the experimental apparatus. The fission chambers were typically used as shown in Figure 2, and an identical series of measurements were conducted with the fission chambers covered with cadmium to permit measurements of epicadmium fluences in the leakage spectrum. Substantial effort was expended to ensure the correct modeling of the input data for performing the MCNP 3-D Monte Carlo calculations. The use of approximately 280 surfaces for detailing the features of the apparatus exemplify this.
for all measurements is from a 1.5 to 1.9% combined uncertainty from all effort. ENDF-B/V point cross-section data and a "light water" 
$S(\alpha,\beta)$ scattering kernel was used in the calculations in concert with the NIST californium fission neutron spectrum.

Though the root-sum-of-squares uncertainties of the calculated-to-experimental (C/E) ratios are being reevaluated, it is believed that the combined uncertainty of the C/E ratios for all measurements is on the order of 1.9 to 3.6%. The reported C/E uncertainties for all the measurements are derived from a 1.5 to 1.9% combined uncertainty from all known experimental measurement factors and a 0.7 to 3.0% statistical uncertainty only of all the Monte Carlo calculations.

The C/E ratios varied from a low value of 0.931 ± 2.4% for a Cd-covered $^{237}$Np neutron leakage measurement from a 4-in. (101.6-mm)-diam water sphere to a high value of 1.108 ± 2.6% for a Cd-covered $^{235}$U neutron leakage measurement from a 5-in. (127.0-mm)-diam water sphere. Though these variational extremes are about 17.4% from one another, only one trend with water sphere diameter was observed from all C/E values. The single trend was observed for the Cd-covered $^{235}$U fission chamber. The C/E ratios were 1.024 ± 2.3%, 1.050 ± 2.5%, and 1.108 ± 2.6% for the Cd-covered $^{235}$U fission chamber. The C/E ratios were 1.024 ± 2.3%, 1.050 ± 2.5%, and 1.108 ± 2.6% for the 3-, 4-, and 5-in. (76.2-, 101.6-, and 127.0-mm)-diam water spheres, respectively. The odds of this nearly 8% error not being a trend is unlikely considering the uncertainties of the three C/E ratios.

Conclusions drawn from this series of computational and experimental evaluations were the following:

- Further experimental and computational studies should be performed to address, at least, the specific, significant trend between the calculated and measured leakage spectra for water moderators.
- Measurements/calculations should be performed for more finely spaced water sphere radii to examine the thermal and epithermal neutron energy systems.
- A thorough review and reexamination of the bases for neutron scattering and the slowing-down process should be considered. This review should examine:
  - the basic experimental data upon which the scattering kernel is based, and
  - the feasibility of reevaluating or improving the rigor of the $S(\alpha,\beta)$ scattering kernel approximations considering today's computing power.

IV. VARIATIONS IN BASIC DATA AND COMPUTATIONAL METHODOLOGIES

A recent article$^4$ was published within the nuclear criticality safety community that highlighted significant variations in calculated $k_e$ results for homogenous systems of $^{238}$U as mixed with typical materials used in reactor fuel designs and, to a lesser extent, materials of construction in storage or transportation (i.e., zirconium, iron, and aluminum). The calculated systems had aluminum, iron, and zirconium to $^{235}$U atom ratios of 2470, 320, and 103, respectively. The fallible premise presented by the article was that calculational codes that use continuous cross-section data are most likely to produce correct results. This incorrect premise was judged to be valid based upon presenting the results of group-wise calculations that had extreme variabilities in $k_e$ results for systems having alleged $k_e = 1.0$. In one extreme, the $k_e = 1.38$ for $Zr^{235}$U = 320 using one of the four different group-wise neutron cross-section libraries. In the other extreme, the $k_e = 0.80$ for Fe$^{235}$U = 320 using a different group-wise cross-section library. In the extreme, this represents a 52% variation in results.

In a subsequent study,$^4$ various calculational codes were teamed with "continuous-energy, point-energy, ultra-fine energy-group, and fine energy-group" cross-section libraries to compare results for the described systems in reference 5. Sources of variability in the results were examined. It was determined that the base case using "continuous-energy" cross-section data of reference 5 did not provide treatment of unresolved resonances in the cross-section data. Upon appropriate selection, processing, and use of cross-section libraries and proper application of calculational codes for their intended applications, the variability of results was reduced from an extreme variation of 52 to about 11%. The unacceptable 11% variation in results was seen for both groupwise and continuous-energy codes and data. Cross-section data issues still remain with regard to the adequacy of resonance structure and treatments at the neutron energies encountered with these systems.

Typical energy-group cross-section libraries used today are severely tested by such poorly moderated, intermediate-to-epithermal neutron energy systems. From the evolution of neutron cross-section measurements, evaluations, processing and computational code developments, it is clear that calculations of benchmark experiments have had a national emphasis$^{12}$ (government and industry) on properly calculating "thermal" or "fast" systems. This emphasis was based upon needing to ensure adequate safety and designs for research and power-production reactor initiatives. As such, there have been very few, if any, critical experiments that have addressed intermediate to epithermal energy systems like those found in nonreactor environments (e.g., fissionable material fabrication processes, waste collection/processing, transportation and shielding, etc.). In reality, such extreme materials as posed in the article of reference 5 can be found in unmoderated fabrication processes of research reactor and power reactor fuel element fabrication, transport, storage and reprocessing environments.
The primary message derived from this subsequent study was that "... the criticality safety community must continue to encourage nuclear data development and the understanding of the physics of neutron interactions in light of new and ever-changing applications." In particular, without relevant critical experiments for validation of the codes and cross sections one cannot establish the bias of nuclear criticality safety evaluations in circumstances such as these.

V. LEGACY UNCERTAINTIES

Nuclear criticality safety organizations are frequently confronted with having to evaluate fabrication, waste recovery, packaging, storage and transportation of fissionable materials. These evaluations are ideally performed in preliminary design stages, thereby permitting flexibility in the approach for solving the "safety problem." Occasionally the design process will get ahead of the safety evaluation process, or an existing circumstance will be discovered that requires "after-the-fact" analysis and proof of safety. The after-the-fact circumstance can be a previously acceptable situation gone awry, or it can be a need to accommodate more exacting regulatory expectations regarding the demonstration of subcriticality and safety. A circumstance of the later type was recently addressed at the Oak Ridge National Laboratory (ORNL).7

From 1965 to late in 1969, ORNL operated the Molten Salt Reactor Experiment (MSRE) which was moderated with a graphite lattice. When the reactor was directed to be "shut down" in 1969, the molten salt (comprised of 84 weight percent of \( \text{U}^{235} \) as 48.1 kg of UF₆ homogeneously mixed with 1985.3 kg of LiF, 1685.3 kg BaF₂, 989.3 kg ZrF₄, and 0.84 kg PuF₃) was drained into two large drain tanks within a shielded "hot cell" for interim storage (now 25 years). The subcriticality and safety of the drain tanks had previously been evaluated in the "safety analysis report" using diffusion theory techniques that were predicated on the limited data base of "fast" and "thermal" benchmarks that were further supported by the MSRE startup and operation. Over the following years subcriticality evaluations were performed with varying degrees of rigor and documentation. Recently, ORNL has undertaken the initiative to reevaluate the subcriticality and safety of fissionable material conditions at the MSRE. Reference 7 is a part of that initiative regarding the documentation of the subcriticality and safety of the MSRE Drain Tanks that were modeled in 3-D detail, including steam thimbles, salt content, materials of tank construction and cell shielding for a Monte Carlo evaluation.

Due to the paucity of benchmarks regarding such a strange mixture of isotopes and elements, the MSRE was explicitly modeled at temperature in three dimensions including control rods, molten fuel, graphite stringers, base plate for graphite stringers, reactor vessel and shielding. Absolute and relative calculated control rod worths duplicated experimental results when using a particular neutron cross-section library. However, use of other neutron cross-section libraries introduced differences in results on the order of ~5% in calculated \( k_{\text{eff}} \).

Because of the observed neutron cross-section influences on the calculated results for the reactor model, the same suite of neutron cross-section libraries were used in the calculations of the drain tank evaluations. The results of the MSRE Drain Tank calculations varied ~14% in calculated \( k_{\text{eff}} \).

Calculations for the two basic models (i.e., the reactor and drain tanks under normal conditions) were performed with a quality assured version of the SCALE13 CSAS25 sequence. Results are provided in Table 2. As can be observed, the library referred to as the 27GROUPNDF4 provides the largest spread in \( k_{\text{eff}} \) (1.005 - 0.860 = 0.145) between the MSRE reactor model and the fuel drain tank (FDT) model. Additionally, of the various libraries examined, the 27GROUPNDF4 library provides the largest result for the drain tank model. The use of the 27GROUPNDF4 neutron cross-section library is assumed to provide the most conservative result of the libraries examined.

Because of the observed reversing biases when using different cross-section libraries, a review of the neutron energy spectrum causing fission was performed to gain insight. A plot of this variable is provided in Figure 3. Clearly, the MSRE reactor (Rx) model is not a representative benchmark for the MSRE FDT model. However, without the resources of time, money, and people to produce an "iron-clad" theoretical statistical sensitivity analysis of cross-section uncertainties and without readily available critical experiments facilities and materials, the evaluators were limited to the reported analysis approach. Surely, the maximum \( k_{\text{eff}} = 0.86 \) for the FDT is adequately subcritical from the minimum \( k_{\text{eff}} = 0.98 \) for the Rx to assure subcriticality of the FDT system. A greater degree of understanding in the variability of results could be ensured if the recent uncertainty tables of the new ENDF/B-VI were used to perform flux-weighted statistical sensitivity calculations associated with the cross-section data measurement uncertainties. An even greater degree of understanding would be achieved through exponential experiments. Ideally, critical experiments should be performed by using similar materials (the existing material is contaminated with fission products and a large percentage of \( \text{U}^{235} \) and \( \text{Pu}^{239} \) that pose a substantial radiation hazard).

VI. SAFETY CHALLENGES AND SAFEGUARDS INITIATIVES

The discussions of the foregoing physical and computational studies were offered to demonstrate the uncertainties in "known" margins of subcriticality as applied to nuclear criticality safety analyses of "real world" situations.
With the social and political push to consolidate and remediate accumulations of spent fuel and weapons-grade materials, the nuclear criticality safety community is becoming increasingly subjected to circumstances requiring more useable and sophisticated evaluation tools and skills to address the complex issues. Such developing circumstances include:

- Increasing densification of spent fuel in wet pool storage with fixed neutron poisons.
- Increasing densification of spent fuel in dry cask storage.
- Relatively long-term storage of large arrays of neutron-interacting low-enriched uranium shipping packages that were designed for limited-time transport and storage.
- Safeguarding of fissionable materials within heavy-duty, thick-walled vaults or underground environments (e.g., like salt domes) that offer heretofore unexamined neutron reflection characteristics.
- Neutron interaction through large thicknesses of structural and shielding materials having limited or no benchmark experiments such as large quantities and thicknesses of lead, stainless steel, iron, depleted uranium, concrete, polyethylene, and others.
- Increased regulatory expectations to define, justify and defend the determination and uncertainty of calculated values that are used to determine margins of subcriticality for safety analysis purposes.

Each of the above-mentioned developing circumstances have technical issues very similar to the four examples provided in this paper, that is, the interacting neutron energy spectra are skewed into energy regions for which few or no benchmarks exist.

VII. ALTERNATIVE APPROACHES

Alternatives for solving these types of problems are limited to:

- **inexpensive** crude and macroscopic "good-feeling or professional-judgement" approaches (e.g., reference 7),
- **possibly expensive** subcritical reactivity measurements for material characterization or measurement of criticality code calculated parameters (e.g., reference 14),
- **expensive** statistical evaluations of cross-section uncertainties, using perturbation theory, as weighted by the presumed problem-dependent neutron energy spectrum (e.g., reference 15),
- **possibly very expensive** reactor physics measurements for neutron characterization of materials through replacement and material-worth measurements (e.g., use of a physical constants test reactor such as in references 16 and 17),
- **very expensive** differential cross-section measurements of materials, reaction rates, and neutron energies of interest (e.g., reference 18),
- **most expensive** critical experiments of identical or similar materials anticipated to be encountered (e.g., reference 19).

The deceiving elements in these approaches are their alleged costs. As long as the "professional-judgement" approach is successful in supplanting hard statistical or experimental data, the cost is quite low. When this approach is called into question or fails, the costs become very large indeed. Historically, the nuclear criticality safety community has not marshalled the technical and theoretical arguments necessary to require the use of necessary analytical and experimental resources for scientifically addressing and documenting the issues surrounding fissionable material transportation, storage and operational processes. Such resources only came to bear on situations surrounding weapon designs, weapons production processes, research reactors and power-production reactor designs—all of which had substantial fiscal and national security reliability incentives. Criticality safety issues were typically addressed at a lower level of technical expertise with left-over resources.

Recently the Nuclear Engineering Applications Section at ORNL has proposed to the Department of Energy a technical software project that would develop, consolidate, and streamline the computational processes necessary for performing sensitivity analyses using standard perturbation theory approaches, ultimately in 3-D systems. Such an approach should permit the evaluation of the adequacy, or inadequacy, for the application of existing data to nuclear criticality safety evaluations.

VIII. CONCLUSIONS

Obvious situations exist and are developing in the area of nuclear criticality safety evaluations that inadequately define margins of subcriticality for use in nuclear criticality safety analyses. Ignorance of these circumstances or inadequacy of such evaluations are becoming more unacceptable in the view of the public, legislators, regulators, and corporate nuclear workers exposed to liability for operations. The only defensible approach for the nuclear criticality safety community is to move into the world of scientific determination of subcriticality and associated uncertainties and reserve the use of "professional judgements" for the selection of alternative analytical approaches.

The ultimate benefits from scientifically determining subcriticality and associated uncertainties will be the recognition by the nuclear criticality safety community of the
uncertainties of the computational evaluation methods, their lack of understanding, and the potential for nonconservative results which may have been unknowingly included in safety analyses.

The resolution or minimization of the uncertainties, through theoretical analysis of the results, permits improved safety analyses and, in some instances, improved operating efficiencies.

The down side to the scientific approach is that some safety evaluations/analyses and process efficiencies may be adversely affected as a result of inadequate conservatism subsequently recognized by a better understanding of the nuclear processes and improved nuclear data base. However, an example of potential benefit is the recognition that, in many storage array analyses, a 3% change in $k_{eff}$ can result in a 100% change in allowable array capacity. Though such results cannot be guaranteed, they may be possible.

![Figure 1. Density Analog Representation of Bare Arrays Containing Square Circular Cylinders of 93 Weight % UO$_2$(NO$_3$)$_2$ at 415 g U/L.](image1)

![Figure 2. Typical Neutron Leakage Measurement Experimental Setup.](image2)

![Figure 3. Percentage of Neutrons Causing Fissions by Neutron Energy Group.](image3)

### Table 1

<table>
<thead>
<tr>
<th>Fission Chamber Neutron Leakage Measurements</th>
<th>C/E and Uncertainty</th>
<th>Fission Chamber Non-Fluoride Neutron Leakage Measurements</th>
<th>C/E and Uncertainty</th>
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<td>$^{235}$U (0,0)</td>
<td>-1.008</td>
<td>$^{235}$U (0,0)</td>
<td>-0.943</td>
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<td>1.010 ± 2.3%</td>
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TABLE 2
Cross-Section Library Comparisons

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<th>Cross-section Library</th>
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<th>44GROUPNDF5</th>
<th>23GROUPNDF5</th>
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<td>0.8145 (0.0010)</td>
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(Results from preliminary ENDF/B-V SCALE library.)

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