Initial Investigations of the Criticality Safety Validation Basis for HA-LEU Transportation*

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INTRODUCTION

Many proposed advanced nuclear energy systems require so called high-assay low enriched uranium (HA-LEU), which has a ^{235}U enrichment >5 wt% and <20 wt%, where the current fleet of light water reactors (LWRs) utilize enrichments of 5 wt% or less. With few benchmark experiments in HA-LEU enrichment range, the interplay of the cross sections for multiple nuclei become important, especially for LEU experiments where ^{1}H, ^{16}O, ^{235}U, ^{238}U and others are applied simultaneously in the calculation of the integral k_{eff} response. With continuing evidence that nuclear data are tuned to reproduce expected k_{eff} values for selected criticality experiments, which likely do not include those with 20 wt% ^{235}U, particular attention must be given to validation.

This paper describes the impact of recent nuclear data changes on HA-LEU and provides a preliminary sensitivity/uncertainty-based assessment of the applicability of available of criticality experiments for the validation of licensing calculations for the transportation of small quantities of 20 wt% UF_6.

APPLICATION MODEL

The transportation container used in this study was the ES-4100 container designed at the Y-12 National Security Complex [1]. The container is designed to ship fresh research reactor fuel and holds four separate containment vessels. Each vessel has a 5-inch (12.7 cm) inner diameter with an inside height of 58 inches (147.3 cm). The overall package is 34 (86.36 cm) inches in diameter and 71 inches (180.3 cm) tall. A cast ceramic absorber containing boron carbide is included for criticality control, and the Kaolite insulation is also included in the package model. A 3D cutaway view of this model is shown in Figure 1. This package was selected as a potentially representative container for shipping HA-LEU UF_6, though no plans exist at this time for certifying it for this application.

The model includes 20 wt% enriched UF_6 homogenously mixed with water and polyethylene in the containment vessel. The UF_6 was modeled in a homogenous mixture with water and polyethylene because this approach was used in the calculations to demonstrate criticality safety for the package's certificate. An individual package is modeled with the maximum ^{235}U mass allowed in each containment vessel per the ES-4100 certificate, which is 1 kg per cylinder or 4 kg per package. This mass is insufficient for industrial production, but it could be useful for initial process testing.

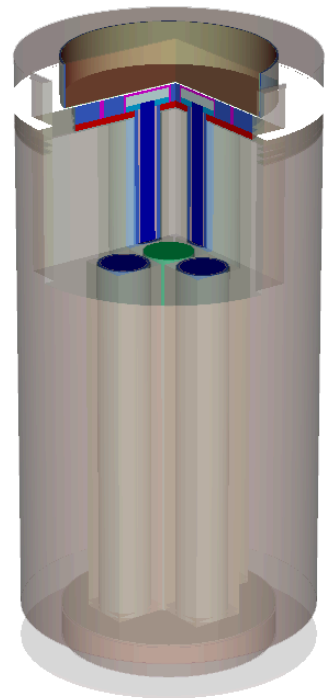

Figure 1. Cutaway view of a single ES-4100 container.

IMPACT OF NUCLEAR DATA TUNING

For files included in the evaluated nuclear data file (ENDF) library, nuclear data evaluations are produced by providing a best-fit representation of differential measurements [2]. However, it is not the "goodness" of a particular fit to differential data that is used as a performance metric of the evaluation; rather, the evaluation's performance is measured against integral quantities of interest. For example, the performance of a new n+^{235}U evaluation may be quantified by comparing the change in the ratio of computed-to-measured k_{eff} values for a series of evaluated benchmark experiments such as those compiled and maintained for the International Criticality Safety Benchmark Evaluation Project (ICSBEP).[3] If the new evaluation does not produce computed results consistent with measured values, then the evaluator may change one or more parts of the evaluation within the differential measurement uncertainties to provide better agreement with the measured integral quantities. Unfortunately, this informed adjustment of the evaluated parameterization may lead to compensation of errors between reaction data sets for a particular isotope. In turn,

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this results in cross section data having cross correlations between isotopes.

The 2018 release of ENDF/B-VIII.0 includes dramatic changes in cross sections for key nuclides relative to the 2011 release of ENDF/B-VII.1. For example, the several percent changes in the ^{235}U cross sections were generated using the Nuclear Data Sensitivity Tool (NDaST) from the Nuclear Energy Agency (NEA) as shown in **Error! Reference source not found.** [4].

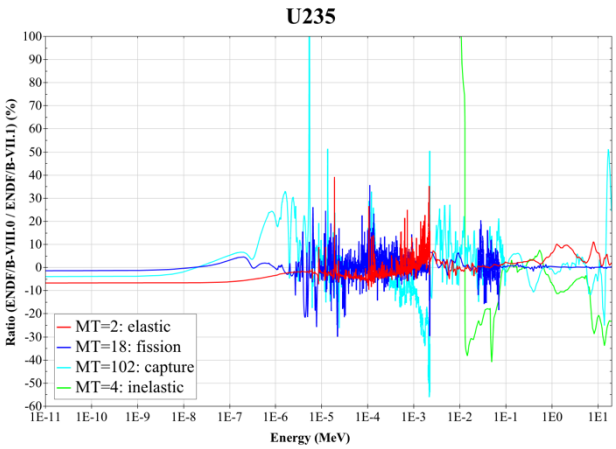

Figure 2. Changes in ^{235}U cross sections from ENDF/B-VII.1 to ENDF/B-VIII.0.

Previously, the error compensation between reaction data sets of a particular isotope has been demonstrated with ENDF/VII.1 and JEFF-3.2 [5], and a similar approach was applied to study the impact of the new set of compensating errors in ENDF/B-VIII.0 that were applied to reproduce the k_{eff} values of some benchmark experiments. For the ES-4100 KENO model, two base sets of continuous energy (CE) nuclear data (ENDF/B-VII.1 and ENDF/B-VIII.0) were combined to perform the computations. Starting with ENDF/B-VII.1, data from each ENDF/B-VIII.0 isotope were systematically replaced with data from the other base set. Once all ENDF/B-VIII.0 isotopic data had been substituted from the other base set, the ENDF/B-VII.1 isotopes were returned to their original base evaluation in the same order of their removal. Figure 3 shows the change in k_{eff} relative to the base result as ENDF/B-VII.1 isotopes were replaced with their ENDF/B-VIII.0 counterparts. The isotopes next to the arrows report the isotopic evaluation being entered into the library at that step. The statistical uncertainty on the computed k_{eff} was less than 10 pcm for all cases.

The base ENDF/B-VII.1 and ENDF/B-VIII.0 k_{eff} results differed by 65 pcm, indicating a good agreement between the integral performance of each base set of isotopic evaluations. However, there is up to a 450 pcm change in the computed k_{eff} due to the use of ^{235}U and ^{238}U from one base set with ^{1}H and ^{16}O from the other base set. Both ENDF/B-VII.1 and ENDF/B-VIII.0 have similar integral performances due to the compensating errors of important differential data isotopic evaluations. This clearly demonstrates a cross correlation between reaction data sets of different isotopes within a library that should be reported in the corresponding evaluations, and it calls into question the use of individual cross sections for predictions of reaction rates and power distributions of interest for nuclear energy applications. This study demonstrated the need for a strong validation basis for safety calculations.

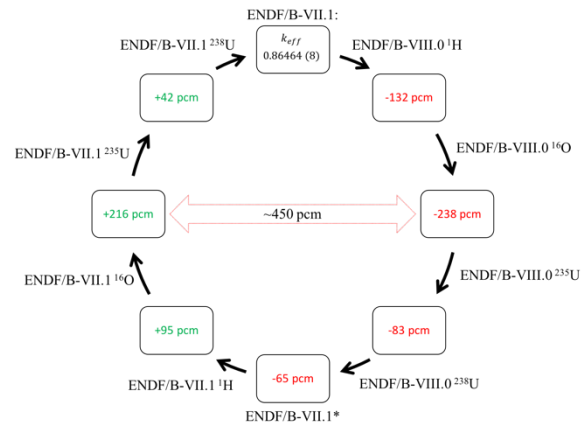

Figure 3. Effects of tuned nuclear data evaluations on HA-LEU transportation package criticality safety calculations.

APPLICABILITY OF CRITICALITY EXPERIMENTS

The TSUNAMI-3D sequence from the SCALE 6.2.3 nuclear simulation suite [6] was used to generate sensitivity data for the ES-4100 models. These sensitivity data were necessary to allow for the quantitative assessment of similarity between the application cases and the suite of benchmark critical experiments. This assessment was performed using the TSUNAMI Indices and Parameters (IP) code: specifically, the c_k integral parameter. The details of the c_k parameter are provided in Rearden et al. 2011 [7]. In this work, c_k values of at least 0.8 are considered to indicate acceptable similarity for use in validation. All results presented here implement the ENDF/B-VII.1 data and covariance libraries from SCALE 6.2.3.

The TSUNAMI approach to validation has been documented in US Nuclear Regulatory Commission guidance for fuel cycle facilities and for transportation and storage of fissile material [8, 9, 10].

CRITICAL EXPERIMENTS CONSIDERED

A suite of 1,584 critical experiments with available sensitivity data was assembled for this effort to identify existing applicable experiments for HA-LEU validation [11]. The majority of the sensitivity data for the experiments are drawn from the ICSBEP Handbook, and the remainder are drawn from the ORNL Verified, Archived Library of Inputs and Data (VALID) [12]. A total

of 181 intermediate-enrichment uranium (IEU) systems are included in the suite. These systems have fast, intermediate, mixed, and thermal spectra, and they are metal, solution, or compound systems. The enrichment range for the systems in the suite categorized as intermediate by the ICSBEP Handbook is 8.88–95.98 wt%. These 181 cases represent the majority of the IEU experiments for which sensitivity data are available from the ICSBEP Handbook. The remaining 1,403 experiments in the suite are fueled (LEU). Fast, mixed, and thermal neutron spectra are represented, as are metal, compound, and solution systems. The enrichment range for these systems is 0.71–10.07 wt%. One case with an enrichment of 93.19 wt% is included in an LEU evaluation and is therefore included in this suite, as well.

APPLICABLE EXPERIMENTS

All 1,584 experiments in the suite are compared in the application to determine which experiments are applicable for validation of k_{eff} calculations of the ES-4100 containing UF_6. While the results may be representative for future shipping packages and should not be interpreted as being generically applicable to all systems, they do indicate the applicability of existing critical experiments for HA-LEU transportation.

The c_k values for all 1,584 experiments compared to the c_k values for the ES-4100 containing 20 wt% UF_6 are shown in Figure 4. The IEU experiments are indicated with circles, and the LEU experiments are signified by diamonds. Different categories of experiments are shown with different color markers. A total of 698 experiments have a c_k value of at least 0.8, and 173 of those have a c_k of 0.9 or higher. The highest c_k values are primarily associated with both IEU and LEU solutions, although some highly applicable experiments are low enriched uranium compound (LEU-COMP-THERM or LCT) systems. Of the 698 cases with c_k values over 0.8, 635 are LEU experiments, and 154 of the 173 highly applicable experiments are LEU. The c_k values for all experiments in the suite are shown as a function of enrichment in Figure 5, and they indicate that c_k does not have a strong dependence on enrichment below approximately 30 wt% ^{235}U. Other fissile material forms and/or other packages may show different numbers of applicable experiments, but these results indicate that enough critical experiments are currently available to perform validation for packages containing HA-LEU UF_6.

The c_k results for the 5 wt% case are qualitatively similar to those of the 20 wt% case. The LCT experiments generally show significantly higher applicability, which is a logical result given that these experiments generally have enrichments at or below 5 wt% ^{235}U. A total of 1,100 experiments have a c_k value of at least 0.8, and 372 have c_k values of 0.9 or higher. These results indicate that the c_k values are not a strong function of system enrichment, further supporting the conclusion that enough applicable critical experiments are available in the ICSBEP Handbook currently to support analysis of HA-LEU systems.

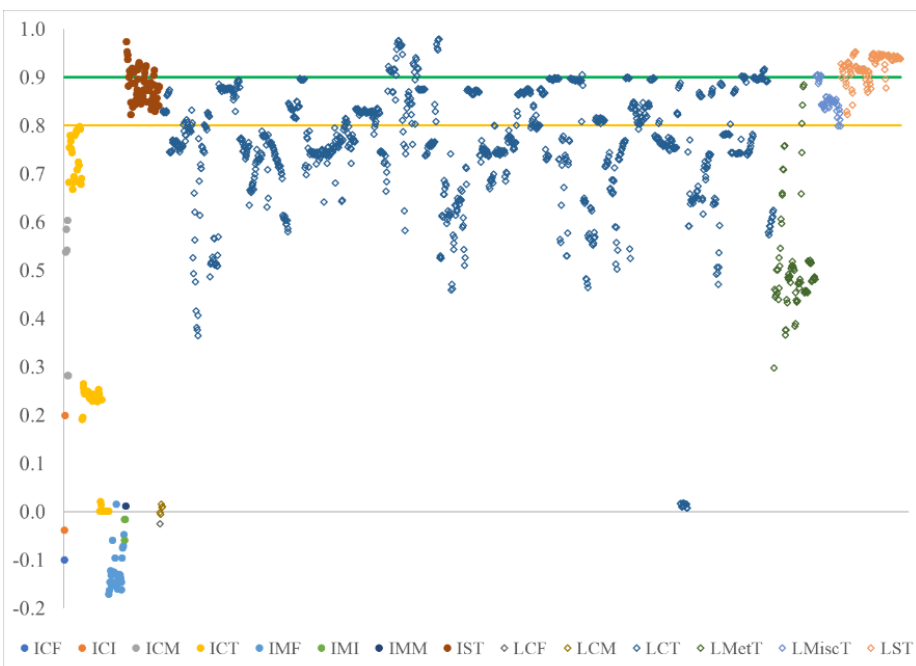

Figure 4. c_k values for 20 wt% UF_6 case.

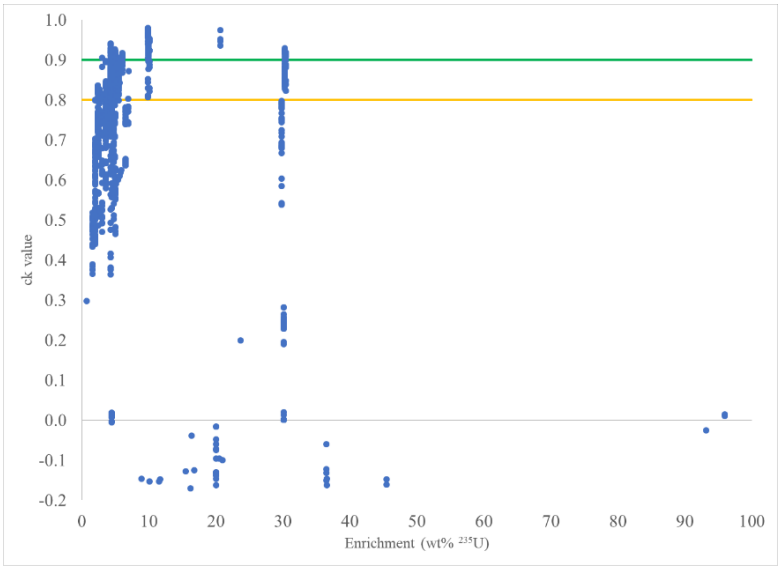

Figure 5. c_k values as a function of enrichment for ES-4100 containing 20 wt% UF_6 indicating that applicability of criticality experiments is not necessarily a strong function of enrichment.

CONCLUSIONS

This paper describes the impact of recent nuclear data changes on HA-LEU and provides a preliminary sensitivity/uncertainty–based assessment of the applicability of available of criticality experiments for validation of licensing calculations for transportation of small quantities of 20 wt% UF_6. This study showed that while recent dramatic changes in the ENDF data due to nuclear data tuning may cause unpredictable behavior in new systems, existing criticality experiments available in the ICSBEP Handbook could provide a sufficient basis for validation, especially where the sensitivity/uncertainty approaches to validation are applied.

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