# RESONANCE REGION NUCLEAR DATA ANALYSIS TO SUPPORT ADVANCED FUEL CYCLE DEVELOPMENT

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The Oak Ridge National Laboratory (ORNL) and the Korean Atomic Energy Research Institute (KAERI) are performing collaborative research as part of a three-year United States (U.S.)–Republic of Korea (ROK) International Nuclear Energy Research Initiative (I-NERI) project to provide improved neutron cross-section data with uncertainty or covariance data important for advanced fuel cycle and nuclear safeguards applications. ORNL and KAERI have initiated efforts to prepare new cross-section evaluations for $^{240}$Pu, $^{237}$Np, and the stable Cm isotopes. At the current stage of the I-NERI project, ORNL has recently completed a preliminary resonance-region cross-section evaluation with covariance data for $^{240}$Pu and has initiated resonance evaluation efforts for $^{237}$Np and $^{244}$Cm. Likewise, KAERI is performing corresponding high-energy cross-section analyses (i.e., above the resonance region) for the noted isotopes. The paper provides results pertaining to the new resonance region evaluation efforts with emphasis on the new $^{240}$Pu evaluation.

KEYWORDS : evaluation, resonance region, $^{240}$Pu, $^{237}$Np, Cm

## 1. INTRODUCTION

The three-year U.S.–(Republic of Korea (ROK) International Nuclear Energy Research Initiative (I-NERI) project is a collaborative effort involving Oak Ridge National Laboratory (ORNL) and the Korean Atomic Energy Research Institute (KAERI) to provide improved neutron cross-section data with uncertainty or covariance data for isotopes important for advanced fuel cycle (AFC) and nuclear safeguards applications. Because nuclear data are basic science data needed for all aspects of the design and deployment of AFC systems, the improved nuclear data will benefit AFC design and deployment efforts as well. In particular, an additional goal of the collaboration will be to assess uncertainties of the nuclear integral parameters due to the cross-section data, to improve safety validation, and to reduce capital cost through system design optimization for AFC developments.

KAERI has worked with ORNL to identify priority nuclides that need improved nuclear data with uncertainty information for AFC analyses and nuclear safeguards applications. Historically, nuclear data improvements have largely focused on improvements for major actinides important for AFC applications, such as $^{235}$U, $^{238}$U, and $^{239}$Pu. In contrast, there has been some effort, albeit at a lower level of focus, on data improvements for other isotopes/nuclides important for AFC and nuclear safeguards applications [e.g., other Pu isotopes (e.g., $^{240}$Pu, $^{241}$Pu, $^{242}$Pu), Am, and minor actinides]. Therefore, the ORNL–KAERI collaboration provided an opportunity to assess the status of the "minor" actinides for supporting AFC and nuclear safeguards applications. To this end, ORNL and KAERI performed a detailed data assessment of the latest cross-section evaluations for $^{240}$Pu, $^{237}$Np, and the stable Cm isotopes. Based on the data assessment, ORNL and KAERI have initiated a collaborative evaluation effort to produce new cross-section evaluations for the noted isotopes. Furthermore, the final product of this work will be improved nuclear data evaluations with covariance information that will be submitted for inclusion in the U.S. Evaluated Nuclear Data File (ENDF/B) system [1]. The paper provides the latest results from the ORNL resonance region evaluation effort.

## 2. NUCLEAR APPLICATION SIGNIFICANCE

The U.S. Fuel Cycle Research and Development (FCR&D) Nuclear Physics Working Group and Nuclear Safeguards Working Group have been investigating

nuclear data needs for supporting safeguards applications. Both working groups recognize that a science-based approach will require integrating experiment, theory, modeling, and simulation to develop technologies needed to enable next-generation safeguards for the nuclear fuel cycle in order to reduce proliferation risks. New measurement and analysis technologies will be needed to monitor and interrogate nuclear systems involving spent nuclear fuel (SNF). Measurement and analysis methods are being developed to safeguard SNF that contains significant quantities of uranium, plutonium, and neptunium isotopes. The ability to simulate the radiation transport processes involving SNF is essential for accurate safeguards analyses, and the modeling and simulation are especially dependent upon the underlying cross-section data. Simulation of particle detector counters in material interrogation for nuclear safeguards applications depends on how well the basic nuclear data and their uncertainties are known.

For safeguards applications, a suite of instruments will be required to achieve the requisite transparency of facility operations. The advanced instrumentation needs include passive neutron counting, active interrogation, ultrahigh-resolution spectroscopy, and process monitoring. Furthermore, the FCR&D working groups have noted that advanced techniques will require nuclear data improvements in a number of areas. For example, a primary candidate isotope for AFC safeguards applications is $^{237}$Np. Neptunium-237 will be coupled with plutonium primarily to enable neptunium recycle, but $^{237}$Np also has a safeguards benefit. Protactinium-233, the decay product from $^{237}$Np, emits an easily detected gamma ray that can facilitate detection and monitoring as well as impede attempts to divert material from a nuclear facility. For active interrogation techniques, detailed nuclear data in the neutron resonance region are needed for key isotopes such as $^{237}$Np, $^{244}$Cm, plutonium isotopes, americium isotopes, and other minor actinides. In addition, cross-section uncertainty or covariance data are not available in ENDF/B for $^{237}$Np or $^{244}$Cm or for many other key isotopes (e.g., $^{241}$Pu, $^{242}$Pu, $^{241}$Am). Improved nuclear data with covariance data are needed to facilitate radiation transport modeling to support nuclear fuel cycle analyses, including safeguards analyses of SNF (e.g., active interrogation). In addition, reprocessed fuel will contain significant quantities of isotopes that spontaneously fission (e.g., $^{240}$Pu and $^{242}$Pu), and these plutonium isotopes do not have cross-section covariance data. In addition to the need for neutron cross-section data, there is a need for accurate gamma-ray energies and branching ratio data that are important for isotopic analyses for AFC materials. Based on the noted safeguards application needs, $^{237}$Np and $^{240}$Pu have been identified as two actinides that need significant improvements to support AFC and nuclear safeguards applications.

With regard to Korean AFC application needs, the share of electricity generation produced by nuclear power plants in Korea is currently more than 42%, and this share will increase in the next several decades because the Korean government plans to construct more than 10 nuclear power plants by 2030. The accumulating SNF from pressurized-water reactor (PWR) operation is an imminent problem in Korea. Likewise, the management of SNF in the United States is an important issue, and both countries have keen interest in efficiently managing SNF. In Korea, it is expected that the spent fuel storage racks in the Korea PWR sites will be fully occupied by 2016. Research for SNF reprocessing has been carried out with a proliferation-resistant nuclear fuel cycle in Korea. Transmutation by an accelerator-driven system (ADS) or by an advanced burner reactor (ABR) is considered to be a viable method for disposal of the high-level radioactive waste. The fuel for the transmutation burner can be manufactured to mixed-oxide powder or metallic alloy through the reprocessing of LWR spent fuel. A minor-actinide–bearing U-Pu-Zr metallic alloy is referenced to the fuel for transmutation burner for high proliferation resistance in Korea. Based on the Korean AFC application needs, accurate neutron cross-section data are essential to the reliable prediction of the behavior of a nuclear system. Moreover, the Korean nuclear data needs encompass both major and minor actinides as well as key fission products that will be present in an advanced nuclear fuel cycle.

Based on the perceived nuclear application needs in both countries, both KAERI and ORNL determined that nuclear data improvements for $^{240}$Pu, $^{237}$Np, and the stable Cm isotopes would benefit AFC and nuclear safeguards applications.

## 3. NUCLEAR DATA ASSESSMENT OF LATEST EVALUATIONS

During the first year of the collaborative project, ORNL and KAERI have performed a detailed assessment and have initiated preliminary evaluation work on $^{240}$Pu, $^{237}$Np, and the stable Cm isotopes. ORNL has performed preliminary cross-section analyses (including covariance analysis) for the resonance energy region, and KAERI has performed complementary analyses at energies above the resonance region. To test the improved cross-section data including covariance data, appropriate benchmarks have been collected and reviewed. The validation of the improved cross-section data and the sensitivity/uncertainty assessments for nuclear integral parameters using new

covariance data will be carried out with various benchmarks.

As part of the data assessment effort, intercomparisons of the latest cross-section evaluations from the ENDF/B library, European Joint Evaluated Fission and Fusion (JEFF) Library, and Japanese Evaluated Nuclear Data Library (JENDL) have been performed (i.e., ENDF/B-VII.0 [1], JEFF-3.1.1 [2], and JENDL-3.3 [3], respectively). As part of the assessment, the latest evaluations have been compared with experimental data where possible. A survey of the available covariance data for the noted isotopes has been performed to identify the current status of the uncertainty information in the cross-section database. The following is a brief assessment summary of the latest evaluations for $^{240}$Pu, $^{237}$Np, and $^{244}$Cm provided in the ENDF/B-VII.0, JEFF-3.1.1, and JENDL-3.3 nuclear data libraries.

### 3.1 $^{240}$Pu Cross-Section Data

Prior to this work, the $^{240}$Pu cross-section data of JEFF-3.1.1 have been most recently evaluated by Bouland, et al. [4]. The resonance parameters were produced with a SAMMY [5] analysis of neutron transmission data measured at the Geel Electron Linear Accelerator (GELINA) by Kolar et al. [6], and the fission cross section was measured at the Oak Ridge Electron Linear Accelerator (ORELA) by Weston et al. [7]. The reaction formalism used in SAMMY is the Reich-Moore formalism, and the analysis was performed in the thermal to 5.7 keV neutron energy range. All the resonances identified in the experimental data were assumed to be s-wave resonances. The covariance matrices of the resonance parameters and cross sections were not given in the previous evaluation. In the Derrien-Bouland evaluation, resonance parameters from 0 eV to 5.7 keV were obtained. The neutron capture was normalized down by 27% compared to previous evaluations that are based in most part on the experiment from Weston. The Derrien-Bouland resonance parameters were adopted in the JEFF library. Also, the Derrien-Bouland data were adopted in the JENDL library but only up to 2.7 keV because the capture cross-section values were too small. In contrast to JEFF 3.1.1 and JENDL 3.3, the ENDF/B-VII.0 evaluation of the resolved resonance region is not the latest resonance evaluation for $^{240}$Pu. The ENDF/B-VII.0 evaluation is based on several measurements by Weston, Spencer, Wisshak, Auchampaugh, Weigman, Kolar, and Block. Due to the large number of experimental data references, the entire set of measurement references are not provided in the paper, and the reader is referred to the ENDF/B-VII.0 evaluation, which provides the complete set of references for the experimental data. Many of the cross-section data used in the ENDF/B-VII.0 evaluation were normalized to the thermal neutron energy region (i.e., Weston capture data).

The thermal elastic cross-section value of ENDF/B-VII.0 is 0.95 barns; it is 2.67 barns in JEFF-3.1.1 and JENDL-3.3. The elastic resonance integrals of ENDF/B-VII.0 are more than 5% smaller than JEFF-3.1.1 and JENDL-3.3. The fission resonance integrals of ENDF/B-VII.0 are about 20% smaller than the other evaluations. Based on comparisons between the experimental cross-section database and the evaluated data, the capture cross sections above 100 keV show large discrepancies among the ENDF/B-VII.0, JEFF-3.1.1, and JENDL-3.3 nuclear data libraries. The fission cross-section data above 10 keV of JEFF-3.1.1 show a discrepancy with ENDF/B-VII.0 and JENDL-3.3. With regard to uncertainty information, the $^{240}$Pu covariance data are included in JENDL-3.3; however, covariance data are not available in the latest ENDF/B or JEFF file systems.

### 3.2 $^{237}$Np Cross-Section Data

The $^{237}$Np fission cross-section data, elastic cross-section data, and delayed neutron production data per fission event of ENDF/B-VII.0 were most recently evaluated by Young, et al. in November, 2004 [1]. The resonance parameters of ENDF/B-VII.0 fission data were taken from JENDL-3.3. The background cross sections for total, elastic, fission, and capture cross sections in the resolved resonance range were deleted. In the unresolved resonance region, the fission width was slightly modified in the 10–35 keV energy range to reproduce the measurements taken at the Los Alamos Neutron Science Center (LANSCE) for the ratio $^{237}$Np/$^{235}$U fission data by Tovesson and Hill [8,9]. The LANSCE data were also used to reanalyze cross sections in the energy ranges between 35 keV and 2 MeV and from 15 to 20 MeV. In the ENDF/B-VII.0 evaluation, the multilevel Breit Wigner (MLBW) resonance formalism is used up to 35 keV. The MLBW resonance formalism is not sufficient to represent the detailed resonance structure that includes resonance-resonance interference effects. As a result, the current $^{237}$Np evaluation may not adequately represent the detailed resonance structure needed to support AFC application needs.

In contrast to the ENDF/B-VII.0 evaluation, the JEF-3.1.1 data were taken from ENDF/B-VI.4 and show large discrepancies in fission and elastic resonance cross sections from ENDF/B-VII.0 and JENDL-3.3 data. The thermal capture cross section of JEF-3.1.1 is about 10% larger than ENDF/B-VII.0 and JENDL-3.3 data; the elastic resonance integrals of JEFF-3.1.1 are about 10% smaller than ENDF/B-VII.0 and JENDL-3.3 data. The fission resonance integrals of JEFF-3.1.1 are one-third those of ENDF/B-VII.0 and JENDL-3.3, although the absolute

fission resonance integrals are very small. The capture cross sections around 1 MeV of JEFF-3.1.1 are slightly larger than the ENDF/B-VII.0 and JENDL-3.3 data. The fission cross-section data in the resonance energy region of JEFF-3.1.1 are smaller than the ENDF/B-VII.0 and JENDL-3.3 data. With regard to uncertainty information, $^{237}$Np covariance data are available from the JENDL-3.3 covariance file; however, covariance data are not available in the latest ENDF/B or JEFF file systems.

### 3.3 $^{244}$Cm Cross-Section Data

The $^{244}$Cm cross-section data of ENDF/B-VII.0 are taken from JENDL-3.3 and were evaluated by T. Nakagawa and T. Liu in 1995 [3]. The JEFF-3.1.1 data have been evaluated by Mann et al. in 1978 [2]. The thermal cross sections of JEFF-3.1.1 are more than 30% smaller than JENDL-3.3. The elastic and capture resonance integrals of JEFF-3.1.1 are ~10% smaller than those of JENDL-3.3. However, the fission resonance integral of JEFF-3.1.1 is ~80% larger than that of JENDL-3.3. Relative to the experimental database, the evaluated cross-section data at all energy ranges exhibit large differences between JENDL-3.3 and JEFF-3.1.1. Regarding uncertainty information, $^{244}$Cm covariance data are available from the JENDL-3.3 covariance file; however, covariance data are not available in the latest ENDF/B or JEFF file systems.

## 4. RESONANCE EVALUATION EFFORT

At this stage of the cross-section evaluation effort for the resonance region, ORNL has completed a preliminary resonance evaluation for $^{240}$Pu with covariance data. In addition, ORNL has prepared a preliminary covariance evaluation for $^{244}$Cm. With regard to $^{237}$Np, ORNL has recently initiated work on a new resonance evaluation for $^{237}$Np that will include new experimental data from LANSCE and the Conseil Européen pour la Recherche Nucléaire (CERN). Because the $^{237}$Np evaluation effort is in the beginning stage, only results from the $^{240}$Pu and $^{244}$Cm evaluation effort are presented in the paper.

### 4.1 $^{240}$Pu Evaluation

The primary objective of the present evaluation is to calculate the Resonance Parameter Covariance Matrices (RPCMs) and the Cross Section Covariance Matrices (CSCMs) needed for the determination of the uncertainties in the group cross sections in the thermal to 5.7 keV energy range.

In the present evaluation new experimental transmission data were added to the experimental database. These new data are the neutron transmission measurements performed at ORELA in 1988 by Harvey and Gwin [10] and which were recently discovered in the ORELA archives. The transmission data of Kolar et al. were obtained with sample thicknesses of 0.00166 at/b and 0.00466 at/b. The sample thickness used by Harvey and Gwin (0.0723 at/b) is 15 times thicker than the thickest sample used by Kolar et al. The experimental fission data were not included. It is assumed that the fission widths, generally very small, were obtained with enough accuracy in the previous evaluation. Preliminary SAMMY analyses of the data allowed the estimation of normalization corrections and of background adjustments. The neutron energies of all the measurements were aligned on the energy of the ORELA data. A consistent experimental database has been obtained by applying appropriate corrections to the data. This consistent database is suitable for a sequential SAMMY Bayes analysis that allows the determination of a unique set of resonance parameters.

The thickness of the samples used by Kolar et al. was too small to allow the identification of the small s-wave resonances and of the p-wave resonances, particularly above the neutron energy of 1 keV. The thick sample of Harvey-Gwin allowed the identification of the small s-wave resonances and of an important number of p-wave resonances. In the previous evaluation, based on the Kolar data, 425 resonances were identified, and all of them are assumed to be s-wave resonances. In the present evaluation, 428 resonances are assumed to be s-wave resonances, and 121 p-wave resonances have been identified. The Harvey-Gwin sample contained a small amount of other plutonium isotopes. The experimental transmission data have been corrected for the contribution of the contaminants by a SAMMY calculation using the ENDF/B-VII.0 resonance parameters, in order to avoid the confusion between small $^{240}$Pu resonances and resonances of the contaminants.

The values of the neutron widths obtained in the present evaluation are very close to those obtained in the previous evaluation in the energy range below about 700 eV. At higher energy there is a tendency to obtain smaller values that could be explained by the reevaluation of the experimental resolution of Kolar data, particularly the exponential tail of the resolution. The total cross-section value at 0.0253 eV, calculated with the new resonance parameters, is 284.05 b and is in agreement with the Spencer experimental data [11] but is significantly smaller than the value of 289 b recommended by the *Atlas of Neutron Resonance Parameters* [12]. Consequently, the capture cross-section value at 0.0253 eV is also smaller than the recommended value provided by the *Atlas of Neutron Resonance Parameters*. The capture resonance

integrals calculated with the PUFF-IV [13] covariance processing software in the energy range 0.5 eV to 7.7 keV are 8479 b, 8480 b, and 8492 b for the present evaluation, JEFF-3.1.1, and ENDF/B-VII.0, respectively.

The resonance parameters and the corresponding RPCMs were converted to the ENDF/B format. A new evaluation file was obtained by replacing the ENDF/B-VII.0 resonance parameters with the new resonance parameters and by adding the RPCM. An evaluation of the CSCM to complement the RPCM is currently in progress. The new $^{240}$Pu RPCM has been processed with PUFF-IV, and an example of the $^{240}$Pu neutron capture correlation matrix for the resonance parameters is provided in Figure 1. The RPCM in Figure 1 only includes the statistical uncertainty resulting from the SAMMY analysis of the resonance parameters. When the CSCM is completed, the overall uncertainty at thermal energies will increase.

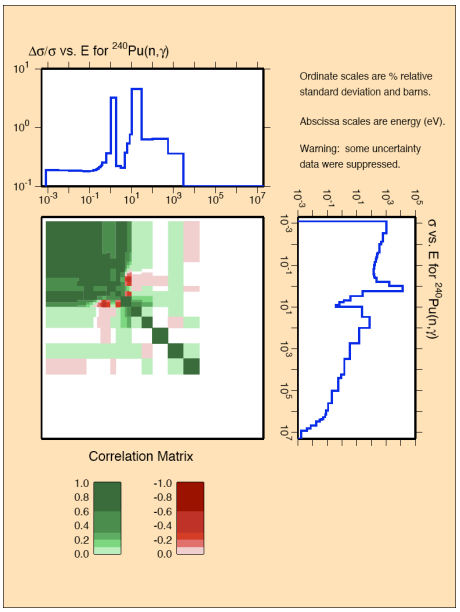


**Fig. 1.** $^{240}$Pu resonance parameter covariance data for neutron capture.

## 4.2 $^{244}$Cm Evaluation

The objective of the $^{244}$Cm evaluation effort is to provide resonance parameter covariance data while preserving the existing resonance parameter data in File 2 of the cross-section evaluation. Moreover, the resonance parameter covariance evaluation will be used to complement the high-energy cross-section and covariance analysis work being performed by KAERI for the Cm isotopes. In the ORNL work, the RPCMs for the resolved region were generated for $^{244}$Cm using the SAMMY R-Matrix software in the energy range from $10^{-5}$ eV to 1 keV. The retroactive approach [14] of the SAMMY software was used to generate the RPCM for $^{244}$Cm. The retroactive covariance analysis approach can be used to preserve the existing File 2 resonance parameters while providing an estimate for the RPCMs. In the current work, the RPCMs were generated based on the resonance parameters of the JENDL 3.3 evaluation. The uncertainties on the thermal cross section and capture resonance integral provided in the *Atlas of Neutron Resonance Parameters* were used in the RPCM analysis. Using the retroactive approach, the SAMMY software has been used to produce $^{244}$Cm RPCM data in the ENDF/B File 32 RPCM format. Subsequently, the PUFF-IV covariance processing software was used to process the $^{244}$Cm RPCM data. To demonstrate the results, Figure 2 provides the correlation matrix for $^{244}$Cm elastic scattering.

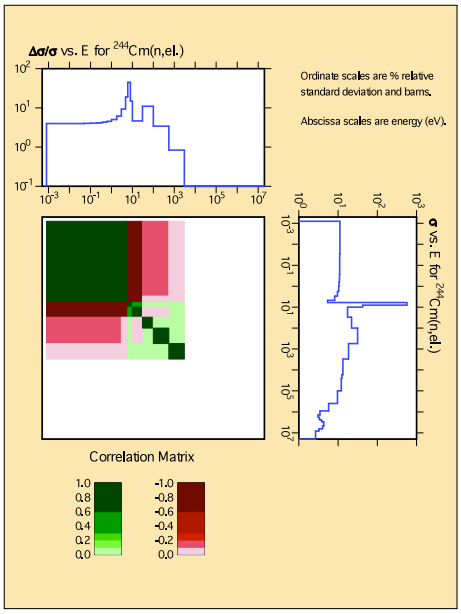


**Fig. 2.** $^{244}$Cm resonance parameter covariance data for elastic scattering.

## 5. SUMMARY

ORNL and KAERI are in the second year of a three-year U.S.–ROK I-NERI project to provide improved neutron cross-section data with uncertainty or covariance data for isotopes important for AFC and nuclear safeguards applications. To this end, ORNL and KAERI have identified and performed a detailed data assessment for $^{240}$Pu, $^{237}$Np, and the stable Cm isotopes that are priority

isotopes needing improved nuclear data with uncertainty information for AFC safeguards and design applications. Based on the data assessment, ORNL and KAERI have initiated a collaborative evaluation effort to produce improved nuclear data evaluations for the noted isotopes. This paper provides the current status of the I-NERI evaluation effort. Specifically, ORNL has produced a preliminary resonance evaluation with covariance data for $^{240}$Pu and a resonance parameter covariance evaluation for $^{244}$Cm. Efforts are in progress to produce a new resonance evaluation for $^{237}$Np. The final product of the ORNL/KAERI collaboration will be improved and tested nuclear data files with uncertainty information that will be submitted for inclusion in the U.S. ENDF/B system.

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