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REVIEW OF MONTE CARLO NEUTRONICS ISOTOPIC VALIDATION STUDIES ON NUCLEAR FUEL DEPLETION

Sean P. Martinson[1] and Sunil S. Chirayath[1,2]

Department of Nuclear Engineering, Texas A&M University, College Station, TX 77843-3473
Center for Nuclear Security Science & Policy Initiatives, Texas A&M University, College Station, TX 77843-3473 [1]smartinson91@tamu.edu [2]sunilsc@tamu.edu

ABSTRACT

Monte Carlo (MC) neutronics codes are popular tools that have been used in academic and industrial applications for over three decades. MC neutronics codes are often coupled with depletion solvers/codes to perform nuclear fuel depletion simulations. MC simulations can model complex nuclear power cores to simple theoretical infinite slab geometries. Within these models, the code may calculate neutron multiplication values or isotope production in nuclear reactor fuel. As is the case with any code simulating reality, benchmark studies must be conducted to test for accuracy. MC codes are commonly validated or benchmarked through criticality experimental results from reactors, but isotopic validation studies are performed less frequently because of the costs. Isotopic validation studies require chemical processing of depleted nuclear fuel and/or material which in turn requires special tools and expertise that may not be widely available. Therefore, several studies have published isotopic inventories from depleted nuclear material for anyone to use for their own validation studies. This work gathers and examines fuel depletion benchmarks and validation studies used to support the accuracy and operation of MC-based fuel depletion neutronics codes. It is imperative to understand strengths and limitations for the integrity of MC codes. MC codes have been shown to accurately predict (< 10% RE) nuclides such as ^{235}U, ^{236}U, ^{238}U, ^{241}Am, ^{243}Am, ^{99}Tc, ^{101}Ru, ^{103}Rh, ^{133}Cs, ^{134}Cs, and ^{137}Cs. Conversely, MC codes have shown difficulty in predicting (< 20% RE) ^{237}Np, ^{242}Cm, ^{155}Gd, ^{125}Sb, ^{140}Ce, and ^{142}Ce. Measuring and predicting these isotopes have practical applications outside of validation studies, such as international safeguards. This work demonstrates strong and weak MC code-derived accuracies of signature isotopes that have implications in nuclear forensics and in nuclear material accountancy.

INTRODUCTION

Nuclear reactors are technological marvels that have helped shaped the latter half of the 21st century. To optimize their designs, innumerable calculations, algorithms, and codes have been developed. Neutronic computer codes are examples of such a development. These codes are used to perform nuclear reactor core modeling, neutron transport calculations, dose and shielding calculations, and isotope production, to name a few. Several popular neutronics codes are built based on Monte Carlo (MC) statistics. These MC codes include Monte Carlo N-Particle (MCNP) [1], Monteburns [2], Serpent [3], etc. The MC neutronics code randomly sample from the probability density functions of various physical process contained in the Boltzmann radiation transport equation. A random sampling scheme is used to simulate the key parameters of a neutron as it travels through and interacts with materials. The key parameters are position, velocity, energy, interaction type, and history. The MC codes have the advantage over other codes, such as lattice-based, by their capacity to model complex geometries and account for neutron energy spectra

variations across reactor cores. The major disadvantage is that MC codes can require high computational resources and be time consuming.

For the MC codes to be fully effective, they must perform well against experimental results. This is accomplished via benchmark and validation studies. In the scope of this review a benchmark uses historical data, typically from a database. While a validation study uses data gathered by the researchers themselves, mostly using experiments. There may be overlaps between these two types, benchmarks and validation studies. Benchmarks and validation studies often measure criticality, though, this review will focus on fission product and actinide isotopic generation measurements.

On their own, MC codes do not calculate fuel depletion or isotope generation. They must be coupled with depletion codes/solvers, which are usually built around the Bateman equations[4]. The two most common depletion codes are CINDER90 and ORIGEN2. MCNPX, and its successor MCNP6, have CINDER90 as their default solver. While ORIGEN2 is commonly coupled with Monteburns and OpenMC.

Experimental measurements can be broken into two analyses: destructive assay (DA) and non-destructive assay (NDA). DA analysis involves the altering of a sample's physical and/or chemical form, such as in the case of a mass spectrometry. Conversely, samples undergoing NDA retain their physical and chemical form. Gamma spectrometry is a type of NDA.

In 2018, Osborn et al. from Texas A&M University developed a nuclear forensics methodology capable of calculating three key parameters (reactor type, burnup, and time since irradiation) of interdicted special nuclear material [5] The methodology, dubbed the Maximum Likelihood method, is built on density functions developed by sampling millions of data points generated through MCNP reactor simulations. The utility of the method is linked to MCNP's ability to accurately calculate isotope generation values. Therefore, methods such as this demonstrate the importance of validated and benchmarked MC codes to nuclear forensics.

BENCHMARK AND VALIDATION STUDIES

In 2012, Čerba et al. published a study of benchmarking MCNPX [6]. The historical data they used was the from the Organisation for Economic Co-operation and Development Nuclear Energy Agency (OECD NEA) Burnup Credit Benchmark. The IV-B phase of the benchmark examined MOX fuel in light water reactors. For the neutronics, Čerba et al. used MCNPX2.7, which uses CINDER90. For the nuclear data library Čerba et al. used three: ENDF/B-VII, JEFF3.1.1, and JENDL4.0.

Čerba et al. calculated the isotopic generation values for 31 nuclides, 16 of which were actinides, and the rest were fission products. They provided relative error (RE) values on bar graphs, which are summarized in Table 1. Between the three data libraries, they all performed well, with 14 out of 31 nuclides having RE values less than 5%. Only four nuclides had poor RE (greater than 20%). The ENDF data library performed the best overall. The authors attribute the discrepancies of the poorly estimated nuclides to inaccuracies in the code or variations in the cross-section evaluation. On the other hand, the results provide strong support for MCNPX's ability to calculate isotope production values.

Table 1. The relative error (RE) values for poor and excellent isotopic estimations [6].

Poor Estimates (>20% RE)	Excellent Estimates (<5% RE)
^{237}Np	$^{235, 236, 238}$U
^{242}Cm	$^{238\text{-}242}$Pu
^{149}Sm	$^{241, 243}$Am
^{155}Gd	^{99}Tc
	^{101}Ru
	^{103}Rh
	^{133}Cs

The next study published by Sternbentz et al. in 2015 is a validation study [7]. The Advanced Gas Reactor (AGR) project was sponsored by the U.S. Department of Energy in collaboration with Idaho National Laboratory (INL) and Oak Ridge National Laboratory (ORNL). The goal of the project was to develop, manufacture, and study tri-structural isotropic (TRISO) nuclear fuels. The TRISO fuels contained LEU UCO kernels. Approximately 4100 TRISO microparticles make up a single fuel cylindrical fuel compact similar in size to traditional LWR fuel pellets. A total of 72 fuel compacts were studied in the Advanced Test Reactor at INL, then they performed a post-irradiation examination (PIE) using gamma spectrometry and mass spectrometry, specifically, inductively coupled plasma mass spectrometry (ICP-MS).

Sternbentz et al. employed JMOCUP, which used MCNP5 for radiation transport and ORIGEN2.2 for depletion calculation, to simulate the compacts' irradiation history. For nuclear data, they used the ENDF/B-VII.1 library. Table 2 shows Calculated/Experimental (C/E) activity ratios for six fission products that were measured experimentally by gamma spectrometry. The two major deviations are from ^{154}Eu and ^{125}Sb, which the authors attribute to "slight measurement or calculation bias," meaning they were overpredicted by the code. Table 3 shows 14 fission products and actinides that were measured experimentally via ICP-MS. Five of the nuclides were overestimated by over 20%, four of which were plutonium isotopes. The authors attributed the difference to either code over-calculations or incomplete sample recovery from dissolution.

Table 2. Activity ratios for isotopes measured via gamma spectrometry [7].

Nuclide	Activity Ratios (C/E)
^{137}Cs	1.0013±0.0319
^{134}Cs	1.0190±0.0506
^{144}Ce (^{144}Pr)	0.9939±0.0367
^{154}Eu	1.1835±0.0525
^{106}Ru	1.0404±0.0432
^{125}Sb	1.3955±0.0860

Glennon et al.'s work can be characterized as a validation study. They measured quantities and ratios of samarium (Sm) isotopes from irradiated nuclear material [8]. UO_2 samples were irradiated in two reactors, High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory and the University of Missouri Research Reactor (MURR). The irradiations differed in enrichment, burnup, and neutron spectra. The irradiated samples were then chemically separated and analyzed

by liquid column chromatography and ICP-MS, respectively. Glennon et al. reported various Sm isotopes as functions of simulated (S) and experimental (E) values. The simulated values were generated with MCNP6, which used CINDER90 for depletion calculations, and ENDF/B-VII.0 as the nuclear data source.

Table 3. C/E averaged values for isotopes measured via ICP-MS [7].

Isotope	Average C/E
^{234}U	1.03
^{235}U	0.98
^{236}U	1.08
^{238}U	1.08
^{239}Pu	1.29
^{240}Pu	1.26
^{241}Pu	1.22
^{242}Pu	1.23
^{139}La	0.95
^{140}Ce	1.27
^{141}Pr	0.90
^{142}Ce	1.28
^{145}Nd	0.89
^{146}Nd	1.08

Glennon et al. reported 21 and two ratios for the HFIR and MURR samples, respectively. Out of 21 HFIR ratios, only two had RE greater than 20% between the S and E values, while nine had RE of 10% or less as shown in Table 4. The two poorest predictions were ^{148}Sm/^{151}Sm and ^{151}Sm/^{152}Sm. Further, all ratios containing ^{151}Sm have differences greater than 13%. The authors state that a possible source of discrepancy may be from isobaric interference, specifically ^{151}Eu, which would affect ICP-MS measurements. For the MURR ratios, ^{150}Sm/^{149}Sm and ^{152}Sm/^{149}Sm, the authors reported RE values of 1.6% and 6.4%, respectively.

Table 4. Poor and excellent RE values from HFIR samples [8].

Poor Ratio Estimates (>20% RE)	Excellent Ratio Estimates (<5% RE)
^{148}Sm/^{151}Sm	^{150}Sm/^{149}Sm
^{151}Sm/^{152}Sm	^{147}Sm/^{149}Sm
	^{149}Sm/^{154}Sm
	^{147}Sm/^{154}Sm
	^{150}Sm/^{154}Sm

ANALYSIS

MC neutronics codes are powerful tools that have been tested and used globally. In the studies discussed here, MC codes have performed well at predicting many fission products and actinides. A few of the isotopes which were well predicted with RE values of 10% or less include ^{235}U, ^{236}U, ^{238}U, ^{241}Am, ^{243}Am, ^{99}Tc, ^{101}Ru, ^{103}Rh, ^{133}Cs, ^{134}Cs, and ^{137}Cs. Conversely, there were several isotopes that were poorly predicted with RE values above 20%, such as ^{237}Np, ^{242}Cm, ^{155}Gd, ^{125}Sb, ^{140}Ce, and ^{142}Ce. For ^{125}Sb, discrepancy may come from ^{125}Sb from numerous production/destruction pathways, which have uncertainties that combine. For trans-uranic isotopes, the main source of uncertainty may be low concentrations from low-burnup conditions. Thus, it may be expected that trans-uranic isotopes may better predicted from higher burnup environments. Another source of deviation may be from the cross-section library used with the MC codes. Only Čerba et al. used data libraries outside of ENDF, but ENDF did not out-perform the other libraries across all isotopes. Therefore, there may be benefit from using multiple data libraries, rather than a single one. It is important to note that data libraries are continually updated, which will only improve isotope generation calculations. Finally, there were also experimental or systematic errors in the studies. Therefore, the predictions may have been good, but could not be experimentally verified. To address this concern, researchers could repeat experiments and avoid previous mistakes.

CONCLUSION

The MC neutronics codes are excellent resources in simulating and understanding nuclear systems. MC codes allow users to calculate criticality and isotope generation with high accuracy. Some isotopes (^{235}U, ^{236}U, ^{238}U, ^{241}Am, ^{243}Am, ^{99}Tc, ^{101}Ru, ^{103}Rh, ^{133}Cs, ^{134}Cs, and ^{137}Cs) can be predicted with less than 10% RE. While other isotopes (^{237}Np, ^{242}Cm, ^{155}Gd, ^{125}Sb, ^{140}Ce, and ^{142}Ce) should be considered with caution. Uncertainties for poorly predicted isotopes may come from multiple sources that need to be properly addressed to ensure better accuracy. Good simulation accuracy is paramount to nuclear forensic and nonproliferation applications, such as the Maximum Likelihood method from Texas A&M University.

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REFERENCES

[1] Werner, C.J. (editor), 2017. MCNP® User's Manual: Code Version 6.2, Los Alamos National Laboratory Report No. LA-UR-17-29981.

[2] Poston, D.I, and Trellue, H.R.,1999. Development of a Fully-Automated Monte Carlo Burnup Code Monteburns, Los Alamos National Laboratory Report No. LA-UR-99-42.

[3] Rearden, B.T. and Jessee, M.A. (editors), 2018. SCALE Code System Version 6.2.3, Oak Ridge National Laboratory Report No. ORNL/TM-2005/39.

[4] Bateman, H., 1910. The solution of a system of differential equations occurring in the theory of radiaoctive transformations, Mathematical Proc. of the Cambridge Philos. Soc., 15, 423-427.

[5] Osborn, J.M., Kitcher, E.D., Burns, J.D., Folden III, C.M., and Chirayath, S.S., 2018. "Nuclear Forensics Methodology for Reactor-Type Attribution of Chemically Separated Plutonium, Nuclear Technology," 201:1, 1-10.

[6] Cerba, S., Hrncir, M., & Necas, V., 2012. Verification of the depletion capabilities of the MCNPX code on an LWR MOX fuel assembly. ICEEE 2012.

[7] Sterbentz, J. W., Harp, J. M., Demkowicz, P. A., Hawkes, G. L., & Chang, G. S., 2015. Validation of the Physics Analysis used to characterize the AGR-1 TRISO Fuel Irradiation Test (No. INL/CON-14-33982). Idaho National Lab (INL), Idaho Falls, ID (United States).

[8] Glennon, K. J., Osborn, J. M., Burns, J. D., Kitcher, E. D., Chirayath, S. S., & Folden, C. M., 2019. Measuring key Sm isotope ratios in irradiated UO 2 for use in plutonium discrimination nuclear forensics. Journal of Radioanalytical and Nuclear Chemistry, 320(2), 405-414.