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Monte Carlo and deterministic simulations of activation ratio experiments for ^{238}U(n,f), ^{238}U(n,g) and ^{238}U(n,2n) in the Big Ten benchmark critical assembly

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Introduction

In 2008, the Nuclear Data Group at Lawrence Livermore National Laboratory (LLNL) released a new Evaluated Nuclear Data Library, ENDL2008.[1] Conjointly, in recent years, there has been a concerted effort to build and expand a test suite of benchmark experiments to test data evaluations in cross-section libraries needed by particle transport codes. Several integral measurements were measured in "classic" criticality safety benchmark assemblies such as Godiva, Jezebel, Big Ten, Flatop-25 and Flatop_Pu, where the k_{eff} measurements are probably the most widely used for validation purposes.[2] Central fission ratios and activation ratios were also measured in these assemblies for several neutron reactions, and that data gives additional information on the behavior of specific cross-sections. The two main data sets are found in the Cross Section Evaluation Working Group (CSEWG) specifications and from the Chemical Science and Technology Division at Los Alamos National Laboratory (CST-LANL).

Almost ten years ago, Frankle and Briemeister published extensive comparisons between central-fission and activation ratio measurements, and MCNP simulations run with several cross-section libraries. Recently, MCNP5 simulations performed with ENDF.B-VII.0 were compared to previously unpublished LANL measurements.[3][4][5] We are not aware of peer-reviewed reports describing the experiments or radiochemical analysis. Nonetheless, they allow a preliminary estimation of the quality of several cross-sections of interests.

This report describes the first Mercury and Amtran simulations of the central-fission ratio and activation ratios in the Big Ten assembly for three reactions: ^{238}U(n,f), ^{238}U(n,2n), and ^{238}U (n,g).

Experimental Data

There are two available sets of data describing the BigTen activation experiments – those reported in private communications by CST-LANL and a compilation published through CSEWG. Presumably this is a re-analysis of the early CST-LANL data. There are some modest discrepancies between the two sets. We will use data reported through CSEWG when making comparisons against calculations, except for ^{238}U(n,2n), which is only reported through CST-LANL. Table 2 gives a summary of the different experimental data.

Simulations

The simulations presented here were done with two Monte Carlo codes, MCNP5 and Mercury, and a deterministic Sn code, Amtran. MCNP5 ran with the ENDF.B-VII.0 data library while Mercury and Amtran ran with ENDL2008.1 [5][6][7][8] Note that the cross-sections for Z=92 are the same in both data libraries. The simulations ran as k-eigenvalue problems with reaction rate tallies. Finally, for debug purposes, a similar model was developed for COG but the COG results were obtained using the ENDF.B-VIr7 cross-section library.[9]

Big Ten benchmark Model

The specifications for the Big Ten benchmark assembly are defined in the International Handbook of Evaluated Criticality Safety Benchmark Experiments.[2] It is an unmoderated Intermediate Enrichment Uranium assembly (IEU_MET_FAST_007). Our model consists of two homogenized zones: a core of uranium with approximately 10% ^{235}U, surrounded by a reflector made of depleted uranium. A 1D-spherical model was developed by Scott McKinley. The 2D cylindrical model is described in reference 1 and has a benchmark k_{eff} of 0.9948±0.0013.

Monte Carlo Simulations

Following the approach described by Frankle et al., the flux is tallied in a 1cm-radius sphere located at the center of the critical assembly, the code then calculates the desired reaction rates based on the simulated flux.[3] Finally, to obtain the fission and activation ratios, these reaction rates are normalized by the reaction rate for $^{235}U(n,f)$ in the same assembly. The codes used the continuous-energy representation of the cross-sections.

Reaction Rate Tally

COG and MCNP5 have similar implementations of a reaction rate tally. The user needs to define a theoretical material made solely of the isotope of interest to define a standard reaction rate tally and to specify the reaction on the selected isotope. MCNP5 uses a standard F4 tally modified by an FM card, and COG uses a standard tally modified by a reaction rate detector response function. MCNP5 gives the reaction rate in the central region in [# reactions], while in COG, the units for the reaction rate is given in [#reactions/cm^3 of isotope].

The approach implemented in Mercury differs significantly and since the code is in development, it might change in the future. The code is set-up to calculate material depletion rather than standard reaction rates. Several parameters must be changed in the Control, the Material and the Standard Tally sections. In the "Control" section, Material_depletion must be set to analog or expected_value. In the "Material" section, for the material studied, the reaction_list of the isotope of interest must be set to ALL instead of NONE. In addition, all species resulting from possible interactions between a neutron and this isotope must be included in the list, and their atom fraction set to 0.0. The standard tallies include a material_depletion tally that needs to be ON. An example of the modified input deck is given in figure 1. To collect the flux/material depletion in a given region of the core, one must define a second material assigned uniquely to the region of interest, with identical composition to the rest of the assembly core. Otherwise, since results are reported "per material", the reaction rate will be given for the whole core. In a given material, the code calculates the total mass of isotope (in grams) lost or gained for each reaction. These results must then be normalized by the isotopic concentration in the material.

Sn code: Amtran

A new capability was implemented in Amtran to calculate reaction rates on both a per atom basis for a specified isotope as well as a per atom basis for the local mixture at

the user specified location.[8] The user needs to define the isotope(s) and list of reaction(s) of interest at one or more locations.. 1D calculations were run with an S24 quadrature set and 2D calculations used an S12 quadrature set. Both calculations used an 87 group energy representation of the cross sections with a P2 representation of the scattering kernel. Amtran uses spatial AMR and the zoning at the finest level was set to 1 mm, but the only area that was dezoned was the air surrounding the device. An example of the modified input deck is given figure 2.

Results

The central fission ratio and activation ratios in the Big Ten critical assembly were first simulated with MCNP5 and COG to establish a baseline, then they were simulated with Mercury and Amtran. The results were compared to CST-LANL data for ^{238}U(n,f), (n,g) and (n,2n), and to CSEWG experimental data with uncertainties for ^{238}U(n,f), and ^{238}U (n,g).

COG and MCNP5 results for ^{238}U(n,f), (n,g) and (n,2n) of the ENDF.BVIr7 data library are compared to experimental ratios in table 1. Even for a fairly small number of cycles and histories, MCNP5 and COG results are fairly consistent, they differ by 5.4% for ^{238}U(n,f), 2.4% for ^{238}U(n,2n) and 1.6% ^{238}U(n,g). Noticeably, the simulated ratios differ by more than 15% when compared to the CST-LANL experiment for ^{238}U(n,2n).

The results for the two models of the Big Ten critical assembly are given in table 2 and are plotted in figure 3. They were calculated with MCNP5, Mercury, and Amtran using the ENDF.B_VII.0 data library. For ^{238}U(n,f), the two Monte Carlo codes underestimate the CST-LANL central fission ratio by ~5.5%, however, the result from the deterministic code differs by less than 3%. These differences decrease to 5 and ~2.4% when the simulations are compared to the CSEWG data. The ^{238}U(n,f)/ ^{235}U(n,f) is a measure of the hardness of the spectrum and it seems to indicate that the continuous-energy representation of the cross-section leads to a softer spectrum in the assembly than the multigroup representation. For ^{238}U(n,2n), Amtran and Mercury results are greater than the CST-LANL experiment by about 5% and by 3% for ^{238}U(n,g), while the MNCP results agree within 0.7% for these two reactions. Finally, when compared to the CSEWG data, the Amtran and Mercury results for ^{238}U(n,g) are well within the experimental error.

The central scalar flux for the Monte Carlo simulations is shown in figure 4. We checked that the geometry of the source and the number of initial cycles did not have a significant effect on the final flux in the central region and on the activation ratios: the code gave similar results for a point source and a volumetric source of spherical shape, or for 10 to 100 initial cycles.

The geometry of the model had some effect on the Mercury results. For the 1D model, they were within 1% of experimental results for ^{238}U(n,g), and 2.5% for ^{238}U(n,2n) as shown in table 2. Plus, the k_{eff} was higher than the k_{eff} calculated with the 2D model, and within one standard deviation of the benchmark value. The geometry of the problem had no effect on the Amtran results, but it did increase calculation time by a factor of *~300X.*

For the Monte Carlo simulations, the single most important factor was the number of histories per cycle, and the total number of active cycles since we were running in criticality calculation, i.e the simulation is sensitive to the total number of histories. The MCNP5 and Mercury results differ significantly from the experimental ratios for ^{238}U(n,2n) when running 10^5 histories/cycle. Also, when the number of active iterations is increased from 200 to a 1000, the difference between experiment and simulation increases from 2.5 to 5.5%, revealing important variations in the statistical error of the flux in the energy range above a few MeV. The energy threshold for ^{238}U(n, 2n) is 6.18 MeV, in a region where the flux decreases rapidly as shown in figure 4. Statistical fluctuations in the flux will lead to large variation in the activation ratio: there is no avoiding the fact that a large number of histories have to be run to estimate the flux in that region.

The 2D problem seemed to run more slowly for Mercury than for COG and MCNP. As a final note, the current implementation of Mercury does not allow easy reaction rate calculations for isotopes other than the ones in the assembly. For example, we cannot calculate ^{238}U(n,f) or ^{63}Cu(n,g) in a Pu assembly such as Jezebel. It may be worthwhile to add a reaction rate tally similar to those implemented in COG and MCNP5. By adding an isotope to a material and assigning it zero atom fraction, Amtran is able to calculate reaction rates for isotopes not appearing in the assembly.

Summary

We have compared calculations of critical assembly activation ratios using 3 different Monte Carlo codes and one deterministic code. There is excellent agreement. Discrepancies between the different Monte Carlo codes are at the 1-2% level. Notably, the deterministic calculations with 87 groups are also in good agreement with the continuous energy Monte Carlo results.

The three codes underestimate the ^{238}U(n,f) reaction , suggesting that there is room for improvement in the evaluation, or in the evaluations of other reactions influencing the spectrum in BigTen.

Until statistical uncertainties are implemented in Mercury, we strongly advise long runs to guarantee sufficient convergence of the flux at high energies, and we strongly encourage comparing Mercury results to a well-developed and documented code such as MCNP5 and/or COG. It may be that ENDL2008 will be available for use in COG within a year. Finally, it may be worthwhile to add a “standard” reaction rate tally similar to those implemented in COG and MCNP5, if the goal is to expand the central fission and activation ratios simulations to include isotopes that are not part of the specifications for the assembly material composition.

References

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```
Control
  ….
  Material_depletion analog (or expected_value)
…
end_control
…….
……
material
#  Material Data
#  -------------
  mat Uranium_10
    iso U234  za 92234  atom_fract 0.00105108  react_list None  end_iso
    iso U235  za 92235  atom_fract 0.101745    react_list None  end_iso
    iso U238  za 92238  atom_fract 0.897204    react_list None  end_iso
  end_mat
  mat Uranium_11
    iso U234  za 92234  atom_fract 0.00105108  react_list None  end_iso
    iso U235  za 92235  atom_fract 0.101745    react_list All  end_iso
    iso U238  za 92238  atom_fract 0.897204    react_list All  end_iso
    iso  U232 za 92232  atom_fract      0.0     react_list None  end_iso
    iso  U233 za 92233  atom_fract      0.0     react_list None  end_iso
    iso  U236 za 92236  atom_fract      0.0     react_list None  end_iso
    iso  U237 za 92237  atom_fract      0.0     react_list None  end_iso
    iso  U239 za 92239  atom_fract      0.0     react_list None  end_iso
    iso  FP   za 99120  atom_fract      0.0     react_list None  end_iso
  end_mat
end_material
…….
……

tally
#  Tally Data
#  ----------
  standard
    tal scalar_flux
      active on
      scope Cell
      output Plot_File
    end_tal
    tal material_depletion
      active On
      scope material
      output Out_File
    end_tal
  end_standard
end_tally
```

Figure 1. To calculate the loss/gain of isotopes in a material, three sections of the Mercury input decks must be modified. The changes are shown in bold.

```
probname = "bigten_1d"
probinfo = "1D version of Big Ten"

idgroup = 4                    #87 energy group set

mmax = 12                      # SN order (2*mmax)
ievt = 1                       # criticality (k eignevalue) calculation
ndim = 1                       # dimensionality

ixsecs = 1                     # use ENDL
genopt = 2                     # generate spherical shells
mfpmult   = 1.0                # mean free path multiplier for AMR zoning
legendrexp = 2                 # expansion order of legendre scattering term (P2)
temp_effects = 1               # use temperature dependent cross sections
epsin = 1.0e-4                 # convergence criteria
plot_now = 1

#problem limits
xlimit = { { 45.72, 0.} }
#zonal limits
ixlimit = { { 448, 1} }

#isotopic compositions for each material (material #1 is index 0, #2 is index 1, etc.)
isofrac[0] = { {92234, 0.00105108}, {92235, 0.101745}, {92238, 0.897204},
{94239, 0.0}}
isofrac[1] = { {92235, 0.00208030}, {92238, 0.997920} }

#set background temperature for each material (in MeV)
tempi[0] = 2.05e-6
tempi[1] = 2.05e-6

watch_point[0] = {0.0}                #location of watch point
watch_point[1] = {0.5}                #location of watch point
#activation calculated at each watch point {isotope, C number}
activation[0] = {92235, 15}
activation[1] = {92238, 12, 15, 46}
activation[2] = {94239, 12}

#inner and outer radius of each region
bigr[0] = { 0.0, 30.48 }
bigr[1] = { 30.48, 50.}

#density at inner and outer radius for each region (linear interpolation)
rhoi[0] = { 18.7799, 18.7799}
rhoi[1] = { 19.0015, 19.0015}

#material number associated with each region
iregsphr[0] = 1
iregsphr[1] = 2
```

Figure 2. Example of an Amtran input deck for reaction rate calculations. The differences with a deck used for a simple k_{eff} calculation are shown in bold.

Table I: COG and MCNP5 simulations are compared to CST_LANL experimental ratios for the Big Ten critical assembly. COG and MCNP5 simulations used the two-homogenized zones model of the assembly, and the ENDFB.VI cross-section library. Both runs were done with 20 initial cycles, 100 settle cycles, and 5e+04 histories/cycles.

Reaction	Experiment	MCNP	COG
^{238}U(n,2n)	1.73250E-03	1.458E-03	1.423E-03
^{238}U(n,f)	3.75000E-02	3.828E-02	4.036E-02
^{238}U(n,g)	1.06119E-01	1.052E-01	1.069E-01

Table II: The central activation ratios for ^{238}U(n,2n), (n,g), and (n,f) were calculated for the Big Ten assembly. Most results refer to the two-homogenized zones model. Mercury and Amtran simulations used ENDL2008 and MCNP, ENDFB.VII.0. The cross-sections for Z=92 should be the same in both libraries.

Reaction	Experiment CSEWG	Experiment CST-LANL	MCNP 2D		Amtran 1D	Amtran 2D	Mercury 1D	Mercury 2D	Mercury 2D
#histories/cycle * #active cycles			10^5*200	10^6*1075			10^6*1000	10^5*200	10^6*1000
^{238}U(n,2n)	-	1.73250E-03	1.8984E-03	1.7436E-03	1.83E-03	1.82E-03	1.7653E-03	2.0589E-03	1.8167E-03
^{238}U(n,f)	0.03739 ± 0.9%	3.75000E-02	3.5455E-02	3.5462E-02	3.66E-02	3.64E-02	3.5381E-02	3.5910E-02	3.5902E-02
^{238}U(n,g)	0.11±0.003	1.06119E-01	1.0683E-01	1.0677E-01	1.09e-01	1.09E-01	1.0699E-01	1.0826E-01	1.0893E-01
k_{eff}	0.9948±0.0013	-	0.9949±1e-4	0.9950±2e-5	0.9822*	0.9804*	0.9960±4e-5	0.9929±3e-4	0.9929±4e-5

- note: Amtran results are with a database that contains prompt nubar only.

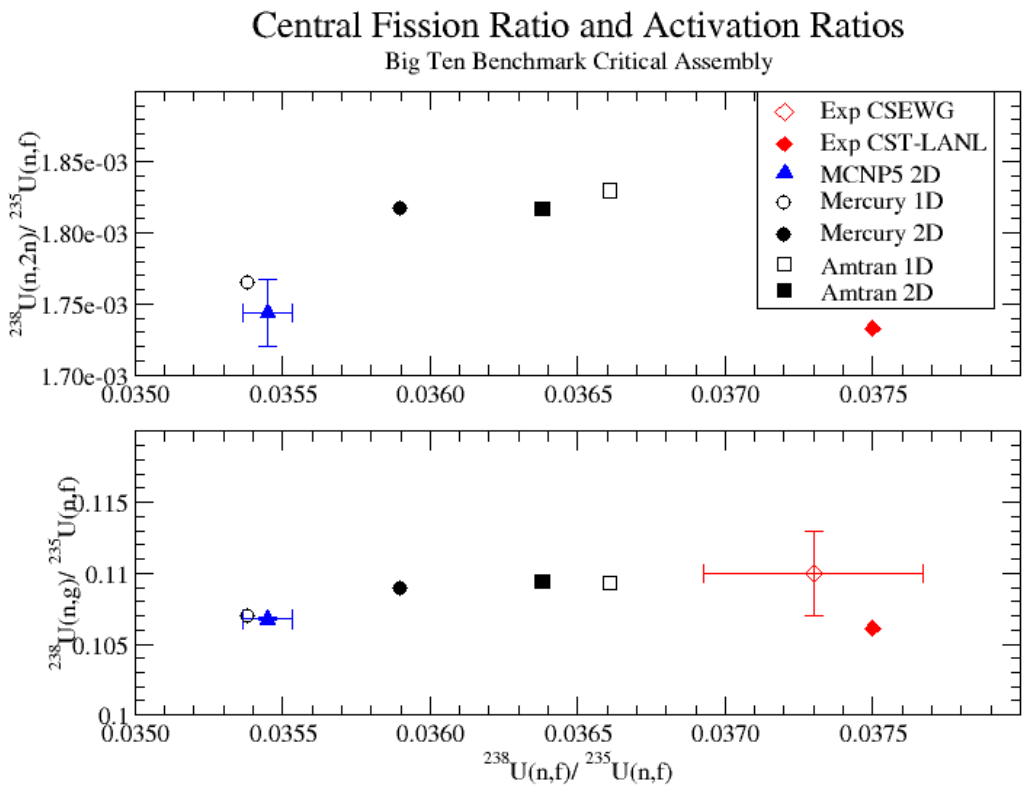

Figure 3. Simulated activation ratios in the Big Ten critical assembly are compared to experimental results from CST-LANL published by Wilkerson et al., and from CSEWG. Mercury and Amtran simulations were performed with ENDL2008.1, and MCNP5 simulations with ENDF.B-VII.0. There is no CSEWG data for the ^{238}U(n,2n) activation ratio. The MCNP5 results are shown with their statistical error.

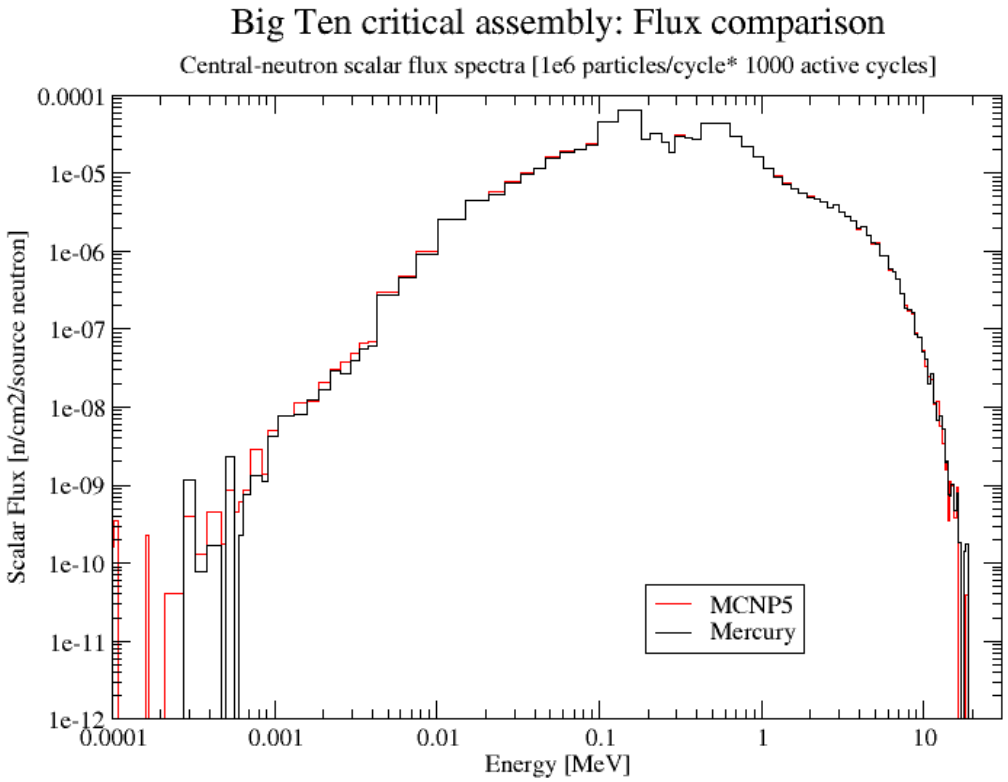

Figure 4. This plot shows the scalar flux in the central region of the 2D cylindrical model of the Big Ten critical assembly for MCNP5 and Mercury simulations run with the ENDF-B.VII.0 cross-section library. We tallied the flux in the standard bdfls 174 energy group-structure.