Godiva IV Central Cavity Neutron Environment Characterization with Threshold Neutron Detectors

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Abstract. Godiva IV is a cylindrical fast burst reactor comprised of approximately 65 kg of highly enriched uranium that is operated by Los Alamos National Laboratory and sited at the National Criticality Experiments Research Center at the Nevada National Security Site in Nevada in the United States. Godiva IV is typically operated at delayed critical and in the regime spanning from sub-prompt to super-prompt bursts. Godiva IV is used for sample irradiations, criticality safety demonstrations, dosimetry studies, and for studying super-prompt behavior. In preparation for both an upcoming experiment to reduce uncertainties in the prompt fission spectrum for ^{235}U using threshold neutron detectors, and for future research using Godiva IV, it was desired to exercise the process of the selection of threshold neutron detectors/activation foils, radiation metrology, and the subsequent adjustment of the neutron spectrum. For this exercise, nine high purity threshold neutron detectors/activation foils were irradiated in a Godiva IV burst. The foils were then analyzed using a high-purity germanium detector in the NCERC counting laboratory to determine end of irradiation specific activities for available IRDFF-II reactions. This work summarizes the Godiva IV foil irradiation, radiation metrology results, and adjusted neutron spectrum. The results of this exercise ultimately characterized the neutron environment inside the sample irradiation cavity inside Godiva IV to a higher degree than previously performed, informed decisions for the upcoming larger scale experiment, and will inform future neutron spectrum characterizations at NCERC.

1 Introduction

1.1 Motivation

In preparation for the Prompt Fission Uranium Neutron Spectrum (PFUNS) experiment at the National Criticality Experiments Research Center (NCERC) [1] and in support of future sample irradiations and research utilizing Godiva IV, it was desired to perform a

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spectral characterization exercise using threshold neutron detectors, or activation foils, in a Godiva IV burst irradiation. The PFUNS experiment, which was originally proposed in 2011, utilizes more than 20 activation foils and fission foils to reduce the uncertainties in the high-energy region of the prompt fission neutron spectrum (PFNS) in ^{235}U especially above approximately 5 MeV. The results from the foil activations are then planned to be used to infer an updated PFNS based on the spectral adjustment results. The foils were then counted, and the results were then used in the neutron spectral adjustment code STAYSL_PNNL [1]. This exercise informed decisions of the PFUNS experiment. In this work, the process of foil selection, irradiation, retrieval, gamma spectroscopy, through to the spectral adjustment process was exercised using Godiva IV.

1.1 Godiva IV general description

Godiva IV, shown in Figure 1, is a fast burst reactor with approximate dimensions of 7" in diameter and 6" in height comprised of approximately 65 kg of bare highly enriched uranium metal that is alloyed with 1.5 % molybdenum for strength [2]. Godiva IV is sited at NCERC in the Device Assembly Facility at the Nevada National Security Site after being relocated from the Los Alamos Critical Experiments Facility (LACEF) in Los Alamos, New Mexico USA. NCERC is a world-class facility offering unprecedented flexibility in critical experiments with the Godiva IV and Flattop critical assemblies as well as the Planet and Comet vertical lift machines [3,4].

Historically Godiva IV has been used to study super-prompt-critical behavior in addition to irradiations and demonstrations. In addition, Godiva IV is a benchmark in the International Criticality Safety Benchmark Evaluation Project (ICSBEP) handbook, as HEU-MET-FAST-086, and is also currently being revised with new measurements [5].

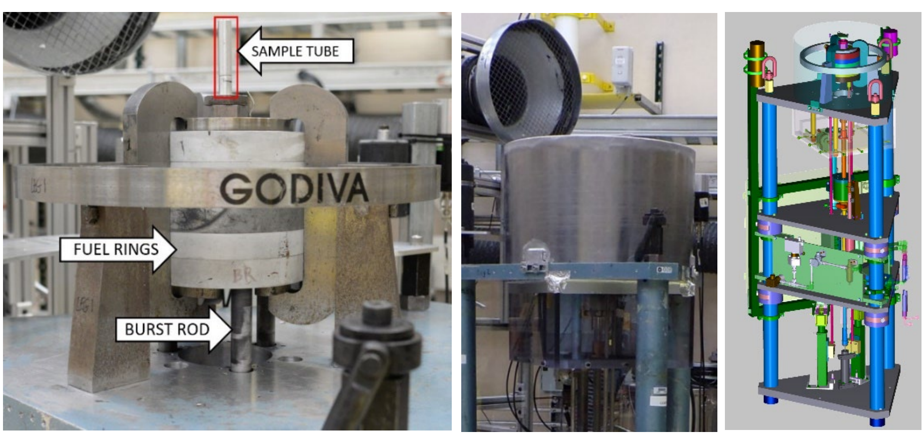

Fig. 1. (Left) Godiva IV with the 'top hat' removed and an aluminum sample tube inserted. (Middle) Godiva IV shown with the 'top hat' replaced. (Right) 3D CAD rendering of Godiva IV and associated mechanical drives and support structures.

2 Methodology

2.1 Godiva IV burst operations and irradiations

Burst operations on Godiva IV are typically characterized by a measured fuel temperature increase for a given reactivity insertion. Typical burst sizes for Godiva IV are 70, 150, and

250 °C of fuel temperature increase, which correspond to approximately $1.04, $1.07, and $1.10 of inserted reactivity respectively. For this work, a 250 °C burst was performed. Godiva IV super-prompt critical bursts of the same reactivity insertion were recently confirmed to be reproducible at NCERC to within approximately 3% uncertainty [6]. Before performing each burst the excess reactivity is verified using the Inhour equation and the measured period. During material irradiations, small scale samples, such as activation foils, can be accommodated through an aluminum sample tube, seen in Figure 2, that is inserted through the top of the assembly into a small diameter cavity referred to as the 'glory hole'. An aluminum contamination control shield referred to as the 'top hat' is also typically in place for Godiva IV irradiations. Samples inside of the tube are generally exposed to approximately 10^{14} neutrons/cm^2 on the scale of tens of milliseconds from approximately 10^{16} total fissions, depending on the burst increment. The samples can then be retrieved by personnel after properly allowing the dose rates to subside and relocated to the radiation metrology laboratory at NCERC.

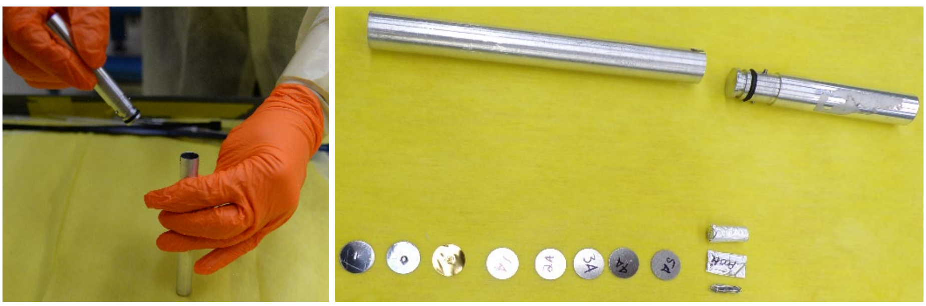

Fig. 2. (Left) An aluminum sample tube used in Godiva IV 'glory hole' irradiations. (Right) Example foil loadout for a Godiva IV burst irradiation.

Generally, the aluminum sample tube for Godiva IV is a limiting factor in the number of foils to be deployed. Therefore, a selection of 9 activation foils measuring 0.5 inch (12.7 mm) in diameter with varying thicknesses were utilized for this exercise and are recorded in Table 1. The process of predicting the foil activities post-irradiation, often needed for planning purposes and to ensure sufficient counting statistics for reactions of interest, was also carried out and is documented in other work [7].

Table 1. Activation foils used in the Godiva IV irradiation with mass uncertainty from vendor.

Foil material	Reported foil mass (mg)	Foil thickness (mil)
Gold (Au)	126.8(1)	2
Cobalt (Co)	66.0(1)	2
Copper (Cu)	141.3(1)	5
Iron (Fe)	124.8(1)	5
Nickel (Ni)	281.1(1)	10

Titanium (Ti)	141.9(1)	10
Magnesium (Mg)	28.6(1)	5
Niobium (Nb)	298.2(1)	10
Molybdenum (Mo)	90.6(1)	3

2.3 NCERC radiation metrology laboratory

The radiation metrology laboratory at NCERC contains a wide variety of quality tools to measure gamma spectra from samples. The laboratory contains multiple high-purity germanium (HPGe) detectors of varying intrinsic efficiencies, an automated sample changer with tungsten loaded polymer shielding, as well as recently installed Kolga shields coupled to HPGe detectors for reducing background. Each Kolga shield also has a sample planchet holder with varying distances available. The automated sample changer with a HPGe detector and shielding as well as the Kolga shields installed in the NCERC counting laboratory can be seen in Figure 3.

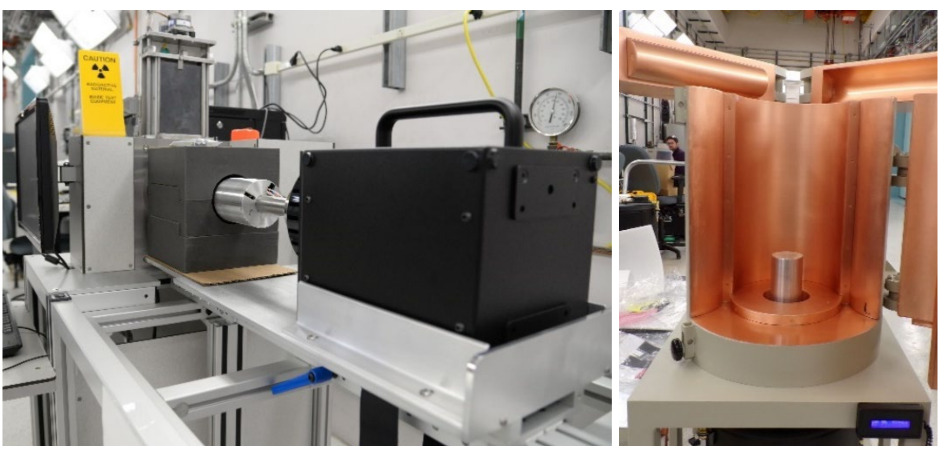

Fig. 3. (Left) Automated sample changer, HPGe detector, and tungsten loaded polymer shielding. (Right) Kolga shield with integrated HPGe and integrated Mobius liquid nitrogen cooler.

After irradiation, the activation foils fielded in the Godiva IV pulse were then removed from the sample tube and then affixed to sample planchets. The planchets containing the samples were then loaded into the automated sample changer with a 50% relative efficiency HPGe detector shielded by tungsten loaded polymer, as shown in Figure 3. After initial counting at NCERC, some foils with low activity and long half-lives were shipped back to Los Alamos National Laboratory for longer term counting.

To obtain end-of-irradiation specific activities, multiple measurements of fixed time were taken. Gamma peak fitting software, with correction options, was then used to determine counts in the specific peak of interest for each time bin. The values of each time bin were used in conjunction with the predetermined detector efficiency, branching ratios, and decay correction to determine the best fit to an exponential (or two if overlapping lines) with a fixed decay constant lambda. A Python code was used to determine the lines of best fit with uncertainty propagation. The uncertainties in counting were combined in quadrature with the previously determined systematic uncertainty of 2.4%.

2.2 STAYSL_PNNL dosimetry least squares neutron spectral adjustment

STAYSL_PNNL is a software package developed by Pacific Northwest National Laboratory that solves the linear least squares formulation of the neutron spectral adjustment problem using IRDFF-II standard reactions and covariances [8]. The package contains many user-friendly features that can be employed for convenience. For example, the software allows the user to correct for things like the irradiation history, the alloying fraction, gamma-ray self-attenuation based on sample shape and thickness, and neutron shielding and self-shielding. In this work, the measured specific activities in conjunction with their associated uncertainties, the well-informed *a priori* neutron spectrum estimate, and necessary corrections were used to obtain an adjusted neutron spectrum and fluence values.

2.3 MCNP6.2 neutron transport simulations

For the *a priori* neutron spectrum that is required by the STAYSL_PNNL dosimetry least squares neutron spectral adjustment code, the pre-existing and recently updated simplified model of Godiva IV in MCNP6.2 [9] originally used in the HEU-MET-FAST-086 ICSBEP benchmark, shown in Figure 4, was utilized to obtain the neutron energy spectrum. The neutron energy spectrum was obtained inside the aluminum sample tube at the bottom where the foils were physically located. To mitigate any potential re-binning errors/bias, the 100-group energy structure used by STAYSL_PNNL was used in conjunction with ENDF/B-VIII nuclear data for the MCNP6.2 tally. A neutron flux tally using UKAEA 709-group energy structure was placed at the same location used in previous work and can be seen in Figure 4 for illustrative purposes.

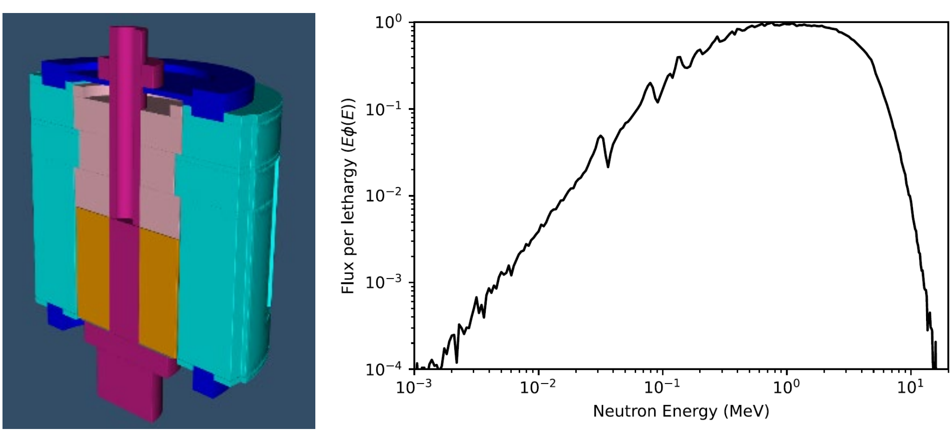

Fig. 4. (Left) *Gxsview* cross sectional 3D rendering of the MCNP6.2 model of Godiva IV with the safety block (orange) inserted that was used in this work. (Right) UKAEA 709-group structure fluence tally (scaled) at the foil locations for illustrative purposes.

3 Results

3.1 Radiation metrology

Of the IRDFF-II reaction standards that were available for the foil loadout in this work, 15 reactions obtained results that were deemed acceptable for this work. A notable reaction of issue that was ultimately removed from the analysis was the ^{197}Au(n,2n)^{196}Au reaction that was shown to contribute highly to the chi-squared metric. Uncertainties in the specific activities of each of the 15 viable IRDFF-II reaction standards listed in Table 2 were propagated in quadrature to include uncertainties in: the mass reported by the vendor (~0.1%), detector counting efficiency (~1-2%), counting systematic uncertainty (~1-2%), counting statistics (~0.1-1%), gamma-line branching ratio uncertainties (~1-5%), sample self-attenuation (~1-5%), and so on. In the future experiments and in the PFUNS experiment, uncertainty in the mass of each foil will be more rigorously reported through repeatability measurements and through more precise scales. It was found, however, that the primary drivers of uncertainty in the final reported specific activity are more closely related to the counting statistics and the systematic uncertainty. It is noted that for the neutron spectral adjustment process, it is crucial to not underestimate the uncertainties as the solution space will be unphysically constrained. Over-constrained adjustment often produces artificialities in the adjustment at energies with large cross-section differences between reactions to compensate for measured reaction rates that disagree by only a few percent when the constraining uncertainty is smaller.

Table 2. Measured specific activity from IRDFF-II reactions, estimated saturation values, and the associated propagated uncertainty in specific activity.

IRDFF-II reaction standard	Measured specific activity (Bq/g_{foil})	*STAYSL_PNNL* σϕ value ($atoms_{prod}/atom_{target}$-s)	Uncertainty (%)
^{197}Au(n,g)^{198}Au	5.5953E+05	6.149E-11	3.2
^{58}Fe(n,g)^{59}Fe	7.4239E+00	1.361E-12	4.4
^{59}Co(n,g)^{60}Co	1.5563E+02	3.655E-12	3.5
^{24}Mg(n,p)^{24}Na	1.0073E+05	3.985E-13	3.3
^{46}Ti(n,p)^{46}Sc	2.9189E+02	2.938E-12	3.2
^{47}Ti(n,p)^{47}Sc	1.1619E+04	5.182E-12	3.0
^{54}Fe(n,p)^{54}Mn	3.5678E+02	2.202E-11	3.5
^{54}Fe(n,a)^{51}Cr	4.1678E+01	2.284E-13	3.9
^{59}Co(n,2n)^{58}Co	8.2278E+01	7.112E-14	3.5
^{59}Co(n,p)^{59}Fe	7.2113E+02	3.914E-13	3.5

$^{59}Co(n,a)^{56}Mn$	3.4157E+03	4.477E-14	3.5
$^{58}Ni(n,p)^{58}Co$	2.3071E+04	2.918E-11	3.5
$^{60}Ni(n,p)^{60}Co$	6.5943E+00	5.882E-13	4.2
$^{63}Cu(n,a)^{60}Co$	4.0133E+00	1.470E-13	3.6
$^{92}Mo(n,p)^{92m}Nb$	1.4128E+03	1.928E-12	4.0

3.2 STAYSL_PNNL neutron spectral adjustment

Specific activities in Table 2 were used in conjunction with the STAYSL_PNNL's SigPhi calculator to provide final inputs for the STAYSL_PNNL spectral adjustment tool. The adjusted spectrum and *a priori* input are shown in Figure 5. The percent adjustment from the simulation is shown in Figure 6. The results from STAYSL_PNNL obtained a reduced chi^2 metric of 0.84628. The results from this exercise also correspond well with previous results.

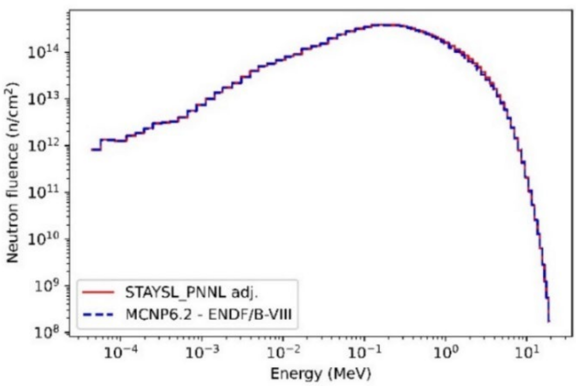

Fig. 5. STAYSL_PNNL adjusted spectrum and *a priori* spectrum from MCNP6.2 using ENDF/B-VIII data.

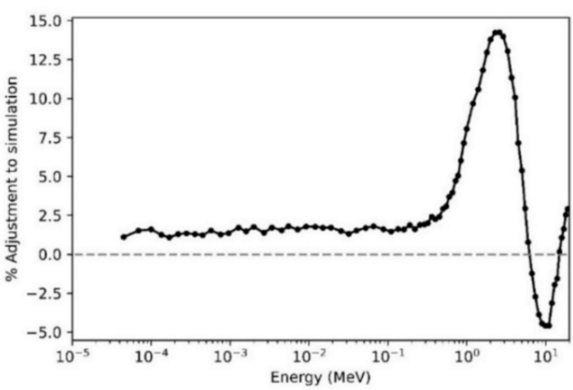

Fig. 6. Adjustment to simulation, STAYSL_PNNL output versus MCNP6.2 using ENDF/B-VIII.

4 Conclusions

This work summarizes the characterization of the neutron spectrum inside the Godiva IV cavity for a 250ºC burst using a suite of activation foils. Ultimately the neutron spectrum was minimally adjusted in lower energies but was subject to a much higher adjustment in the 500 keV to a few MeV region. The spectral adjustment obtained a reduced chi^2 metric of 0.864628 and compared favorably with other previous work [11,12]. After STAYSL_PNNL spectral adjustment, the updated total fluence with respect to the new energy spectrum was estimated as 5.068×10^{14} with an uncertainty of 3.03 %. Oscillations in the adjustment in higher energies, as seen in Figure 6, are also seen in previous work [11,12].

This work was supported by the U.S. Department of Energy (DOE) Nuclear Criticality Safety Program (NCSP), which is funded and managed by the National Nuclear Security Administration (NNSA) for the DOE. Los Alamos National Laboratory is operated by Triad National Security, LLC, for the National Nuclear Security Administration of the US Department of Energy under Contract No. 89233218CNA000001.

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