Isotope Production and Materials Irradiation Research Studies to Support HFIR LEU Conversion Assessment[1]

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INTRODUCTION

This paper presents results from a series of high-impact isotope production and materials irradiation research studies to support the conversion of the High Flux Isotope Reactor (HFIR) from highly enriched uranium (HEU) to low-enriched uranium (LEU) fuel. Conversion is expected to take place around 2040. HFIR's mission capabilities must be maintained or enhanced after conversion to sustain the facility's world-leading scientific capabilities. The Shift [1,2] and ORIGEN [2] codes are used to characterize isotope production and materials irradiation research metrics in the flux trap irradiation facility with the HEU-fueled core and a proposed LEU core with a uranium silicide fuel design.

High Flux Isotope Reactor and Flux Trap

HFIR is an 85 MW_{th}, HEU-fueled (U_3O_8-Al, ~93 wt %), pressurized, light-water–cooled and –moderated, beryllium-reflected research reactor operated at Oak Ridge National Laboratory (ORNL) on behalf of the US Department of Energy (DOE) Office of Science. HFIR is used for neutron scattering research, isotope production, materials and fuels irradiation research, neutron activation analysis, and other radiation-based research.

HFIR was designed with a compact high power density core to promote neutron leakage into experiment regions, enabling unparalleled in-vessel irradiation facility flux environments (e.g., $\phi_{th,max} > 2\times10^{15}$ n·cm^{-2}·s^{-1}). A central over-moderated flux trap is nested inside two cylindrical fuel elements which are surrounded by two concentric control elements and a large beryllium reflector (Fig. 1).

A basket-type flux trap design includes 37 vertical target positions, with 31 inside the basket and 6 around the periphery (Fig. 2). Each aluminum target tube is typically loaded with up to 8 individual capsules or a stack of ^{252}Cf production pellets. The peripheral targets are typically loaded with capsules containing materials irradiation specimens that require high fast neutron fluxes. The 31 interior positions comprise a hydraulic tube and 30 targets containing isotope production and materials irradiation capsules. Thermal (E_n < 0.625 eV) and fast (E_n > 0.1 MeV) flux distributions are illustrated in Fig. 2 for a representative flux trap loading.

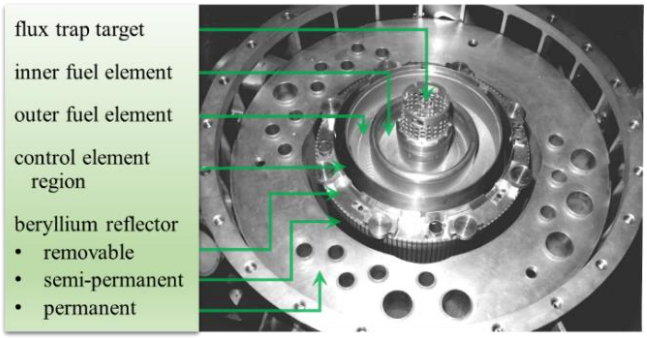

Fig. 1. HFIR core mockup [3].

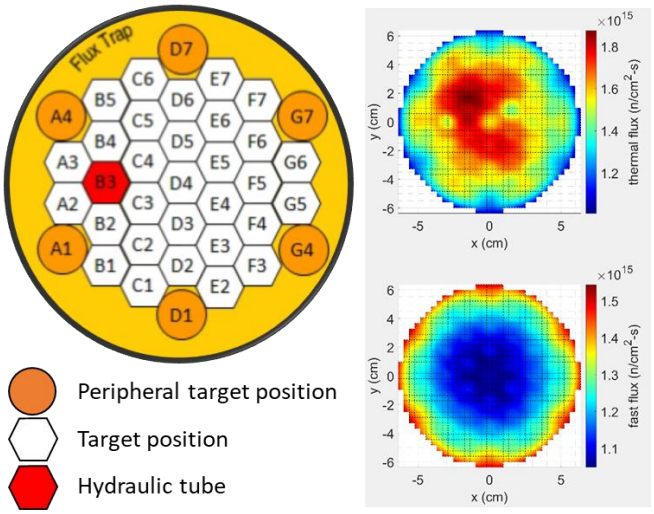

Fig. 2. Flux trap map (left), thermal (top right) and fast (bottom right) flux distributions on core midplane.

Low-Enriched Uranium Conversion

As part of the DOE National Nuclear Security Administration Office of Material Management and Minimization's mission to eliminate the use of HEU in civilian nuclear applications to the greatest extent possible, HFIR is evaluating conversion to LEU fuel. LEU silicide dispersion (U_3Si_2-Al, 19.75 wt %) became the primary fuel form candidate for HFIR conversion in 2019. Several U_3Si_2-Al designs with various fuel fabrication features and

fuel densities have been developed and documented by Bae et al. [4] and Betzler et al. [5]. These U_3Si_2-Al designs meet key performance metrics (KPMs) if power is increased from 85 to 95 MW. The KPMs are evaluated in line with the design study depletion calculations and include cycle length, cold source cold flux, thermal flux in select isotope production irradiation facilities, ^{252}Cf production rates, and fast flux in select materials irradiation facilities [6]. An LEU-fueled core design must maintain or exceed the HEU core KPMs to be considered a candidate. In this paper, higher fidelity performance comparisons are presented to provide additional confidence that a 95 MW LEU core can maintain 85 MW HEU-like core performance.

Isotope Production and Materials Irradiation Research

Isotopes are produced in the HFIR flux trap for energy, medical, industrial, security, and research purposes. The ^{252}Cf isotope is used as a source for a variety of unique applications such as fuel enrichment evaluations, reactor startups, shipping container inspections, well logging analyses, and university education and research. The ^{63}Ni isotope is used for national security applications, ^{75}Se is used for commercial and industrial gamma radiography, and ^{133}Ba is used to monitor oil, gas, and water flow data in the oil and gas industry. Medical isotopes for treating conditions like cancer, arthritis, bone metastases, and restenosis include ^{89}Sr, ^{117m}Sn, ^{177}Lu, ^{188}W, ^{227}Ac, and ^{229}Th [7]. Additional isotopes such as ^{238}Pu, which is used in the form of PuO_2 to fuel radioisotope power systems for NASA's deep-space and planetary missions, are produced in the beryllium reflector.

The flux trap facilities are also used to study the effects of neutron irradiation damage on materials. Materials irradiation research provides insight on key phenomena such as irradiation-induced hardening, embrittlement, phase instabilities, precipitation, creep, and growth. Materials irradiation research is performed to support efforts to extend the life of the current fleet of fission-based reactors and to evaluate materials proposed for use in future fission- and fusion-based reactors [7].

STUDY METRICS AND METHODS

The thermal flux in the flux trap is calculated during the LEU design studies [4,5] as a KPM to capture at a high level a design's ability to maintain HEU-like isotope production capabilities. However, isotope production depends on the full energy spectrum; therefore, higher fidelity isotope production metrics are assessed in this study. The fast flux in the flux trap is used as a KPM to capture at a high level a design's ability to maintain materials irradiation research capabilities. A major contributor of neutron irradiation damage in metals is the displacements per atom (dpa) from their normal lattice sites [9]; therefore, the number of times an atom has been displaced during irradiation is a higher fidelity radiation damage metric and is assessed in this study.

The Shift Monte Carlo–based radiation transport and depletion tool with ORIGEN was deployed for neutron transport, fuel depletion, control element withdrawal and activation, and select isotope production target activation calculations. The beginning-of-cycle explicit HEU fuel and representative experiment loading input [8] was used as the basis for the HEU calculations. The low-density (4.8 gU/cm^3) U_3Si_2-Al fuel design, referred to as the *optimized design* in Betzler et al. [5], was used for the LEU calculations. The two models are equivalent except for the fuel plate internals (e.g., fuel and filler lengths, shapes, and material compositions). Each transport step simulated 100 million active histories and 5 million inactive histories.

Additional tallies were defined in the depletion calculations to characterize intracycle fluxes and radiation damage metrics. Neutron flux mesh cell tallies were used to calculate the 252-energy group flux in the 8 axial capsule positions within each of the 37 flux trap target positions. Iron-based dpa results were also calculated on the same spatial mesh. The response function detailed in the American Society for Testing and Materials E693–17 standard [9] and the light-water reactor ex-core study by Davidson et al. [10] was used. The dpa cross section and flux trap average neutron flux spectrums are illustrated in Fig. 3.

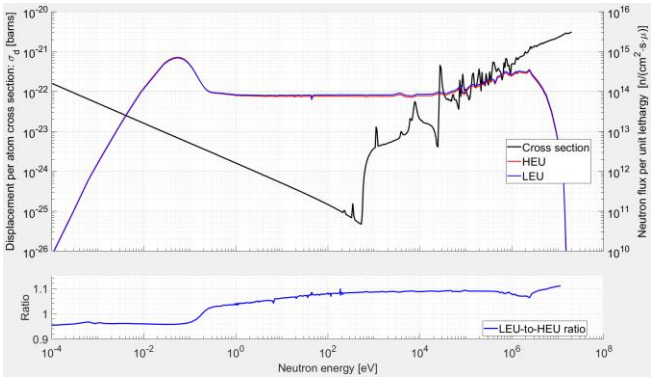

Fig. 3. Iron dpa cross section and flux trap flux spectrums.

Five full-length targets loaded with actinide oxide dispersion pellets were modeled in the flux trap and irradiated in the single-cycle Shift simulations to produce ^{252}Cf and other heavy isotopes. Standalone ORIGEN (SCALE V6.3.0) was then used for follow-on isotope production and sensitivity coefficient studies to evaluate materials not explicitly modeled in Shift. Cycle-averaged fluxes (total and 252-energy group) were calculated from the Shift data discussed in the above paragraph to generate the ORIGEN cross section libraries and to irradiate the subject materials.

ISOTOPE PRODUCTION RESULTS

The Shift-calculated HEU and LEU cycle lengths are 26.0 days (2,210 MWd) and 27.4 days (2,603 MWd), respectively. Production of the ^{252}Cf and ^{249}Bk isotopes is slightly increased with the LEU core as illustrated in Fig. 4.

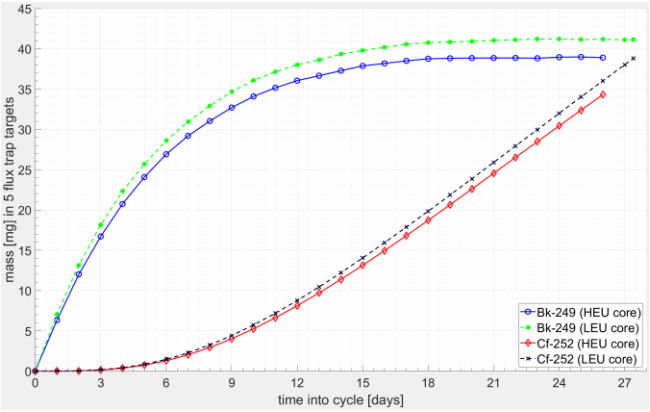

Fig. 4. ^{249}Bk and ^{252}Cf mass vs. time into cycle.

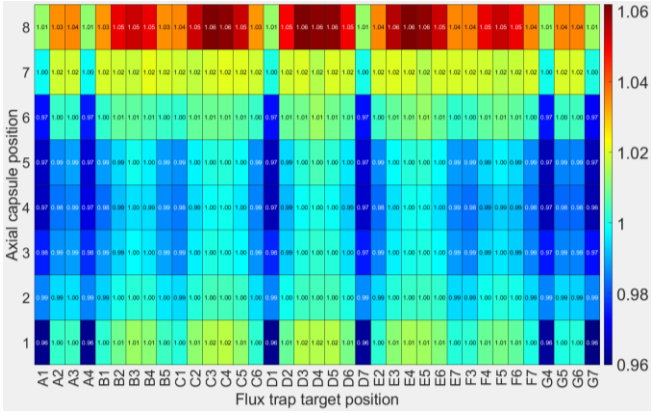

Fig. 5. Flux trap map of LEU-to-HEU ^{63}Ni mass ratios.

Standalone ORIGEN calculations were also performed to assess the effects of LEU conversion on a select set of high-priority isotopes in the flux trap. The product, feed, and irradiation histories evaluated are provided in Table I. The ^{177}Lu and ^{227}Ac isotopes are produced in the hydraulic tube in less than a full cycle of irradiation. The other isotopes are produced in 1–14 cycles. Outages between cycles were assumed to last for two weeks, and production values were estimated in all flux trap positions. The results indicate that conversion will not significantly affect the production of these isotopes. However, the production of select isotopes in select positions is reduced by up to 7% with the LEU core.

TABLE I. Flux Trap LEU-to-HEU Production Summary

Product isotope	Feed isotope(s)	Irradiation time	LEU-to-HEU production ratio*
^{63}Ni	^{62}Ni	12 cycles	0.96–1.06 (1.00)
^{75}Se	^{74}Se	1 cycle	1.02–1.11 (1.04)
^{89}Sr	^{88}Sr	1 cycle	1.02–1.20 (1.07)
^{117m}Sn†	^{117}Sn	1 cycle	1.07–1.37 (1.13)
^{133}Ba	^{132}Ba	8 cycles	0.97–1.14 (1.03)
^{177}Lu	^{176}Lu/^{176}Yb	7 days	0.93–1.07 (1.01)
^{188}W	^{186}W	2 cycles	0.94–1.34 (1.07)
^{227}Ac	^{226}Ra	14 days	1.00–1.11 (1.03)
^{229}Th	^{226}Ra/^{228}Ra	5 cycles	0.99–1.15 (1.03)

* ratios are listed as minimum–maximum (average)
† ^{117}Sn(n_{fast},$n'\gamma$)^{117m}Sn inelastic scattering

Figure 5 is a flux trap map illustrating the LEU-to-HEU ^{63}Ni production ratios after 12 irradiation cycles, or about 329 irradiation days with the LEU core (31,236 MWd) and 312 days with the HEU core (26,520 MWd). Production of ^{63}Ni is approximately maintained in its typical irradiation positions (i.e., interior targets near the core midplane). The ratios in axial positions 2–7 are close to 1.00. However, the ratios in position 8 are greater because the LEU fuel length is 55.88 cm, whereas the HEU fuel length is 50.80 cm. Additionally, the bottoms of the 6 peripheral targets are about 4.5 cm below those of the 31 interior targets, explaining the axial offset observed for these 6 targets in Figs. 5 and 6.

ORIGEN was also used to calculate the sensitivity coefficients using adjoint-based depletion perturbation theory to better understand the reaction and decay channels important to the production of these isotopes. The reaction loss coefficients greater than 0.1, provided in Table II, are similar for the HEU and LEU cores. The results are spatially dependent; however, the average results for all flux trap positions are provided. The coefficients are expressed as the relative change in isotope production (e.g., ^{63}Ni) per change in the reaction channel (e.g., ^{62}Ni*(n,γ)*).

TABLE II. Primary Reaction Loss Sensitivity Coefficients

Product isotope	Reaction(s)	Coefficients HEU | LEU
^{63}Ni	^{62}Ni*(n,γ)*	0.75 | 0.75
	^{63}Ni*(n,γ)*	−0.38 | −0.38
^{75}Se	^{74}Se*(n,γ)*	0.87 | 0.86
	^{75}Se*(n,γ)*	−0.40 | −0.41
^{89}Sr	^{88}Sr*(n,γ)*	1.00 | 1.00
^{117m}Sn	^{117}Sn($n,n'\gamma$)	1.00 | 1.00
^{133}Ba	^{132}Ba*(n,γ)*	0.87 | 0.87
^{177}Lu	^{176}Lu*(n,γ)*	−0.23 | −0.20
^{188}W	^{187}W*(n,γ)*	0.97 | 0.97
	^{186}W*(n,γ)*	0.80 | 0.79
^{227}Ac	^{226}Ra*(n,γ)*	0.91 | 0.91
	^{227}Ac*(n,γ)*	−0.55 | −0.54
^{229}Th	^{226}Ra*(n,γ)*	0.69 | 0.68
	^{228}Th*(n,γ)*	0.35 | 0.33
	^{229}Th*(n,γ)*	−0.50 | −0.53
	^{229}Th*(n,f)*	−0.20 | −0.21

MATERIALS IRRADIATION RESEARCH RESULTS

The single-cycle cumulative dpa values were calculated in all 296 flux trap capsule positions. Table III summarizes the single-cycle flux trap cumulative dpa values for the HEU and LEU cores. On average, the LEU core results in a 16% increase in dpa per cycle. The combination of LEU fuel and the power uprate leads to increased fast fluxes (Fig. 3) and dpa values. Figure 6 is a flux trap map of the LEU-to-HEU

dpa ratios. The ratios in axial positions 3–5 are approximately 1.10, but the ratios at the axial ends (i.e., positions 1 and 8) vary between 1.16 and 1.39 because of the fuel length differences discussed in the previous section.

TABLE III. Flux Trap LEU and HEU Iron dpa Summary

	HEU (dpa)	LEU (dpa)	LEU-to-HEU ratio
minimum	0.47	0.62	1.10
maximum	1.97	2.17	1.39
average	1.31	1.49	1.16

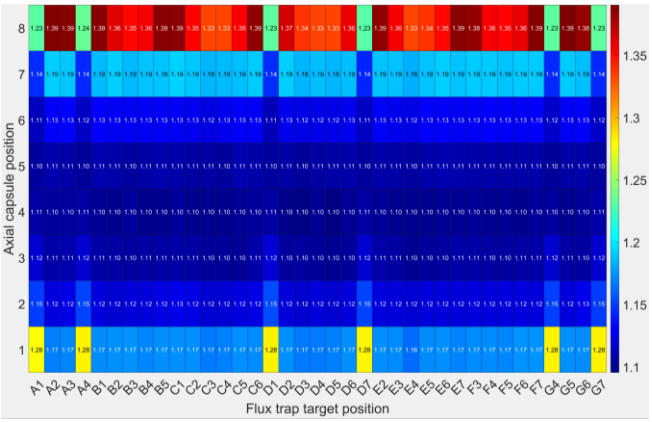

Fig. 6. Flux trap map of LEU-to-HEU neutron dpa ratios.

CONCLUSIONS AND FUTURE WORK

ORNL is evaluating HFIR conversion to LEU U_3Si_2-Al fuel and has developed fuel designs that meet HEU-like KPMs (e.g., few group fluxes in various experiment facilities) defined to capture HFIR's capabilities at a high level. The SCALE Shift and ORIGEN codes were used to develop methods and characterize isotope production and materials irradiation research metrics with the HEU core and a proposed LEU U_3Si_2-Al fuel design. The evaluations indicate that conversion and a simultaneous power uprate from 85 to 95 MW will enhance materials damage capabilities to support materials irradiation research and approximately maintain key isotope production capabilities.

This study, which includes a detailed HEU-to-LEU fuel comparison but does not include a detailed validation effort, is the first step in assessing isotope production and materials irradiation research metrics. Measurement data from irradiation campaigns of the studied isotopes are being collected to determine validity of the results and to determine if additional studies are required. Ongoing studies are being performed in collaboration with the Plutonium-238 Supply Project to validate multicycle ^{238}Pu production calculational methods. Validation of ^{252}Cf and other heavy isotope production is being initiated as part of these LEU conversion efforts. Explicit modeling and simulation of pertinent isotope production and materials irradiation capsules in the flux trap may also be performed to better characterize neutron fluxes, heat deposition and isotope production rates, and material-specific damage rates with the HEU and proposed LEU cores. These studies will be the subjects of future publications.

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