Studies of Plutonium-238 Production at the High Flux Isotope Reactor

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INTRODUCTION

The High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory (ORNL) is a versatile 85 MW_{th}, pressurized, light water-cooled and –moderated research reactor. The core consists of two fuel elements, an inner fuel element (IFE) and an outer fuel element (OFE), each constructed of involute fuel plates containing high-enriched-uranium (HEU) fuel (~93 wt% $^{235}U/U$) in the form of U_3O_8 in an Al matrix and encapsulated in Al-6061 clad. An over-moderated flux trap is located in the center of the core, a large beryllium reflector is located on the outside of the core, and two control elements (CE) are located between the fuel and the reflector. The flux trap and reflector house numerous experimental facilities which are used for isotope production, material irradiation, and cold/thermal neutron scattering.

Over the past five decades, the US Department of Energy (DOE) and its agencies have been producing radioisotope power systems used by the National Aeronautics and Space Administration (NASA) for unmanned, long-term space exploration missions. Plutonium-238 is used to power Radioisotope Thermoelectric Generators (RTG) because it has a very long half-life ($t_{1/2} \approx 89$ yr.) and it generates about 0.5 watts/gram when it decays via alpha emission. Due to the recent shortage and uncertainty of future production, the DOE has proposed a plan to the US Congress to produce ^{238}Pu by irradiating ^{237}Np as early as in fiscal year 2011 [1]. An annual production rate of 1.5 to 2.0 kg of ^{238}Pu is expected to satisfy these needs and could be produced in existing national nuclear facilities like HFIR and the Advanced Test Reactor (ATR) at the Idaho National Laboratory (INL). Reactors at the Savannah River Site were used in the past for ^{238}Pu production but were shut down after the last production in 1988. The nation's ^{237}Np inventory is currently stored at INL [1].

The process of producing ^{238}Pu includes: (1) fabricating $^{237}NpO_x$-Al targets, (2) irradiating the targets in a nuclear reactor which includes two reactions: $({}^{237}_{93}\mathrm{Np} + {}^{1}_{0}\mathrm{n} \rightarrow {}^{238}_{93}\mathrm{Np})$ then $({}^{238}_{93}\mathrm{Np} + {}^{0}_{1}\mathrm{e} \rightarrow {}^{238}_{94}\mathrm{Pu})$, and (3) recovering the ^{238}Pu in the targets via chemical extraction. The HFIR complex is ideal for these operations because it is home to both the reactor, which produces a very high neutron flux for irradiations, and the Radiochemical Engineering Development Center (REDC), which would be used to perform all of the post-irradiation processing activities.

At HFIR, ^{238}Pu production could occur in the vertical experiment facilities (VXF) in HFIR's permanent beryllium reflector. Sixteen small (radius ≈≈2.012 cm) and 6 large (radius ≈≈3.599 cm) VXFs are located within the reflector. The ^{237}Np target rods (radius = 0.3937 cm and height = 50.8 cm) have been conceptualized (not yet produced) to hold 35 pellets (radius = 0.3175 cm and height = 1.4478 cm) with helium as gap filler. The pellets are to be composed of 20 vol% NpO_x (impurities include Pu, Th, U, etc.) in an aluminum matrix. The small and large VXF target bundles are to be assembled in an 8 target polar array and a 31 target triangular array, respectively as illustrated in Figure 1 [2]. The figure was taken from the created VESTA model.

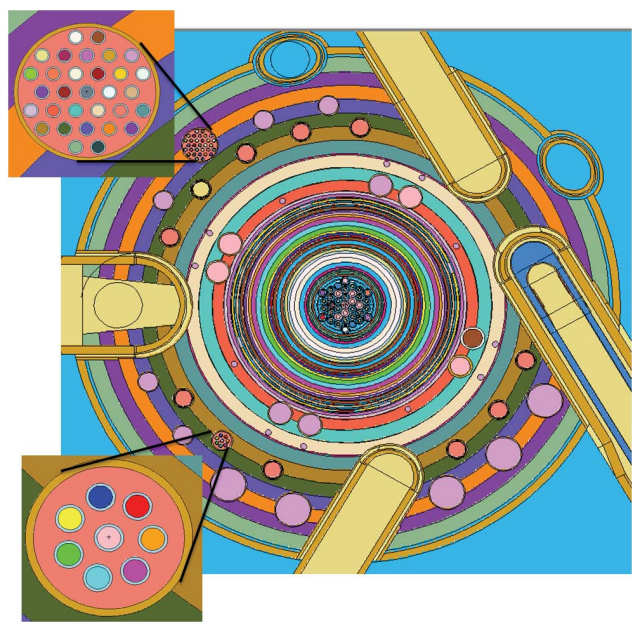

Fig. 1. Cross-sectional view of the HFIR illustrating the small (lower left) and large (upper left) VXF target arrays simulated in this study.

Studies are currently being performed to understand the production rates of ^{238}Pu in HFIR without diminishing reactor performance or affecting its varied scientific missions. The quality of the Plutonium (content of ^{238}Pu within the Pu vector), fission gas buildup, and heat generation rates are also being studied. This is one of several studies aimed at better understanding the impact of a variety of factors that can significantly affect reaction rates in the target regions. Primarily, target loadings,

target arrangements, flux levels, control element position, all versus space and time.

METHODOLOGY

Version 6 of the Standardized Computer Analyses for Licensing Evaluation (SCALE) code package [3] and VESTA Version 2.0.2 [4] were utilized in this study. SCALE is developed and maintained by ORNL and VESTA is developed and maintained by L'Institut de Radioprotection et de Sûreté Nucléaire (IRSN) in France. The TRITON depletion sequence in SCALE, which couples the three-dimensional Monte Carlo transport code KENO-VI and the depletion and decay code ORIGEN-S, was employed along with the 238-group ENDF/B-VII neutron cross section library for burn-up calculations. VESTA is a Monte Carlo-based depletion tool that couples MCNP (Version 5 in this case) to ORIGEN 2.2 and was also used for depletion calculations along with the ENDF/B-VII continuous energy cross section library.

The TRITON and VESTA inputs were created by modifying the inputs documented in References 5 and 6, respectively, by modeling the target bundles in the VXFs and setting up the appropriate irradiation/decay history. These inputs were used to produce the output in the Results section. In both code inputs, the IFE and OFE are discretized into 8 and 9 radial regions, respectively, in order to model the varying, spatially-averaged ^{235}U concentrations. The fuel elements in the VESTA input are divided into 19 axial regions since fuel burnup and control element withdrawal are simulated as a function of cycle time. They are modeled as one axial region in the TRITON input since fuel burnup is not modeled. Both inputs use fresh fuel and do not model any impurities in the Be reflectors.

The KENO-VI/TRITON input was originally constructed to activate permanent beryllium reflector number 3 throughout its duration in HFIR (Jan. 1984 – Oct. 2000) to study its composition at discharge as well as during post-discharge decay. Since a 16 year period was modeled, fuel depletion/replacement wasn't performed and borated water was used to simulate the control elements and control reactivity. The fluxes in the beryllium reflector were shown to be slightly greater (~9 and ~18% for the ISVXF and LVXF, respectively) than the approximate, cycle-averaged fluxes [5].

Rather than modeling 35 pellets in each target, the target rods were modeled as a single pin within the Al clad. Each target pin was assigned a different material number such that each target pin was depleted individually. The targets were irradiated for 24 days, which is a "typical" HFIR cycle length. From out the output, we could gather how much ^{238}Pu could be produced from different targets over a single cycle and then estimate the yearly production rates.

RESULTS

Using the modified inputs, the single-cycle ^{238}Pu production rates in one small VXF array (8 targets) and one large VXF array (31 targets) as calculated with VESTA and TRITON are shown in Figure 2. For a 24-day cycle starting with fresh targets, the VESTA calculation shows that 10.8 and 15.1 grams of ^{238}Pu can be produced in a small and large VXF array, respectively, and the SCALE/TRITON calculation shows that 11.0 and 17.1 grams of ^{238}Pu can be produced, respectively.

The conversion ratio (ratio of final ^{238}Pu mass to initial ^{237}Np mass) is 1.99% in the large VXF and 5.46% in the small VXF. The production rate does not scale proportionately with the number of targets because the large VXF array is more tightly packed (self-shielding effects) and further from the core (lower fluxes) in comparison to the small VXF array The beginning-of-cycle and end-of-cycle reactivity worth of one loaded large and one small VXF target array with respect to no loaded targets (the original input) as calculated with VESTA/MCNP5 are -0.5 ± 2 and -1.8 ± 3 cents, respectively.

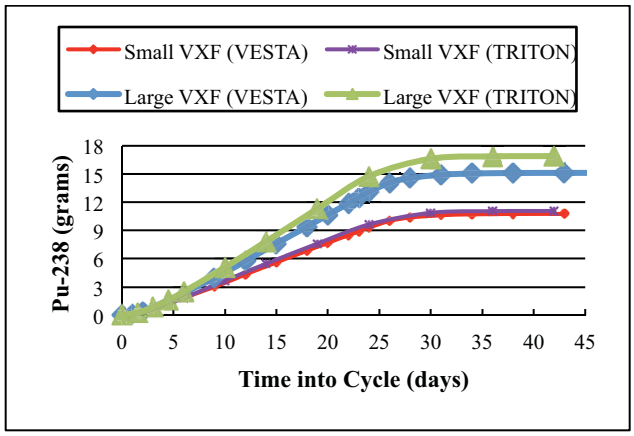

Fig. 2. Estimated single cycle ^{238}Pu production in small and large VXF targets using VESTA and TRITON.

CONCLUSIONS

A plan for producing ^{238}Pu at US research reactor facilities such as the High Flux Isotope Reactor at ORNL has been initiated by the US DOE and NASA for space exploration needs. Two Monte Carlo-based depletion codes, TRITON (ORNL) and VESTA (IRSN), were used to study the ^{238}Pu production rates with varying target configurations in a typical HFIR fuel cycle. Preliminary studies have shown that approximately 11 grams and within 15 to 17 grams of ^{238}Pu could be produced in the first irradiation cycle in one small and one large VXF facility, respectively, when irradiating fresh target arrays as those herein described.

Important to note is that in this study we discovered that small differences in assumptions could affect the production rates of Pu-238 observed. The exact flux at a specific target location can have a significant impact upon production, so any differences in how the control elements are modeled as a function of exposure, will also cause differences in production rates. In fact, the surface plot of the large VXF target Pu-238 production shown in Figure 3 illustrates that the pins closest to the core can potentially have production rates as high as 3 times those of pins away from the core, thus implying that a cycle-to-cycle rotation of the targets may be well advised.

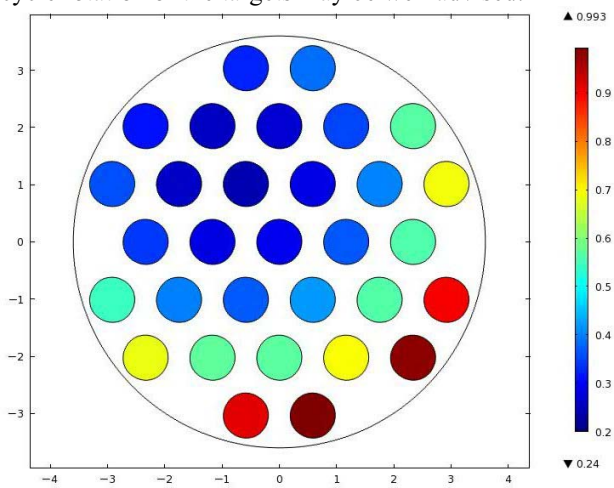

Fig. 3. Spatial dependency of ^{238}Pu production in large VXF targets. Axes denote location in cm, color scale denotes grams of Pu-238 produced per target rod.

A methodology for generating spatially-dependent, multi-group self-shielded cross sections and flux files with the KENO and CENTRM codes has been created so that standalone ORIGEN-S inputs can be quickly constructed to perform a variety of ^{238}Pu production scenarios, i.e. combinations of the number of arrays loaded and the number of irradiation cycles. The studies herein shown with VESTA and TRITON/KENO will be used to benchmark the standalone ORIGEN.

REFERENCES

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