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# Increasing transcurium production efficiency through directed resonance shielding

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ABSTRACT

The Radiochemical Engineering Development Center at Oak Ridge National Laboratory is the world's leader in production of $^{252}$Cf. This and other heavy actinides are produced by irradiation of mixed curium/americium targets in the High Flux Isotope Reactor. Due to the strong dependence of isotopic cross-sections upon incoming neutron energy, the efficiency with which an isotope is transmuted is highly dependent upon the energy spectrum and intensity of the neutron flux. There are certain energy ranges in which the rate of fission absorptions in feedstock materials is reduced relative to the rate of $(n,\gamma)$ captures. Using a variety of computational models it is shown that by perturbing the flux spectrum, it is possible to alter the net consumption of curium feedstock, as well as the yields of key isotopes for the heavy element research program, such as $^{249}$Bk and $^{252}$Cf. This flux spectrum perturbation is accomplished by means of focused resonance shielding through the use of filter materials. It is further shown that these perturbations can alter the target yields in a significant way, increasing the amount of $^{252}$Cf produced per unit curium consumption by over 40%.

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## 1. Introduction

The Radiochemical Engineering Development Center (REDC) at the Oak Ridge National Laboratory (ORNL) is the world's top supplier of heavy actinides such as $^{252}$Cf and $^{249}$Bk, which are produced by the irradiation of actinide targets in the High Flux Isotope Reactor (HFIR). The current tools and methods in place for modeling this process do not employ optimization evaluations. It is up to the user to choose the target composition and irradiation scheme. The current methods for heavy actinide yield calculations, performed using an in-house Fortran code called TCOMP (Alexander, 2007), use a set of generally unperturbed fluxes that are not dependent upon the composition of the target (TCOMP assumes no thermal flux depression), and a set of two-group cross-section parameters that includes a rather simple resonance self-shielding factor based on isotopic mass. However, these calculations do not take into account the impacts that one nuclide may have on another, or on the physical arrangement of the target materials. While these calculations are useful for predicting yields within an acceptable margin of error given starting masses of the key isotopes, they are not able to identify changes within the transmutation scheme as a result of inter-isotope interactions.

The overall process of producing heavy actinides involves many losses due to decay, fission, or transmutation to other undesirable isotopes. For the case of $^{252}$Cf production, only a primitive optimization method is currently performed prior to an irradiation campaign. This method uses the $^{252}$Cf production rate during each cycle to determine the endpoint of the irradiation when the consumption of $^{252}$Cf equals the production rate. The supply of curium feedstock useful for heavy actinide isotope generation is limited, and efficient use of this material is prudent to preserve materials for future needs.

An initiative has recently been undertaken to analyze the transcurium isotope production process and to identify possible means of optimizing production of the heavy actinides relative to consumption of the curium feedstock. With each irradiation cycle, fission losses reduce the total actinide inventory. However, due to the energy dependence of neutron capture and fission cross-sections, the likelihood of fission per absorption in the target actinide is greater for certain energies of an incoming neutron. Therefore, it is proposed that by depressing the neutron flux present in the actinide targets within these key energy ranges, the total number of fissions per absorption can be reduced (and the total number of $(n,\gamma)$ reactions per fission can be increased), resulting in greater product yield per consumption of feedstock material. The purpose of this work is to use computer modeling to analyze the materials used for transcurium isotope production, identify the areas of primary feedstock loss, and examine the potential to reduce feedstock losses via directed flux spectrum perturbation.

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## 2. Materials and methods

This work attempts to best model spatial and transient behavior of the neutron flux in the actinide target sites of the HFIR in order to identify the effects of the neutron flux on the actinide reactions. This involves using a variety of modeling techniques in an attempt to isolate the primary factors affecting the yields. At different points in the computational process, the modeling technique used is dependent upon the known and desired parameters as well as the accuracy required and the computing time required (or tolerated). Two primary means of modeling the transmutation process have been employed: (1) a full-reactor 3-D Monte Carlo model and (2) a 1-D discrete-ordinates model of the flux trap utilizing a fixed source to represent the flux within the target, combined with a matrix-exponentiation depletion calculation.

### 2.1. Monte Carlo evaluation

In this work the Monte Carlo code KENO-VI (Hollenbach et al., 2009), a module of the SCALE (Bowman, 2011) code package developed by ORNL, was used to develop a 3-D model of the HFIR, including full detail of the flux trap and transcurium target sites.

The inner and outer fuel elements of the HFIR are composed of involute plates in the inner and outer fuel elements containing 15.18 g ± 1% and 18.44 g ± 1% $^{235}$U, respectively. This fuel meat is distributed along the arc of the fuel plate in varying thicknesses, with the remainder of the plate composed of aluminum. The plates are separated by 0.050 in. water gaps. This involute plate geometry results in a fuel–water ratio that varies with the radial distance from the flux trap, as the weight percentages of fuel meat and aluminum vary. In this model the individual plates were not modeled; instead, the fuel region itself was represented by a series of cylinders, with axial and radial layers, each approximating the average fuel-water ratio in that region of the fuel element. The involute fuel plate geometry is shown in Fig. 1 (left), as well as a construct for the fuel as represented by the concentric cylinders modeled in KENO-VI (right).

The flux trap is the central region of the HFIR core, inside the fuel element region. The flux trap region consists of a cylindrical basket and 37 vertical experimental target sites that typically contain heavy actinide curium production targets, solid aluminum dummy targets, shrouded aluminum dummy targets, stainless steel targets, and other experiments. The flux trap is the area of primary interest to this research, as it is the location of the production of heavy actinides.

In the KENO-VI model, the target sites are subdivided into six radial regions per target: the experimental specimen, specimen cladding, target shroud, target site cylinder, coolant between the specimen cladding and target shroud, and coolant between the target shroud and the target site cylinder. The experimental specimen is further divided into two axial regions, one for collection of centerline data and one for the remaining axial region. All of the coolant within the flux trap is homogenized with the structural materials of the flux trap basket and target site cylinders.

The flux trap, as modeled for cycle 400 of HFIR operation, is shown in Fig. 2. The image is cut away at the top, as well as from the front quarter, to show both axial and radial cross-sectional views. The solid cylinders represent targets of either aluminum or stainless steel which have been fully homogenized with the surrounding cladding and moderator, while the multi-region cylinders represent shrouded aluminum targets. The larger cylinders around the periphery of the flux trap are the peripheral target position (PTP) targets, and also represent shrouded aluminum targets (Hogle and Maldonado, 2011).

The control element region consists of two concentric cylinders, the inner control element (ICE) region and outer control element (OCE) region surrounding the fuel. Each element is composed of four radial arc segments comprising a cylinder with small gaps between each radial arc. Each element is also composed of four axial control regions, aluminum at the top and bottom, a black control region composed of aluminum with europium oxide, and a grey control region composed of aluminum with tantalum oxide. The control elements are moved vertically in opposite directions over the course of the reactor cycle, with the ICE moving down and the OCE moving up. This movement reduces the amount of negative reactivity being provided by the control elements for the duration of the cycle.

The control elements were modeled in KENO-VI as two cylinders with four axial regions each. Each axial region was homogenized with the flow through holes and spaces between the radial arcs.

The HFIR contains three reflector regions: the removable, semi-removable, and permanent beryllium reflector regions. The reflector regions are modeled in KENO-VI as a series of concentric cylinders. Although the composition of the beryllium used in each reflector region is identical, the presence of structural materials

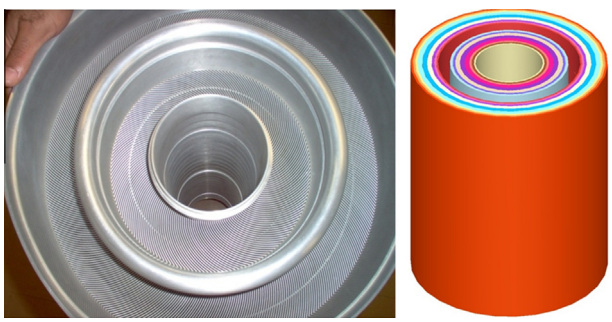

**Fig. 1.** Involute fuel plates of the HFIR core (left) and as modeled in KENO-VI (right).

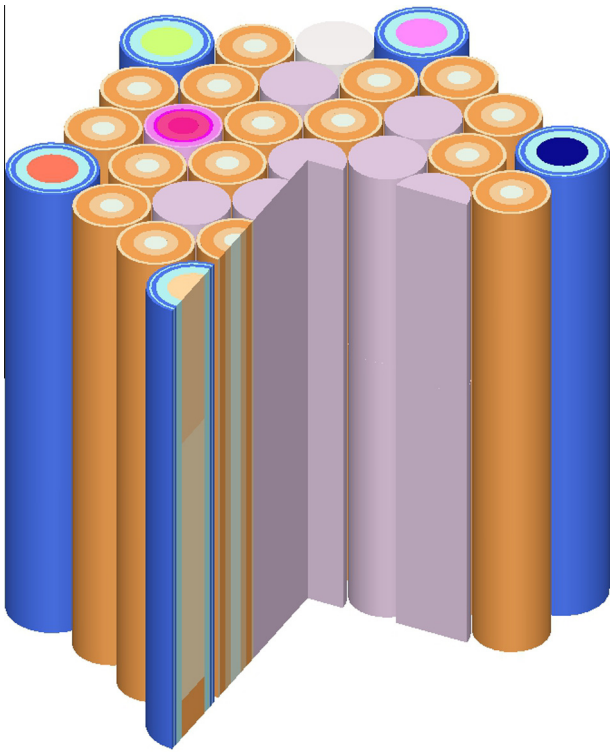

**Fig. 2.** Flux trap as modeled in KENO-VI.