Analysis of Improved ^{232}U Production Through Modification of Neutron Flux Spectra

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INTRODUCTION

^{232}U has been identified by the scientific community as a potential tracer isotope and as a proliferation deterrent for use in uranium-based fuels [1]. As a tracer isotope, ^{232}U can be added to a system, the movement of the ^{232}U can then easily be tracked by its radioactive decay. This is particularly useful when performing quotative uranium radiochemistry analyses, as the alpha particles emitted from ^{232}U have substantially different energies from naturally occurring uranium isotope (^{234}U, ^{235}U, and ^{238}U). ^{232}U also has several decay products, including ^{208}Tl, which emits a 2.614 MeV gamma ray. This gamma-ray tracking technique is particularly useful for security applications of uranium fuel [1].

Extending this to security and nonproliferation, the previously discussed high energy gammas make attempting to conceal weapons material more difficult. Current fuels are much easier to conceal behind lead shielding, making them difficult to locate and track. Secondly for nonproliferation, the addition of ^{232}U helps to hinder enrichment efforts as enriching ^{235}U with traditional methods also results an increased enrichment of ^{232}U, increasing the unattractiveness of the resulting uranium vector [2].

Several current concerns exist for the large-scale implementation of ^{232}U in the uranium fuel cycle and other research and manufacturing applications. One of the primary concerns is the need for an effective production method that can accommodate the scale of ^{232}U needed. Several previous studies have examined potential production with the use of ^{231}Pa and ^{230}Th as potential target materials [1][3][4][5]. Due to the rarity of protactinium, with an average terrestrial concentration in the magnitude of parts per trillion [6], this research has decided to focus on ^{230}Th as our target material.

The method evaluated in this research is the potential use of shielding material or other techniques to filter or shift the neutron spectrum to a more beneficial range for ^{232}U production. This study performs a computational experiment using Oak ridge National Laboratory's (ORNL) High Flux Isotope Reactor (HFIR), or another research reactor.

Figure 1 shows the production path from ^{230}Th to ^{232}U in blue (dashed) highlighting the desired neutron interactions (positive), while in red (solid) the undesired interactions (negative) is also presented. This figure is a simplified example intended to demonstrate the various interactions that can occur and some of the interactions we wish to limit with filtering material.

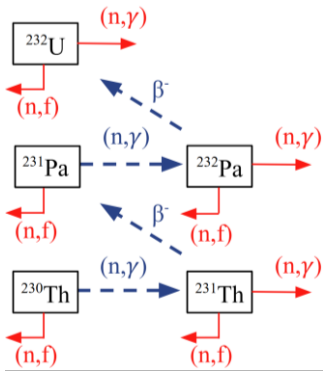

Figure 1. Production path for ^{232}U from a ^{230}Th target.

PRODUCTION CROSS SECTIONS

The first step taken to evaluate shielding material was to examine the neutron energy dependent cross sections for each interaction that leads to ^{232}U. To do this, ENDF/B VIII.0 [7] data was imported into Excel using the JANIS online tool [8]. An average was taken of the cross sections for positive interactions, those that lead to ^{232}U, and negative interactions, those that lead away from ^{232}U, and plotted in Figure 2.

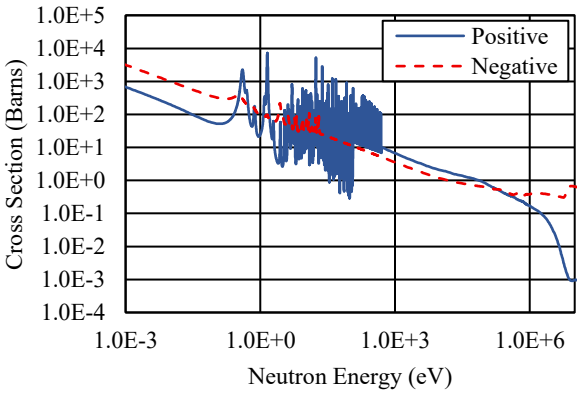

Figure 2. Positive and negative neutron cross sections.

Figure 2 shows the areas in which the average for the positive interactions approaches or overtakes the negative interactions will be where the most ^{232}U is produced. From this we speculated that the best region for neutron energies to produce ^{232}U would be the epithermal region to the lower end of the fast region. In the figure this correlates to an energy range of roughly 0.5 to 100,000 eV. This observation serves as a basis to select neutron filter and shielding materials.

NEUTRON FILTERS AND SHIELDS

Monte Carlo N-Particle 6.3 (MCNP) code [9] was used along with a MCNP model of HFIR provided by ORNL [10] to evaluate the flux profile inside of the target facilities. Several experiment facilities exist, and several were examined during this project with the most time dedicated to location D4. Figure 3 is taken from the HFIR user guide [11] and shows the location of D4 in the center of the flux trap region. It is important to note that D4 was chosen at first due to its relatively high particle flux helping to improve simulation uncertainty and acts as a proof of concept. For the planned experiments in HFIR, one of the six large removable beryllium facilities, seen as 6, 14, 16, 17, 19, and 21 in Figure 3, will likely have to be used to accommodate the required shielding thicknesses.

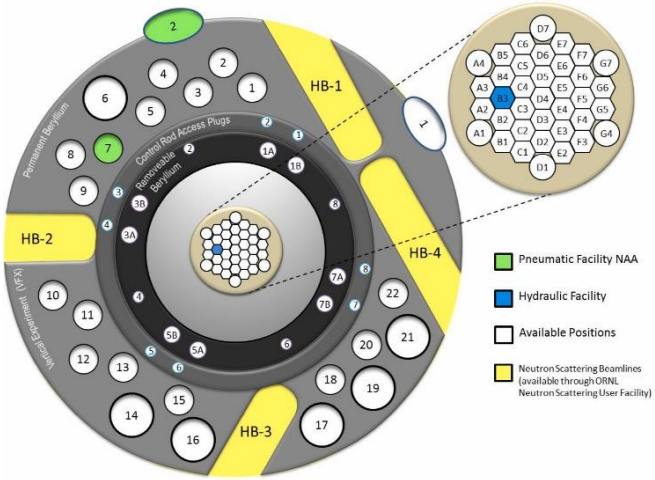

Figure 3. Experimental locations in HFIR. [11]

An initial flux tally was performed on the D4 irradiation facility and shown in Figure 4 where it has been plotted (in green) alongside the positive and negative interaction data from Figure 2.

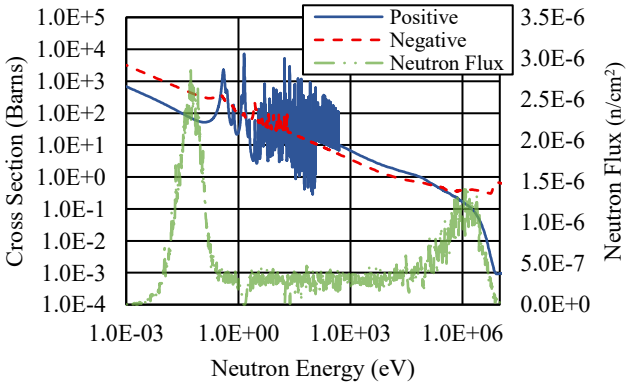

Figure 4. Initial unfiltered target flux.

Based on the Figure 4 we determined two areas of concern to be addressed with filtering materials. First the thermal peak can be seen as being in an area where the negative cross section is much higher than the positive cross section, and secondly a similar problem can be seen for the fast peak.

To address these concerns filter materials were first tested to eliminate the thermal peak. For this cadmium and gadolinium were both used with varying thickness until gadolinium with a thickness of 1 mm was chosen for its superior filtering capacity. Figure 5 shows the target flux with the added gadolinium shielding.

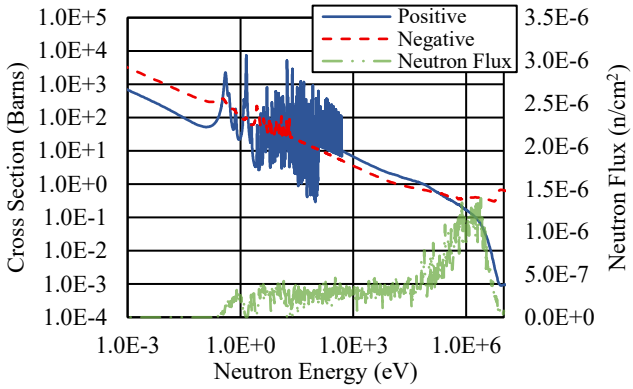

Figure 5. Gadolinium (1 mm) shielded target flux.

To limit the fast flux, a moderator was introduced as an additional shielding layer added before the gadolinium. Several moderators were simulated including H_2O, D_2O, high density polyurethane, boron, beryllium and graphite all at varying thicknesses. Of all the moderators tested, 10 cm of graphite was found to be the most promising. Not necessarily because of its superior moderating ability but instead because it seems to have had the strongest effect for shifting the thermal flux peak up in energy. Figure 6 shows the neutron flux for the graphite and gadolinium combined shielding.

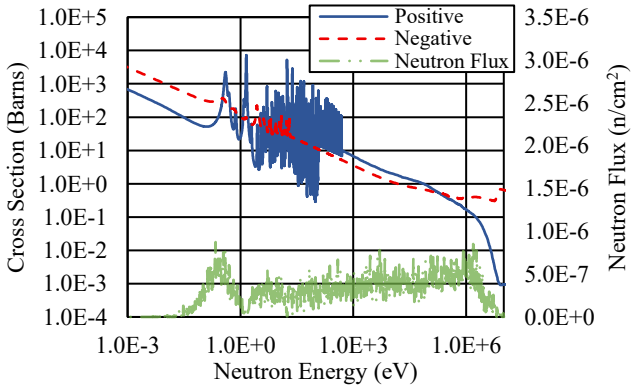

Figure 6. Graphite and gadolinium shielded flux.

Upon investigating the thermal peak shift, attributed to the addition of the graphite, we determined that the effect was caused by absorption hardening. Further simulations were run with just graphite as shielding with varying simulating

temperatures from 0.1 K to 2500 K. The resulting flux from these simulations can be seen in Figure 7.

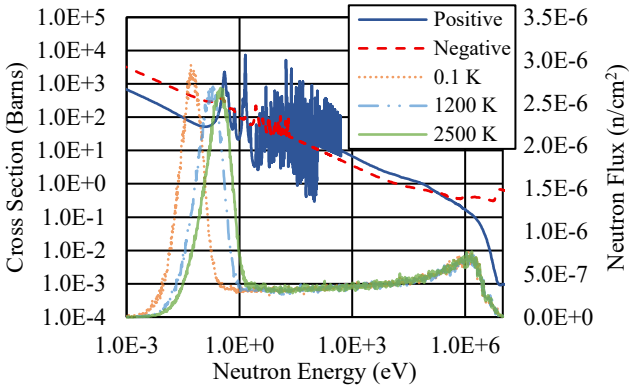

Figure 7. Graphite shielded flux at various temperatures.

From these results we can see that the temperature change has an effect on the flux profile and seems to shift the thermal peak into a region of a high resonance peak for the positive interactions. At 0.1 K we see that compared to the initial flux in Figure 4 that no peak shift occurred.

To evaluate production performance, an energy-averaged cross section was calculated based on each of the flux profiles for the different shielding types. To do this a weighted average was taken of the cross section based on the flux at each energy bin. This was done for both the positive and negative interactions. Of importance in these calculations is the magnitude of the positive cross sections and the difference in positive and negative cross sections. The magnitude will affect the rate of production of ^{232}U while the difference in positive and negative cross sections should affect the amount of ^{232}U produced per gram of target material consumed. Table I summaries the calculated average cross section for each of the discussed shielding materials.

Table I. Energy-averaged cross section with various shielding materials.

Shielding material	Positive (barns)	Negative (barns)	% Positive
No Shielding	57.6	180.7	24.2%
Gd	20.2	86.4	19.0%
Graph. + Gd	59.0	111.7	34.5%
1200 K Graph.	111.3	167.1	40.0%
2500 K Graph.	145.5	154.2	48.5%

BURNUP OF THORIUM TARGETS

Based on the above shielding designs, MCNP's burnup capability was activated to simulate the irradiation of a thorium target in HFIR. The burnup simulates neutron irradiation based on the given power level of HFIR and keeps track of changing material composition as the simulation progresses. Simulations were performed on several of the above-mentioned shielding materials. Each target started with 1 gram of ^{230}Th and was burned for 30 days in HFIR, roughly the length of one operating cycle for the reactor.

To gauge the improvement of adding shielding materials, several metrics were examined. Of these metrics, the first is simply the amount of ^{232}U produced. Each sample was simulated for the same 30-day period so the amount of ^{232}U at the end of those 30 days tells us how the shielding material affects our rate of ^{232}U production. The second metric is the production efficiency (η) which measures the amount of ^{232}U produced per gram of target material consumed, as seen in Eq.(1). This gives us an idea of how cost effective our production method is as ^{230}Th is a relatively expensive target material.

$$\eta = \frac{\text{Mass}_{\text{U,232,Finial}}}{\text{Mass}_{\text{Th,230,Initial}} - \text{Mass}_{\text{Th,230,Finial}}} * 100\% \qquad (1)$$

The last metric used is ^{232}U purity. This measures how much of the resulting uranium produced is ^{232}U, as seen in Eq.(2). This metric is important as other forms of uranium, specifically ^{233}U, may have undesired effects when added as a tracer material.

$$^{232}\text{U Purity} = \frac{\text{Mass}_{\text{U,232}}}{\sum_i^n \text{Mass}_{\text{U,i}}} * 100\% \qquad (2)$$

Table II summaries the results of these metrics for each of the shielding materials.

Table II. Burnup results with various shielding materials.

Shielding material	Mass ^{232}U (gram)	Production Eff. (η)	^{232}U Purity
No Shielding	1.3E-02 ± 3.2%	13% ± 0.5%	91.5% ± 0.3%
Gd	1.0E-03 ± 10%	6% ± 0.6%	98.8% ± 0.3%
Graph. + Gd	1.1E-03 ± 11%	7% ± 0.9%	98.9% ± 0.5%
1200K Graph.	1.7E-02 ± 1.9%	24% ± 0.7%	95.0% ± 0.2%
2500K Graph.	2.0E-02 ± 1.5%	29% ± 0.1%	96.3% ± 0.1%

As sown in Table II, the 2500 K graphite shielding condition led to the highest ^{232}U mass yield and the best production efficiency at 29%, while maintaining a high isotope purity at 96.3%. Notably, even though the gadolinium and graphite offered the highest purity at 98.9%, its overall yield and efficiency were substantially lower. The combination of graphite and gadolinium indicates that thermal suppression must be balanced with spectrum shifting to optimize production.

CONCLUSIONS

In this study, various neutron filtering configurations were evaluated for their effectiveness in enhancing ^{232}U production from a ^{230}Th target with MCNP. The initial

unfiltered flux graph revealed significant thermal and fast peaks, both of which corresponded to energy regions dominated by negative interaction cross sections. Gadolinium shielding effectively suppressed the thermal flux but also significantly reduced the magnitude of positive interactions, as reflected by the drop in the energy-averaged cross section to just 20.2 barns seen in Table I. When graphite was added as a moderator, the thermal peak was shifted up into an energy range where positive cross sections dominate. This shift, most notably observed at 2500 K, yielded the highest energy-averaged positive cross section at 145.5 barns, with a favorable positive-to-negative ratio of 48.5% seen in Table I.

These improvements in cross section values directly translated into improved thorium burnup results as indicated by Table II. The correlation between energy-averaged cross sections and burnup outcomes confirms that strategic spectrum tailoring using temperature-moderated graphite offers a promising path forward for scalable ^{232}U generation. These findings highlight the importance of both neutron flux shaping and cross-sectional targeting in nuclear isotope production design.

ACKNOWLEDGEMENTS

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