Verification of the ^{238}Np Fission Cross Section for HFIR ^{238}Pu Production

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INTRODUCTION

An ongoing investigation to verify the accuracy of the published ^{238}Np fission cross section using neutron activation analysis (NAA) is being conducted by the NAA Lab at the Oak Ridge National Laboratory's High Flux Isotope Reactor (HFIR). It has been speculated that the ^{238}Np fission cross section could be overestimated by as much as 50%. Assessment of the published literature [1, 2] indicates that variations in the thermal and resonance fission cross sections could be as high as 20% and 40%, respectively. These disparities are likely due to the fact that the measurement of ^{238}Np, a transient isotope in the ^{238}Pu production chain, offers inherent complexities associated with measuring the ^{238}Np fission cross section. The ^{238}Pu production pathway and other related reactions are depicted in Figure 1. Because ^{238}Np undergoes transmutation, fission, and decay during neutron irradiation of ^{237}Np, the modeling of its activity depends on many factors besides the fission cross section. Additionally, cross section resolution using gamma spectroscopy of the ^{239}Np, ^{238}Pu, and ^{239}Pu is made more difficult by the relatively high ^{238}Np radioactivity. Nevertheless, measurement of ^{238}Np and its fission products offers significant insight into the relative accuracy of the modeled system, including the fission cross section.

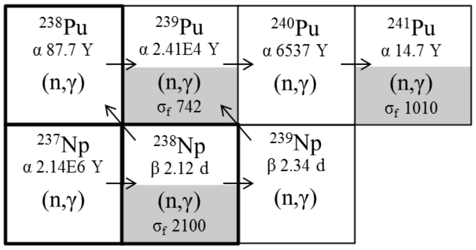

Fig. 1. Reactions associated with ^{238}Pu production.

Crucial isotope production missions at HFIR could be significantly impacted depending on the cross section value used in the neutronics analysis [3] for fission heat generation. Reducing uncertainties in the ^{238}Np fission cross section through experimental measurements could safely increase efficiency of ^{238}Pu production in HFIR. Therefore, the objective of this work is to investigate ^{238}Np and its fission products produced in the Pneumatic Tube (PT) facility of HFIR to assess the agreement with nuclear transmutation modeling codes.

THEORY

The initial evaluation of the ^{238}Np cross section data depends on the comparison between simulated and measured nuclides. Nuclear codes utilize cross section data to model neutron interactions during target irradiations. Modeling codes COUPLE + ORIGEN-S (part of the SCALE modeling suit) [4] provide an accurate computer-generated nuclide portfolio for an irradiation, given that the published cross section and decay data are correct. Due to the well-characterized modeling inputs for the pneumatic tube in the NAA facility, direct comparison between these simulations and measured ^{238}Np in the target after neutron bombardment would reveal any major contradictions in the production or removal cross section data for ^{238}Np. Additional analysis between the modeled and measured fission products produced would reveal potential discrepancies associated with the fission cross section data. Knowing the approximate bias associated with the ^{238}Np capture and fission cross section data would impact the certainty of the HFIR irradiation safety analysis for ^{238}Pu production.

METHODOLOGY

The NAA lab at HFIR has access to a neutron flux in its PT that is similar to that experienced at the ^{238}Pu production target positions. The PT has a thermal flux of about $4.5\text{x}10^{14}$ n/cm^2-s and a total flux of about $6.5\text{x}10^{14}$ n/cm^2-s. Since the large thermal flux quickly generates significant fissions and transmutations in small target samples, a higher certainty can be assigned to specific reaction pathways. Furthermore, the NAA lab has a large shielded cubicle, radiochemical processing hoods, and several High Purity Germanium (HPGe) detectors for sample staging, processing, and measurement. These facilities allow for strategic irradiations and measurements to occur with relative ease and efficiency.

Pneumatic Tube Irradiations

Multiple irradiations were conducted to sufficiently study the production and removal cross sections associated with ^{238}Np. Low fluence irradiations highlighted the ^{237}Np(n,γ)^{238}Np reaction, and helped to verify the accuracy of the data used for the ^{237}Np capture cross section. Higher fluence irradiations were used to examine data incorporated in the ^{238}Np(n,γ)^{239}Np and ^{238}Np(n,f)FP reaction pathways. The target material consisted of a low mass, dilute ^{237}Np wire. Starting ^{237}Np mass was determined by measuring ^{233}Pa activity through gamma spectroscopy, which is an alpha decay product in secular equilibrium with ^{237}Np. Table I shows the starting target mass of each sample as well as the total fluence received by each sample. The flux for Samples 1 and 2 was measured by simultaneous irradiation of a flux monitor. The irradiation time for Samples 3 and 4 was too long to facilitate co-irradiation of a flux monitor. Instead, the flux was measured before and after irradiation with dilute aluminum foils containing manganese and gold.

Table I. Target and irradiations details.

Sample #	^{237}Np mass (ug)	Irradiation Time (s)	Total Fluence on Sample (n/cm^2)
1	38.11	2[a]	1.34E+15
2	13.03	20	1.11E+16
3	10.70	3600	2.31E+18
4	9.41	180000	1.16E+20

[a] *Irradiation time was 20 seconds at 10% reactor power which is equivalent to 2 seconds.*

HPGe Measurements

Measurements of the ^{238}Np and fission product activities after irradiation in HFIR were completed using a well-calibrated position 30 cm away from the end cap of an N-type HPGe detector having 44% relative efficiency. The irradiation of ^{237}Np produced a large quantity of ^{238}Np activity when exposed to high neutron fluences, necessitating a lengthy decay before counting to reduce detector dead time. For all samples, the 984 keV gamma ray of ^{238}Np was measured until reasonably low counting statistics were reached, usually less than one hour. The count rate associated with the measured peak along with the detector efficiency and branching ratio was used to calculate the activity of ^{238}Np at the end of bombardment. The Canberra Genie-2K software and Lynx digital signal analyzer were used to obtain all gamma spectra.

COUPLE/ORIGEN Modeling

Modeled results were generated using COUPLE in conjunction with ORIGEN-S (ENDF/B-VII.0 cross section data) to simulate irradiation, decay, and measurements of the targets by applying a known 238-group neutron spectrum for the HFIR PT-1. COUPLE takes a 238-group cross section library and uses the flux spectrum as weights to generate a 1-group cross section for ORIGEN. ORIGEN uses the created 1-group cross sections and simultaneously calculates concentrations or activities for the generation or depletion of a large number of isotopes by neutron transmutation, fission, and decay. The ENDF/B-VII.0 cross section library associated with SCALE version 6.1 was used in the modeling analysis. Published cross section data [5] and formulations for calculating the energy dependent cross sections [6, 7] of ^{238}Np were implemented to produce the cross section data in ENDF/B-VII.0. Primarily, the experiments conducted in this work were used to evaluate reaction pathways predicted by the ENDF/B-VII.0 ^{238}Np cross section data.

RESULTS AND DISCUSSION

To establish a maximum neutron irradiation time needed, a determination was made to ensure that the majority of the measured fission products resulted from ^{238}Np fission. Using collapsed 1-group neutron cross sections produced in ORIGEN for the HFIR PT-1 position to calculate the activation and depletion of several isotopes associated with ^{238}Pu production, it was found that fissions resulting from ^{238}Np become the majority after only 25 minutes in PT-1. As shown in Figure 2, after 50 hours (3000 min.) of irradiation, the ^{238}Np fissions account for more than 99% of the total fissions, indicating that the bulk of measured fission products come from ^{238}Np. As irradiation time continues past 50 hours, ^{238}Np activity approaches saturation, and the fission rate from products of decay (Pu species) begins to impact the fission products produced. Therefore, an irradiation time of 50 hours was used in the measurement of the fission products.

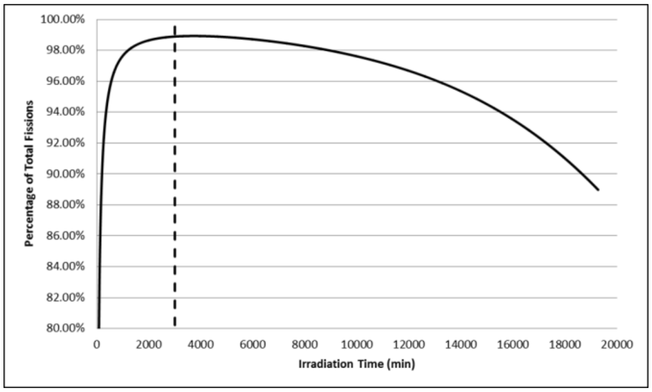

Fig. 2. Percentage of total fission from ^{238}Np.

After quantification of ^{238}Np in the sample at the end of irradiation, the simulated mass of ^{238}Np was compared to the experimentally measured value, and indicated a recovery error less than 1.5%. Table II shows details of the ^{238}Np recovery comparisons for all samples irradiated. While the recovery error increases slightly with increasing neutron fluence on the sample, the recovery error was still reasonably low. Therefore, it appears there was not a significant bias in the cross section data that encompasses the production and removal of ^{238}Np. However, these measurements address the combined effect of fission, decay, and capture reactions of ^{238}Np, so small ^{238}Np recovery errors do not necessarily imply accuracy of the fission cross section. Fission products were measured in Sample 4 (50 hrs. irradiation) to assess any differences in the fission cross section data.

Table II. ^{238}Np recovery comparisons.

Sample #	Simulated ^{238}Np Mass (μg)	Measured ^{238}Np Mass (μg)	Simulated % Recovery Error
1	6.328E-6	6.365E-6	0.58%
2	2.337E-5	2.344E-5	0.32%
3	3.070E-3	3.109E-3	1.23%
4	9.212E-2	9.339E-2	1.36%

Gamma spectroscopy of fission products after 50 hours of irradiation revealed recovery errors of less than 5% for several fission products when compared to ORIGEN modeled production. Recovery for other measurable fission products ranged within 20% of the measured value. Some of the worst recoveries of the fission product isotopes were from volatile iodine and therefore expected to be low. Table III below shows the recovery for the measurable fission products. The overall recovery errors of non-volatile fission products was relatively low implying that the fission cross section data may only have slight deviations from actual. Yet, there remains a large uncertainty in ^{238}Np fission product yield data used in the modeling for some fission products, such as ^{103}Ru, that could affect recovery error.

Table III. Recovery error of measured fission products.

FP Isotope	Photopeak Energy (keV)	Simulated % Recovery Error
^{140}La	1596.2	2.45%
^{95}Nb	765.8	3.86%
^{95}Zr	756.7	2.65%
^{95}Zr	724.2	6.61%
^{132}I	667.7	-12.95%
^{131}I	636.99	-8.55%
^{132}I	630.2	-20.03%
^{103}Ru	610.3	18.26%
^{140}Ba	537.3	-5.43%
^{103}Ru	497.1	12.45%
^{140}La	487	-1.63%
^{131}I	364.5	-12.51%
^{140}La	328.8	-4.42%

CONCLUSION AND FUTURE WORK

These preliminary results using a comparison method give good initial insight into the behavior of the ^{238}Np cross section. The results suggest that the actual cross section of ^{238}Np is most likely not significantly different from the cross section used in ENDF/B-VII.0. However, there exists a large potential for additional work to refine published cross section values. Accordingly, safety and production calculations that utilize nuclear transmutation codes employing ^{238}Np cross section data for production of ^{238}Pu have to be relied upon with measured assurance until more analysis is completed. The recently published ENDF/B-VII.1 libraries have increased the capture and fission cross sections for ^{238}Np by roughly 6.5% to match the cross section published in JENDL 4.0, based on an update from JENDL 3.3 to improve the resolved resonance integral parameters evaluated at JAEA [8]. Further analysis of a replicate set of measurements will be used to confirm the reported results along with additional irradiations using sealed targets to capture volatile fission products. With this continued work, the HFIR can have increased certainty in the accuracy of ^{238}Np fission cross section.

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