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CONTENTS

ABBREVIATIONS

ACTS	Assessment and Commitment Tracking System
ADVANTG	Automatic Variance Reduction Generation
CENDL	Chinese Evaluated Nuclear Data Library
ENDF	Evaluated Nuclear Data File
HFIR	High Flux Isotope Reactor
HFIRCON	High Flux Isotope Reactor Controller
JANIS	Java-Based Nuclear Data Information System
JEFF	Joint Evaluated Fission and Fusion
JENDL	Japanese Evaluated Nuclear Data Library
LAVAMINT	LAVA Model Interrogator
MCNP	Monte Carlo N-Particle
ORIGEN	Oak Ridge Isotope Generation
ORNL	Oak Ridge National Laboratory
TENDL	TALYS-based Evaluated Nuclear Data Library

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ABSTRACT

This study is being completed in response to an assessment and commitment tracking system (ACTS) item that was opened during the production of ^{166m}Ho. To satisfy the ACTS action item, cross section data of ^{166m}Ho has been evaluated. Future ^{166m}Ho production campaigns will employ the latest cross section data.

The isotope ^{166m}Ho is a good calibration source for gamma spectroscopy because of its wide range of gamma energies and its long half-life. As interest in ^{166m}Ho increases, the need to accurately calculate the production yields will also increase. A key factor that contributes to inaccurately predicting the yields is outdated cross section data used in calculations. With the advancement of technology, more precise cross section data evaluation methods become attainable, thus allowing for more accurate predictions of the production yields.

The expected production yield of ^{166m}Ho was calculated and compared with the experimental yield activity from three ^{165}Ho targets that were irradiated in the High Flux Isotope Reactor (HFIR) during cycles 489 and 490A. MCNP-ORIGEN (Monte Carlo N-Particle–Oak Ridge Isotope Generation) and HFIRCON (HFIR Controller) codes were used to calculate the theoretical yield of ^{166m}Ho. The yield was calculated using various cross section libraries.

1. BACKGROUND

The metastable state of ^{166}Ho has a half-life of 1,200 years while the ground state only has a half-life of 26.824 hours. This makes ^{166m}Ho a good candidate for long-term applications. One method of producing ^{166m}Ho is by irradiating natural holmium in the High Flux Isotope Reactor (HFIR). Holmium is a monoisotopic element and naturally exists only as ^{165}Ho. The following nuclear reaction in Equation 1 shows the production route of reactor-produced ^{166m}Ho:

$$^{165}\text{Ho} \xrightarrow{(n,\gamma)} {}^{166}\text{Ho}\ {}^{166m}\text{Ho}\ . \#(1)$$

1.1 CROSS SECTION DATA

Table 1 shows the thermal neutron cross section data for reactions on the Ho isotopes of interest for this work. The Atlas of Neutron Resonances[1] cross section data are based on experimental data. The Joint Evaluated Fission and Fusion File 3.0/A (JEFF-3.0/A)[2] is the default reaction rate library used by ORIGEN. The TALYS-based Evaluated Nuclear Data Library (TENDL) 2019[3] is derived from the TALYS nuclear model code. The Evaluated Nuclear Data File (ENDF)[4] is from the United States and is the default low energy nuclear data employed by MCNP. ENDF/B-VIII.0 is the latest release of the ENDF data library. Since the cross section values for holmium did not change within ENDF/B-VIII.0, ENDF/B-VII.1 was used in the study.

The ^{165}Ho(n, γ)^{166}Ho reaction has a cross section that is over 17 times greater than the ^{165}Ho(n, γ)^{166m}Ho reaction (Table 1), so ^{166}Ho is the main isotope that is created. Because of the high-burnup cross section of ^{166m}Ho, the yield is reduced dramatically if it is irradiated for long periods of time.

Table 1. Thermal neutron cross section data for holmium

	Mughabghab (b) [1]	JEFF-3.0/A (b) [2]	TENDL 2019 (b) [3]	ENDF/B-VII.1 (b) [4]
^{165}Ho(n, γ)^{166}Ho	60.9 ±1.0	66.59	64.31	64.68
^{165}Ho(n, γ)^{166m}Ho	3.5 ±0.4			
^{166}Ho(n, γ)^{167}Ho	—	48.95	3,588.16	—
^{166m}Ho(n, γ)^{167}Ho	10,778 ±600	3163.75	10,796.10	3,600

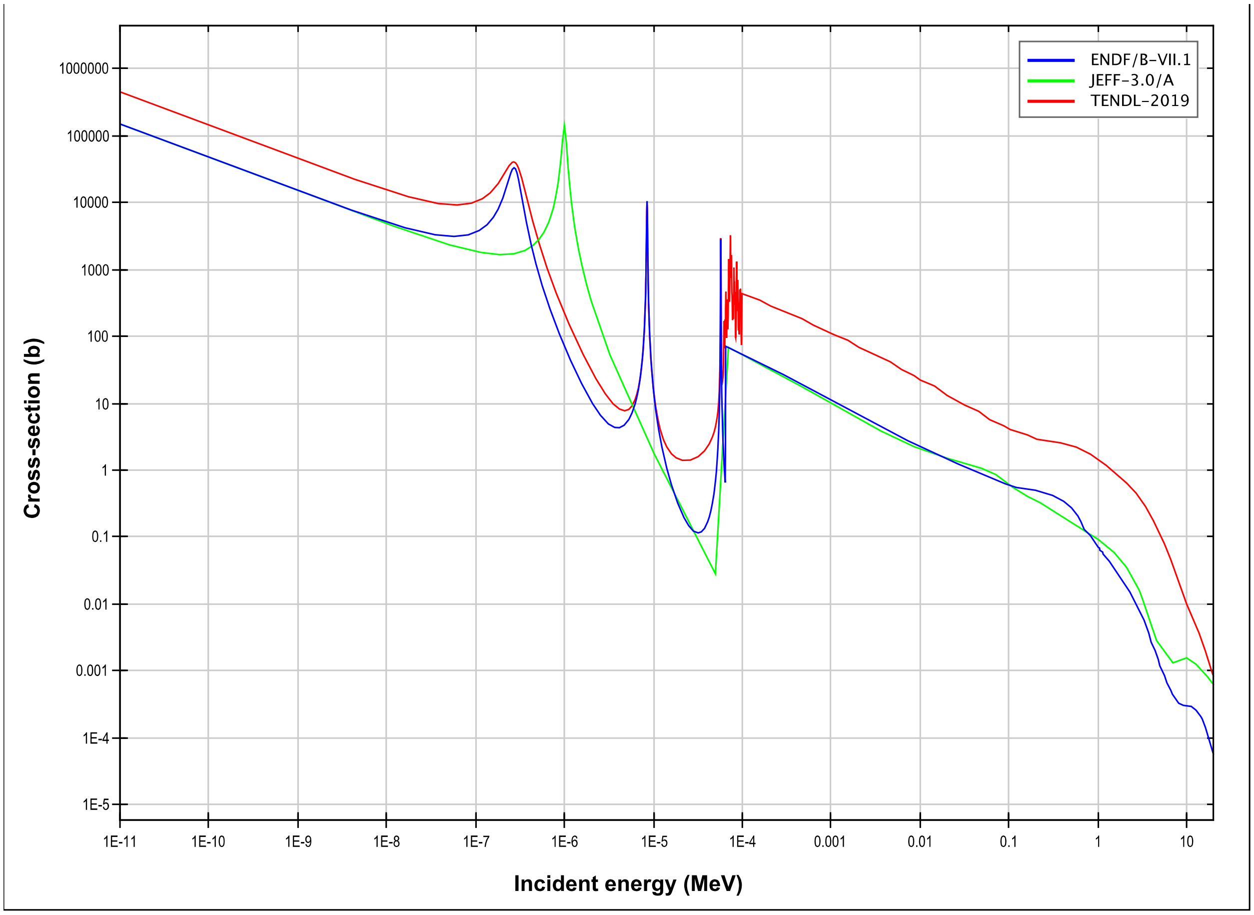


Figure 1. TENDL 2019 vs. ENDF/B-VII.1 vs JEFF 3.0/A cross section burnup comparison for ^{166m}Ho.

Figure 1 shows the comparison of ^{166m}Ho burnup cross section data for the TENDL 2019, JEFF-3.0/A and ENDF/B-VII.1 libraries. Figure 1 was created using Java-Based Nuclear Data Information System (JANIS) [5].

[1] Mughabghab, *Atlas of Neutron Resonances. Resonance Parameters and Thermal Cross Sections*.
[2] Sublet et al., “The JEFF-3.0/A Neutron Activation File - New Features, Validation, and Usage.”
[3] Koning et al., “TENDL: Complete Nuclear Data Library for Innovative Nuclear Science and Technology.”
[4] Chadwick et al., “ENDF/B-VII.1 Nuclear Data for Science and Technology: Cross Sections, Covariances, Fission Product Yields and Decay Data.”
[5] Soppera, Bossant, and Dupont, “JANIS 4: An Improved Version of the NEA Java-Based Nuclear Data Information System.”

1.2 IRRADIATION EXPERIMENTS

Using the assembly drawing JLM06272008-1 Rev. 0, three standard finned rabbits (drawing N3E020977A036 Rev. 0) were loaded with three holmium rings each. Table 2 is a summary of the loaded rabbit assemblies. The three rabbits were irradiated for approximately 7 days each in the center line position of the hydraulic tube (position B3-5) of HFIR during cycles 489 and 490A.

Table 2. Holmium rabbit loading summary

Rabbit #	Pellet ID	Placement	Outer diameter (mm)	Length (mm)	Mass (g)
NM-911	2C	Top	6.46	49.99	6.690
	2D	Middle	6.44		
	2F	Bottom	6.33		
NM-915	3A	Top	6.44	49.50	6.697
	3B	Middle	6.44		
	3C	Bottom	6.38		
NM-916	2E	Top	6.43	49.56	6.671
	3D	Middle	6.41		
	3E	Bottom	6.33		

A combination of MCNP6 (neutron transport) and ORIGEN (activation and decay) with the default ORIGEN cross section library (JEFF-3.0/A) was used to estimate the expected ^{166m}Ho specific activity of 1.32 mCi/g. After irradiation, the targets were dissolved in concentrated nitric acid, sampled, and analyzed using high purity germanium (HPGe) gamma-ray spectroscopy. Three different measurements were completed on a HPGe detector. The results showed that a total of ~12.32 mCi of ^{166m}Ho was produced from the three targets for a total specific activity of ~0.61 mCi/g ^{166m}Ho. Because a lower specific activity was observed during the processing of the ^{166m}Ho, an ACTS item was opened to investigate the discrepancy between the predicted vs. experimental results.

2. SIMULATION METHODS

HFIRCON (HFIR Controller) and ORIGEN-MCNP6 (Oak Ridge Isotope Generation/ Monte Carlo N-Particle) simulations were used to study how the cross-section libraries affect the ^{166m}Ho theoretical yield calculations.

2.1 HFIRCON

HFIRCON is a toolkit that is used to automate many HFIR neutronics calculations. Used by the Research Reactors Division at Oak Ridge National Laboratory (ORNL) to qualify materials prior to irradiation in HFIR, HFIRCON consists of a collection of python routines and C plugins that automate the workflow for multi-cycle fuel and target depletion in HFIR. HFIRCON uses the Automatic Variance Reduction Generation (ADVANTG) code package for all the variance reduction calculations, ORNL-TN/MCNP5 for all transport solutions, ORIGEN/MSX_DEPLETE for all the depletion calculations, and LAVA Model Interrogator (LAVAMINT) to stochastically calculate cell volumes and bounding boxes.[6]

[6] Wilson et al., "HFIRCON Version 1.0.5 User Guide."

HFIRCON Version 1.0.5 uses a hybrid of cross section libraries from ENDF/B-VII.0 and ENDF/B-VII.1, with supplemented data from JEFF-3.1.2, JENDL4.0u (Japanese Evaluated Nuclear Data Library), CENDL3.1 (Chinese Evaluated Nuclear Data Library), and TENDL-2013 to ensure reasonable gamma production data [7].

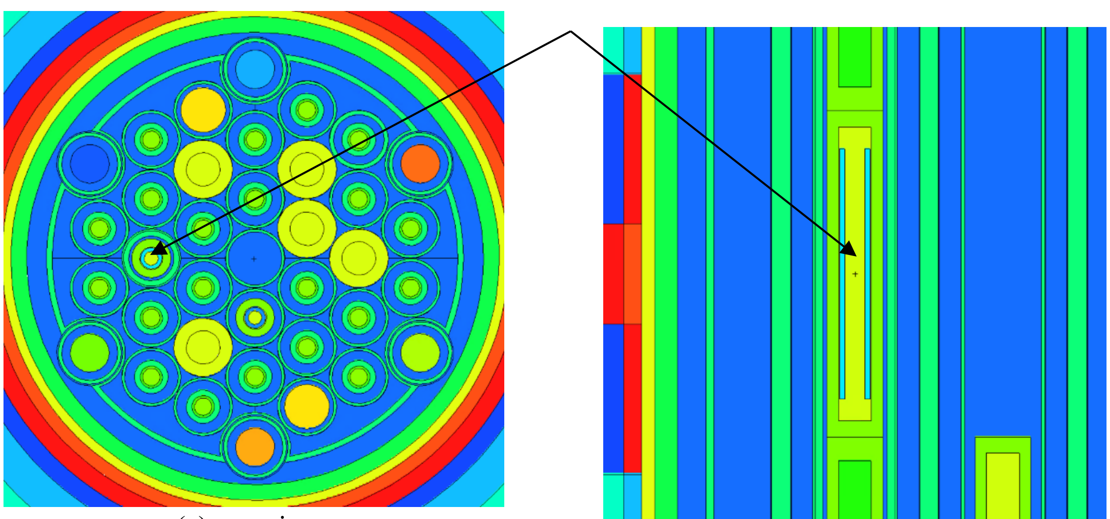
2.2 MCNP6-ORIGEN

In the MCNP6-ORIGEN model a modified version of HFIR cycle 400 run was used to create a 238-group neutron spectrum of the target in the flux trap of HFIR. COUPLE, a nuclear decay and cross section data processing code, was used to create the ORIGEN reaction library by collapsing the 238-group spectrum into a 1-group cross section. COUPLE can also be used to update the cross-section data of any isotope.[8]

In the MCNP6-ORIGEN simulation, the ACE (compressed file format) files for the TENDL 2019 cross section data of ^{165m}Ho were used to create a new 238-group collapsed cross section to investigate how running the ORIGEN calculations with this library compares to running the simulation with ORIGEN's default JEFF-3.0/A data library.

2.3 MCNP MODEL

The base MCNP input file used to model irradiation within HFIR is the highly enriched uranium simplified cycle 400 model [9]. Target NM-911 in the B3-5 position was evaluated. Figures 2(a) and 2(b) show the *x–y* and the *x–z* views, respectively, of the target at the axial centerline of the flux trap. The dark green area is the aluminum rabbit, the yellow area is the helium filler gas, and the light blue area represents the holmium ring(s). For simplicity, the three rings were modeled as one long ring. This MCNP input was used in both the HFIRCON and the MCNP6-ORIGEN simulations.



(a) *x–y* view (b) *x–z* view

Figure 2. Holmium target in position B3-5 (hydraulic tube) of the HFIR MCNP model.

[7] Wilson et al.

[8] Rearden and Jessee, *SCALE Code System*.

[9] Ilas et al., *Modeling and Simulations for the High Flux Isotope Reactor Cycle 400*.

3. SIMULATION RESULTS AND DISCUSSION

Figure 3 is a summary of both the calculated and experimental yield results. Because of the relatively lower (3163.75 b) burnup cross section reported in the default ORIGEN JEFF-3.0/A library, a higher specific activity yield is predicted for ^{166m}Ho. This observation is also shown for the HFIRCON default ENDF/B-VII.1 as well (3,600 b).

The much higher (10,796.10 b) burnup cross section reported in the TENDL 2019 library results in an estimated end of cycle specific activity of 0.556 Ci/g which is much closer to the experimentally determined value of 0.614 Ci/g. It also shows that peak ^{166m}Ho production is reached at 3 days as opposed to the 7-day peak production determined when utilizing the default cross section library.

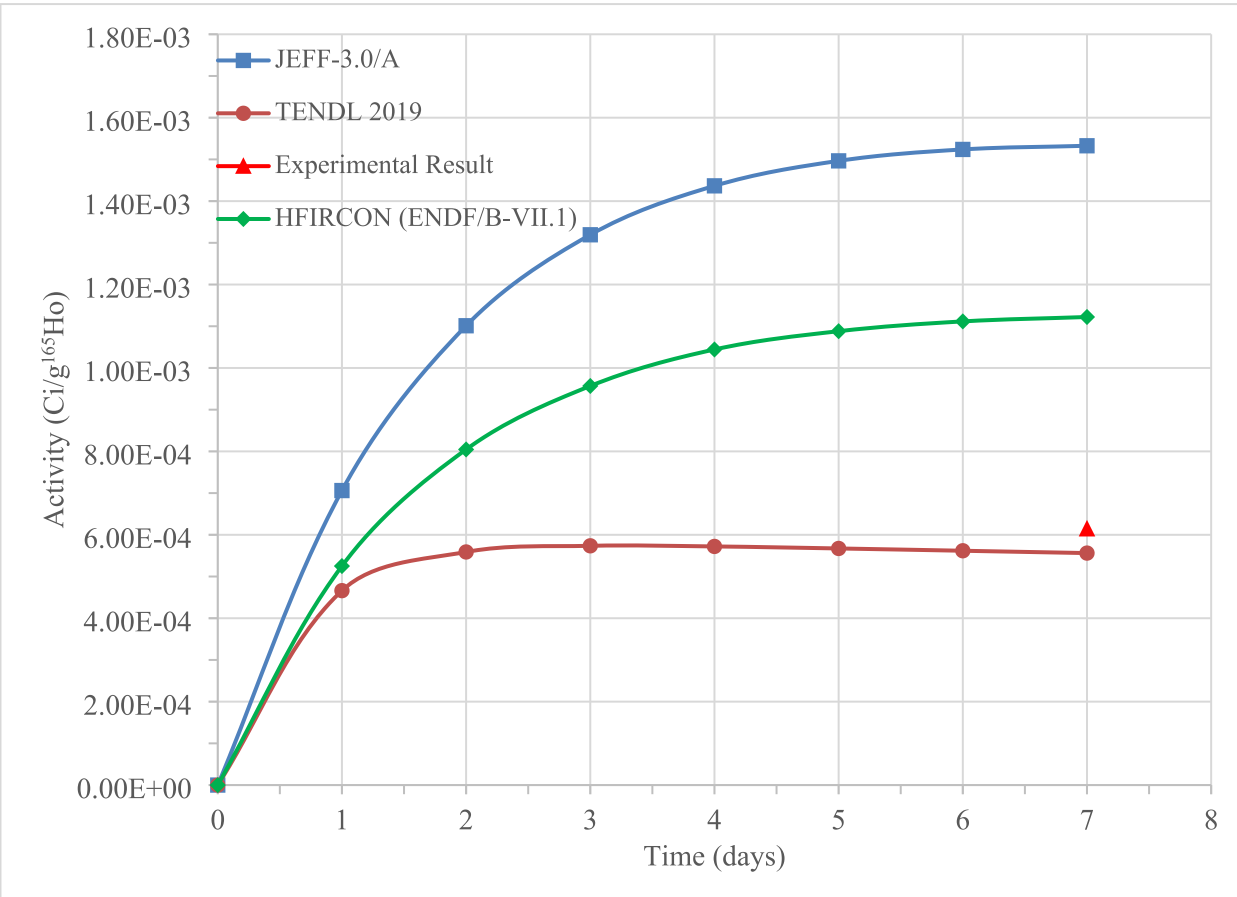


Figure 3. Calculated production of ^{166m}Ho in HFIR. Units are curies per gram of irradiated holmium material.

4. SUMMARY AND CONCLUSIONS

Because the originally estimated specific activity calculation of ^{166m}Ho was much higher than the experimental results. HFIRCON and MCNP6-ORIGEN simulations were run to study and compare how various cross section libraries impact the estimated specific activity of ^{166m}Ho production.

Both MCNP6-ORIGEN with the TENDL 2019 library and HFIRCON yield specific activity estimates closer to the experimental value. The lower-burnup cross section within HFIRCON using the END/B-

VII.1 library for ^{166m}Ho yields a higher production value, whereas the higher-burnup cross section within MCNP6-ORIGEN using the TENDL 2019 library yields a lower production value. It appears that additional experiments would be helpful to confirm the ^{166m}Ho burnup cross section. According to the TENDL 2019 MCNP/ORIGEN simulation, the peak production of ^{166m}Ho occurs at about 3 days of irradiation rather than the originally estimated 7 days. The analysis in this work satisfies the outstanding ACTS action item.

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