

Studies of Past Operations at the High Flux Isotope Reactor

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Introduction

During the past year, two topics related to past operations of the High Flux Isotope Reactor (HFIR) were reviewed in response to on-going programs at Oak Ridge National Laboratory (ORNL). Currently, studies are being conducted to determine if HFIR can be converted from high enriched uranium (HEU) fuel to low enriched uranium (LEU). While the basis for conversion is the current performance of the reactor, redesign studies revealed an apparent slight degradation in performance of the reactor over its 40 year lifetime. A second program requiring data from HFIR staff is the Integrated Facility Disposition Project (IFDP). The IFDP is a program that integrates environmental cleanup with modernization and site revitalization plans and projects. Before a path of disposal can be established for discharged HFIR beryllium reflector regions, the reflector components must be classified as to type of waste and specifically, determine if they are transuranic waste.

End-of-life burnup for HFIR fuel

The HFIR reached full power operation (100 MW) in September of 1966. The fuel cycle length achieved, 2300 MWD, was 40% greater than had been predicted prior to startup of the reactor.¹ The fuel loading and core geometry for HFIR, today, is unchanged from the second production core loaded to the reactor though the maximum approved operating power for the reactor is now 85 MW. In 2005, staff from the Reduced Enrichment for Research and Test Reactors Program (RERTR) contacted ORNL to request studies on the conversion of the reactor to LEU fuel. The end-of-cycle lifetime basis for design of an LEU fuel was selected as 26 days based on the maximum, recent performance achieved (cycles 389 and 397; 2002 and 2003, respectively) and an operating power of 85MW.² Maintaining the current level of reactor performance while converting to LEU leads to the conclusion that reactor operating power must be increased to 100 MW with consequent end-of-life burnup of 2600 MWD.³⁻⁶

The principal concern with increasing the end-of-life burnup of HFIR fuel is the integrity of the fuel clad. Two phenomena impact the clad, buildup and spallation of aluminum oxide on the surface of the fuel and increase in fission product gas inventory in the fuel relative to current and past irradiation exposure. The current methodology for predicting oxide growth⁷ should be valid for either 85 or 100 MW since there is not a strong sensitivity to heat flux and indicates that oxide growth is a function of operating time with the maximum thickness of oxide limited to 75 microns due to spallation. Thus oxide growth in the proposed LEU fuel cycle should not be a concern. For fission gas production, either simulation by computation or experimental data will be needed to resolve safety concerns.

The HFIR cycle length varies over a wide range – in recent history, as low as 1890 MWD for cycle 406 (2006) – depending on the irradiation and isotope production experiments loaded into the reactor. As LEU fuel design studies reach closure for a reference fuel design⁷, HFIR operating data were reviewed to determine the “burnup operational envelope,” i.e. maximum burnup achieved with the current HEU fuel.

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HFIR “end-of-cycle (EOC) packages” were reviewed for EOC burnup values. Values from reactor startup through July 2009 were tabulated and are shown in Fig. 1. The scatter in the data are due to several reasons including: premature shutdowns due to equipment failure or other reasons, changes in experiment loadings in either the central target region or the beryllium reflector, or, as will be discussed below, documentation inconsistencies.

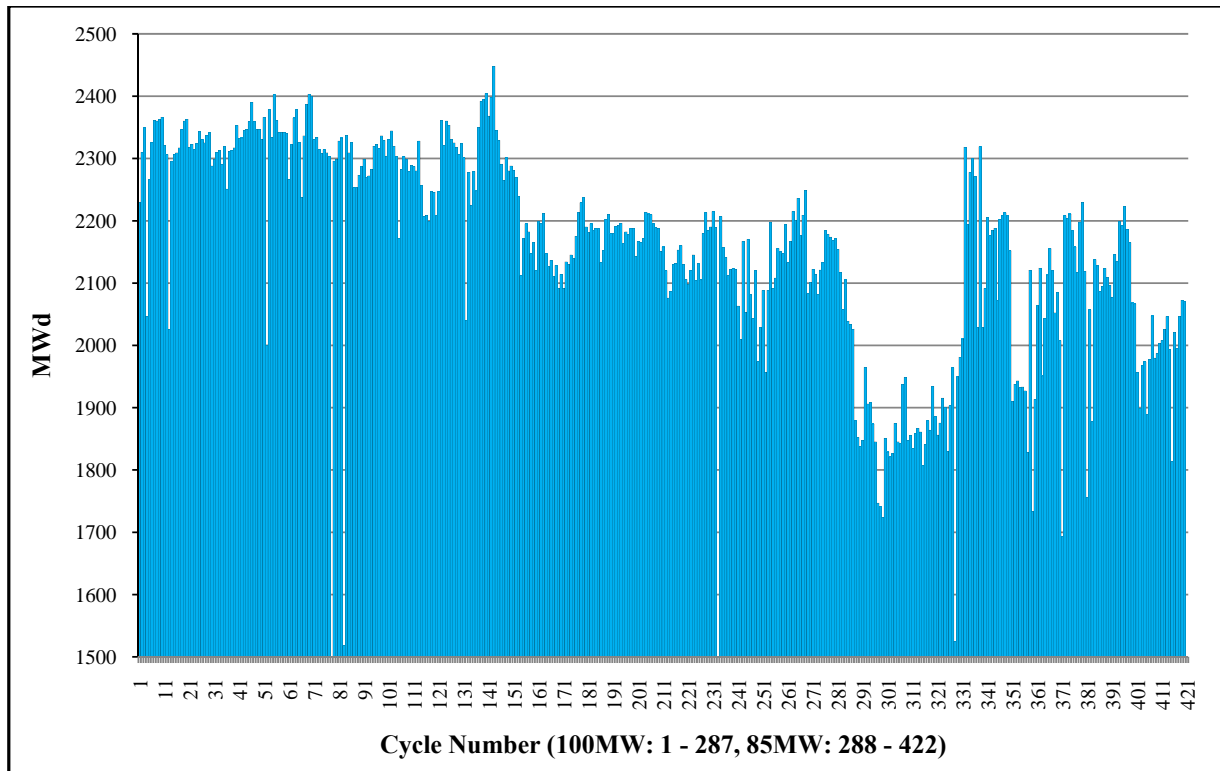


Fig. 1. End-of-cycle exposure for HFIR fuel cycles

Five cycles were reported as achieving burnups greater than 2400 MWD and all of them operated prior to the decrease in power from 100 MW to 85 MW. Two of these cycles (cycles 143 – Feb. 1977 and 267 – June 1985) were reported with burnups close to 2450 MWD, but after further investigation of cycle 267 - reported of reaching 2441 MWD - it was discovered that the core from cycle 266 was reloaded once the core initially loaded in cycle 267 was discharged due to expended fuel. However, after investigating cycle 143, no evidence was found that two elements were irradiated during a single, designated cycle. Operating data shows that cycle 143 operated for 24 days and 11.65 hours at a power of 100 MW with no interruptions. The other cycles that were reported with burnups greater than 2400 MWD include cycles 55, 69, and 140, which were exposed to 2402, 2403, and 2404 MWD, respectively.

Though the purpose of compiling the data shown in Fig. 1 was to assess data/analysis needs for LEU conversion, once viewed as a graphical presentation, a general downward trend in end-of-cycle burnup is observed. Furthermore, three significant but temporary decreases are found at/around cycle periods 287-330, 350-360, and 400-415. One significant increase is observed for the period 332-338.

A simple regression analysis yielded the linear fit shown in Fig. 2. The equation relating the decrease in burnup (y) as a function of cycle number (x) is: $y \text{ (MWD)} = 2353 \text{ (MWD)} - 0.926 \text{ (MWD/cycle \#)} * x \text{ (cycle \#)}$. Investigation of plant records revealed the causes of the three decreases were the insertion of neutron poison filters around selected experiment irradiation locations. Though not the purpose of this study, cycle length reduction provides a basis for the assessment of an economic penalty to a program choosing to sponsor the insertion of a neutron filter in HFIR.

The increase in cycle length for the period 332-338 was investigated and found to be due to the removal of two neutron filters within the removable beryllium reflector. Prior to the startup of cycle 333, two hafnium liners were removed from the removable beryllium reflector and were replaced by aluminum liners. Also, the experiments within these facilities were replaced with beryllium plugs and 11 curium target rods replaced 11 aluminum targets in the flux trap. These changes extended the length of cycle 333 (27.3 days) 3.5 days beyond that of cycle 332 (23.7 days).

End-of-cycle burnup has a mean value of 2303 MWD before cycle 150 (Summer of 1977). If the sets of cycles which included neutron filters are removed from consideration, the mean burnup value is 2141 MWD for cycles after cycle 150. Note that the reactor operating power was 100 megawatts before and after the apparent change. This difference in end-of-cycle burnup corresponds to approximately 1.9 days of operation at 85 MW, 7.6% of a current, nominal operating cycle time of 25 days.

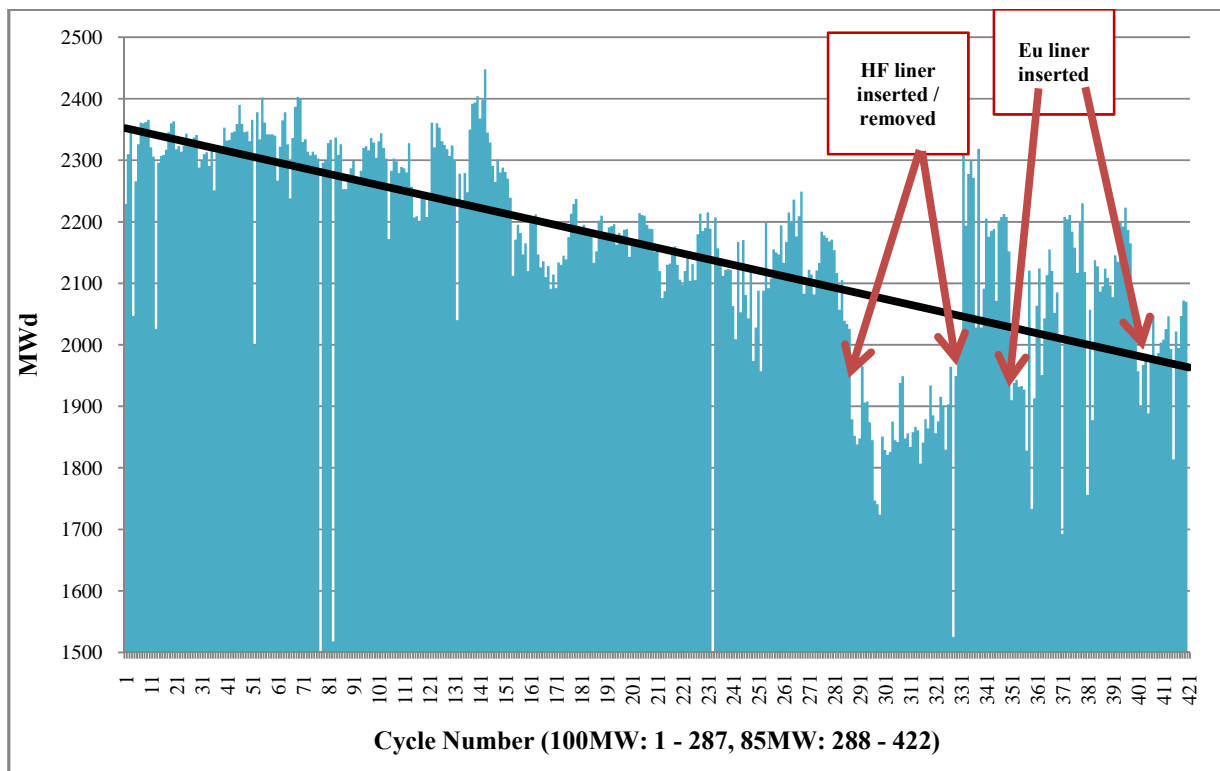


Fig. 2. Presentation of linear regression fit to data and identification of cycles with neutron filters

Given that the HFIR fuel design has not changed over the lifetime of the reactor, factors that could account for a step change in reactivity that was not recovered during subsequent years (thereby reducing cycle lifetime) include replacement of the initial beryllium reflector with subsequent reflectors of poorer quality, a change in coolant water chemistry, or a permanent change in the configuration of the central target region. Control/safety plate design in HFIR is basically unchanged since startup and would not affect cycle length as control and safety plates are fully withdrawn at end-of-cycle.

The HFIR beryllium reflector consists of three annuli. The innermost is the removable beryllium reflector, middle is the semi-permanent reflector, and the outer is the permanent reflector. Typical lifetime values for reflector components are shown in Table 1.

Table 1. Typical lifetimes of HFIR reflector components

Reflector component	Typical lifetimes		
	Exposure (MWD)	Reactor operating time at 85 MW (years)	Calendar time assuming 8 cycles per year (years)
Removable Beryllium	83,700	2.7	4.9
Semi-permanent	167,400	5.4	9.8
Permanent	279,000	9.0	16.5

The irradiation intervals of the components of the beryllium reflector were examined to determine if any significant changes occurred around cycle 150. Fig. 3 shows the reflector replacement intervals plotted with a portion of the data shown in Fig. 1. No changes occurred at or around the time of the apparent reduction in end-of-cycle exposure.

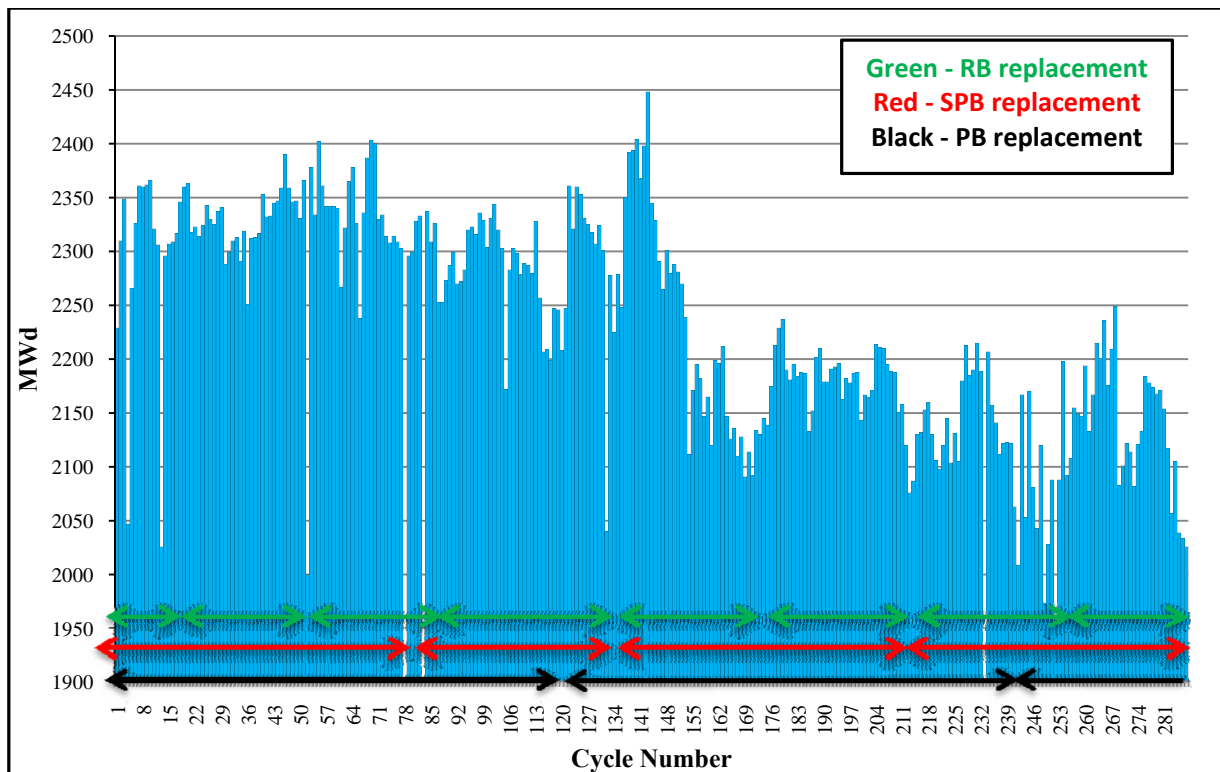


Fig.3. Schedule for change-out of beryllium reflector components

During the time frame spanned by Fig. 3, HFIR coolant water was processed through a demineralizer located on site. There were no changes to the coolant water process during the time span under consideration.

Central target loadings for the cycles shown in Figs. 2 and 3 were investigated with the results shown in Fig. 4. Originally, plutonium targets were loaded into the central region in order to produce californium-252, but after cycle 62 (May 1971), curium targets were loaded into the target region. Recently, fewer transuranic target rods, which contain fissionable nuclides, have been loaded into the central region and more experiments (denoted “other” in Fig. 4) and aluminum rods (available position) have been loaded. There were no significant changes to the target region between cycles 140 and 160. The lack of data for cycles centered around 360 is due to only relative changes in target loadings being documented.

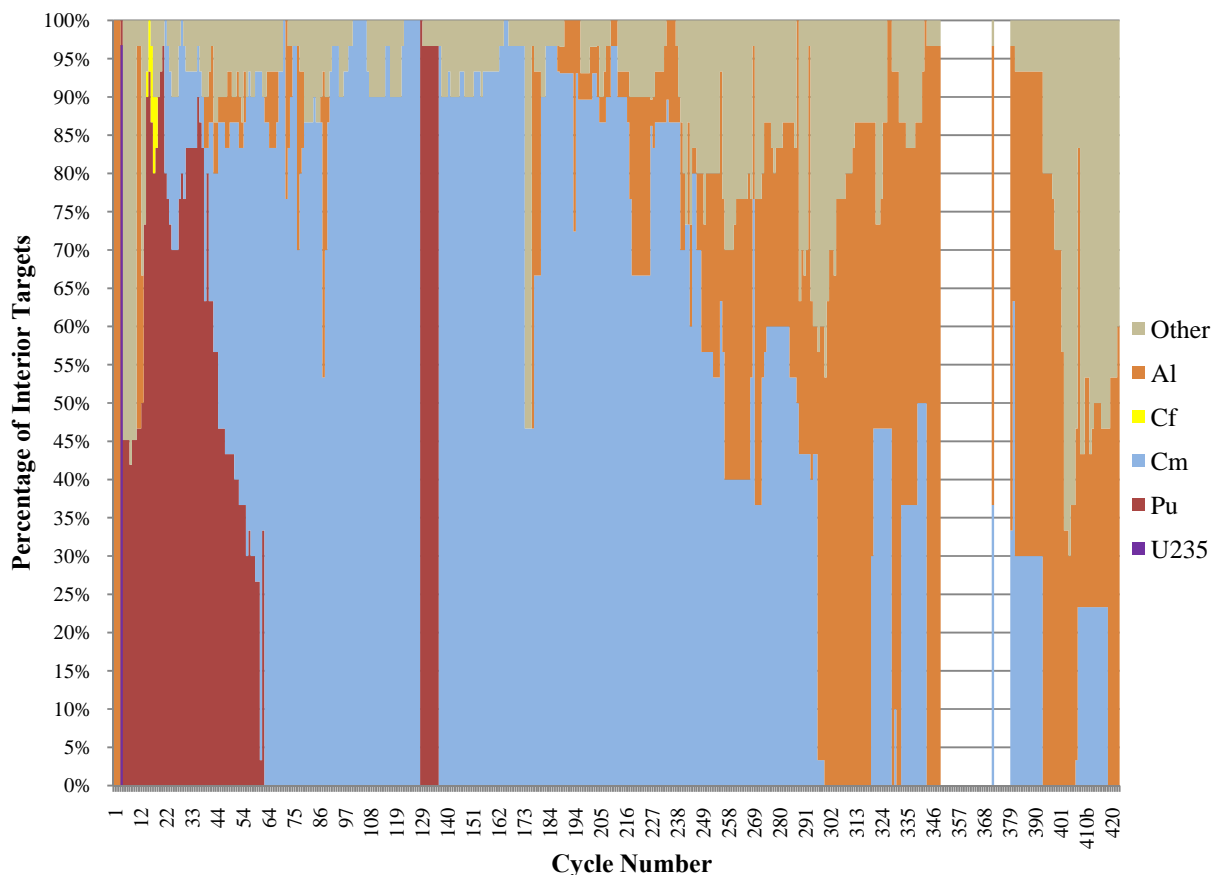


Fig.4. Central target region loadings for HFIR fuel cycles

In conclusion, there appears to be a slight reduction, 8%, in the end-of-cycle exposure for HFIR fuel; the reduction occurring rather precipitously after 15 years of operation. The reason for this apparent change has not been identified but various potential causes have been excluded. In the design of a new, LEU fuel cycle, the National Nuclear Security Agency has stipulated that it will maintain reactor performance during the conversion but that improvement of reactor performance is outside the mandate of the conversion program. The U. S. Office of Science has stipulated that a conversion of the reactor to LEU fuel shall not degrade the performance of the reactor. Debate as to the definition of the end-of-cycle exposure performance of HFIR has been precluded by matching LEU performance to maximum fuel exposure obtained during the past 10 years.

Waste category designation for discharged beryllium reflectors

The Integrated Facility Disposition Project (IFDP) is a program that integrates environmental cleanup with modernization and site revitalization plans and projects. The main objectives of IFDP are to eliminate high risk legacies, complete environmental cleanup, and to enable ongoing modernization of ORNL.

HFIR's permanent beryllium reflectors numbers 2 and 3 are considered legacy waste and await disposition. Currently, reflector number 2 is located in a waste storage area on the Oak Ridge Reservation and reflector number 3 is located in the HFIR fuel storage pool. Fig. 5 depicts the position of the beryllium reflector regions relative to the HFIR core.

Before a path of disposal can be established for the beryllium reflector regions, these components must be classified as to type of waste. Transuranic (TRU) waste is defined as radioactive waste containing more than 100 nanoCuries (3700 Becquerels) of alpha-emitting transuranic nuclides (nuclides with a Z greater than 92) per gram of waste and with half-lives greater than 20 years. According to the Nevada Test Site Waste Acceptance Criteria⁸, ^{237}Np , ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{242}Pu , ^{244}Pu , ^{241}Am , $^{242\text{m}}\text{Am}$, ^{243}Am , ^{243}Cm , ^{245}Cm , ^{246}Cm , ^{247}Cm , ^{248}Cm , ^{250}Cm , ^{247}Bk , ^{249}Cf , and ^{251}Cf are the isotopes that shall be considered when making the transuranic waste determination.

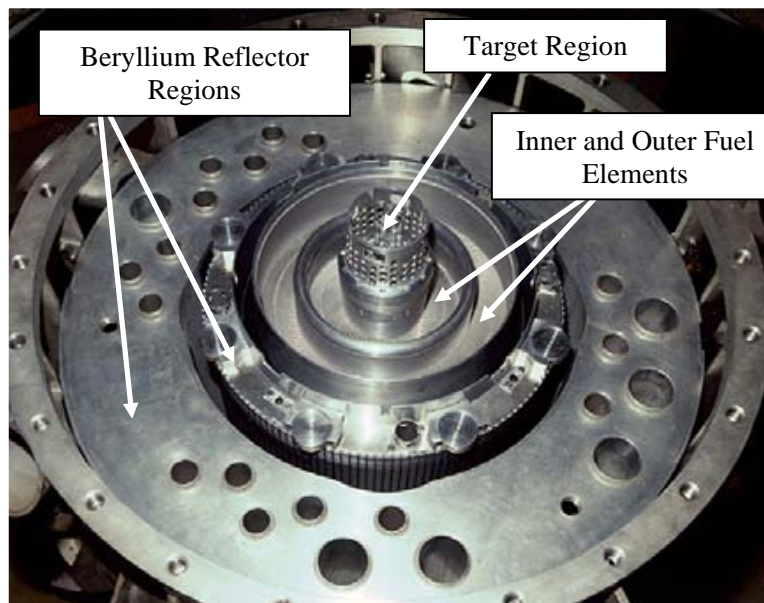


Fig. 5. HFIR core and reflector

Inventories of transuranic nuclides were calculated using the SCALE 6.0 computational system⁹. Inventories were estimated for the irradiation times reported in Table 1 and each of the three reflector regions were considered separately. Irradiation reactor power was 85 MW for the removable beryllium and semi-permanent beryllium but a different simulation was used for the permanent beryllium. The simulation followed the actual history of permanent beryllium reflector 3 and included an initial reactor power of 100 MW, a 3.5 year outage time, and then a reactor power of 85 MW.

The nuclide inventory of trace elements (including uranium) in the beryllium reflector regions was determined via neutron activation analysis (NAA) on three samples of a recent fabrication effort for in-vessel beryllium components. Although these samples were not from the reflector regions being modeled, their composition was judged by L. D. Proctor, ORNL, to be similar. The results of the NAA are listed for all three samples in Table 2. Samples denoted with an “A” and a “B” are duplicates of the same sample.

Table 2. Nuclide inventory in beryllium metal samples

Element	5122A		5122B		5125A		5125B		5127A		5127B	
	µg/g Be	1σ	µg/g Be	1σ	µg/g Be	1σ	µg/g Be	1σ	µg/g Be	1σ	µg/g Be	1σ
sodium	4	1	0	0	0	0	0	0	0	0	2.6	0.8
aluminum	460	104	489	111	442	100	302	68	425	96	487	110
scandium	6.4	0.2	6.1	0.2	6.9	0.2	6.9	0.2	7.6	0.2	7.8	0.3
titanium	86	17	70	15	150	19	132	16	131	19	183	20
vanadium	3.6	0.3	3.9	0.3	3.4	0.2	2.6	0.2	3.8	0.3	4.3	0.3
chromium	54	5	62	5	66	6	72	6	70	6	66	5
manganese	73	2	74	2	82	2	80	2	70	2	75	2
iron	970	79	885	70	981	101	936	100	990	7	909	95
cobalt	4.7	0.5	4.6	0.5	3.3	0.4	2.8	0.4	4.8	0.5	3.6	0.5
copper	83	22	72	20	84	22	71	19	68	19	96	25
tungsten	19	1	19	1	12.5	0.8	12.5	0.8	12.7	0.8	12.9	1
gold	4.1	0.1	3.9	0.1	4.2	0.1	4.1	0.1	4.8	0.1	4.9	0.1
mercury	28	4	30	4	20	3	20	3	20	3	19	3
uranium	16.3	0.9	15.4	0.8	0	0	0	0	0	0	12	1

A lower bound analysis, such that the smallest amount of each of the above nuclides (specifically uranium) is to be inserted into the reflector, is the basis of this calculation. A lower bound analysis was selected for the first step of deciding the disposal path of the beryllium reflector in order to better understand the transmutations inside of the beryllium reflector. If the beryllium reflector is deemed TRU waste via a lower bound calculation and the true U content is greater than the lower bound amount, then the reflector is still TRU waste. The smallest nonzero value listed in Table 3 minus one standard deviation was used for this study. The density of Be was assumed to be 1.845 grams/cm³.

The HFIR beryllium specification calls for the uranium content of the beryllium to be less than or equal to 0.0011 wt. %. However, the uranium content of the permanent beryllium number 3 was determined to be 0.0044 wt. %. The results of the calculations are shown in Fig. 5 where time zero is the start of irradiation of the beryllium in the reactor (RB = removable Be, SPB = semi-permanent Be, PB3 = permanent Be with irradiation cycle for reflector number 3). The initial, non-monotonically increasing portions of the curves are due to the definition of those nuclides included (and therefore those not included) as TRU waste.

If beryllium is procured according to the current HFIR standard, discharged reflector pieces are not transuranic waste for several decades though removable beryllium pieces would become TRU waste approximately 40 years following discharge. Recently available beryllium supplies have a uranium content greater than that stipulated in the standard. Removable and semi-permanent and permanent reflectors fabricated from that material are TRU waste when discharged.

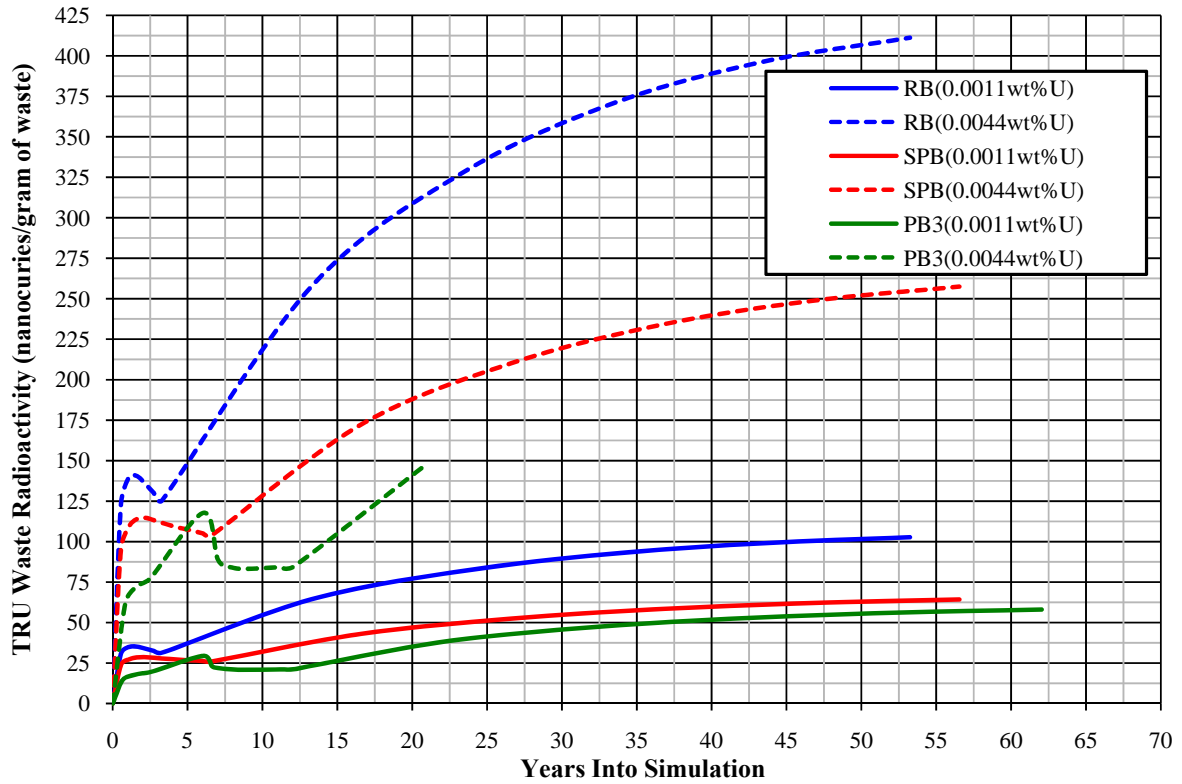


Fig. 5. Transuranic waste activity from discharged HFIR beryllium reflectors

Conclusions

Some degradation in end-of-cycle fuel exposure from HFIR appears to have occurred approximately 25 years ago. The source of degradation is still unknown but does not impact on-going studies of the conversion of the reactor to LEU fuel. If HFIR beryllium reflectors were fabricated according to the HFIR procurement standard for beryllium, most discharged reflector pieces would not be TRU waste but a small quantity would become TRU waste 40 years following discharge. Data which are available for the most recently discharged reflector indicate that all components would be TRU waste upon discharge.

References

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