NUCLEAR TRANSMUTATIONS IN HFIR'S BERYLLIUM REFLECTOR AND THEIR IMPACT ON REACTOR OPERATION AND REFLECTOR DISPOSAL

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NUCLEAR TRANSMUTATIONS IN HFIR'S BERYLLIUM REFLECTOR AND THEIR IMPACT ON REACTOR OPERATION AND REFLECTOR DISPOSAL

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ABSTRACT

The High Flux Isotope Reactor located at the Oak Ridge National Laboratory utilizes a large annular beryllium reflector that is subdivided into three concentric regions and encompasses the compact reactor core. Nuclear transmutations caused by neutron activation occur in the beryllium reflector regions, which leads to unwanted neutron absorbing and radiation emitting isotopes. During the past year, two topics related to the HFIR beryllium reflector were reviewed. The first topic included studying the neutron poison (helium-3 and lithium-6) buildup in the reflector regions and its affect on beginning-of-cycle reactivity. A new methodology was developed to predict the reactivity impact and estimated symmetrical critical control element positions as a function of outage time between cycles due to helium-3 buildup and was shown to be in better agreement with actual symmetrical critical control element position data than the current methodology. The second topic included studying the composition of the beryllium reflector regions at discharge as well as during post-discharge decay to assess the viability of transporting, storing, and ultimately disposing the reflector regions currently stored in the spent fuel pool. The post-irradiation curie inventories were used to determine whether, for disposal purposes, the reflector regions are discharged as transuranic waste or become transuranic waste during the decay period and to determine the nuclear hazard category, which may affect the controls invoked for transportation and temporary storage. Two of the reflector regions were determined to be transuranic waste at discharge and the other region was determined to become transuranic waste in less than 2 years after being discharged due to irradiation of the initial uranium impurity content (0.0044 weight percent uranium). It was also concluded that all three of the reflector regions could be classified as nuclear hazard category 3 (potential for localized consequences only).

Keywords

HFIR, Be, beryllium, He-3, helium, reactivity, waste, TRU, transuranic, disposal, hazard category

1 PROBLEM DEFINITION AND GOAL FOR STUDIES

During the past year, two topics related to the High Flux Isotope Reactor's (HFIR) beryllium reflector were reviewed. The first topic included studying the poison (^{3}He and ^{6}Li) buildup in the reflector regions and its affect on beginning-of-cycle (BOC) reactivity and the startup symmetrical critical control element positions. A reactivity difference of $1.55, which is approximately equal to 0.5 inches in control element position at startup, is the maximum reactivity difference allowed at startup and if exceeded, a technical evaluation is performed to resolve the discrepancy prior to a restart. After a 484 day outage following cycle 407 (Jan. 2006 to May 2007), the reactivity attributed to ^{3}He was erroneously calculated using the current methodology. This occurrence is the motivation for developing a new methodology to predict the reactivity impact ^{3}He has on reactor startup following long outages.

The second topic included studying the composition of the beryllium reflector regions at discharge as well as during post-discharge decay to assess the viability of transporting, storing, and ultimately disposing the reflector regions currently stored in the HFIR spent fuel pool. The post-irradiation curie inventories determine whether the reflector regions are discharged as transuranic waste, become transuranic waste during some period of decay or are not transuranic waste. Categorization is necessary for determination of path to disposal and to establish the nuclear hazard category; the category designation affects the controls invoked for transportation and temporary storage.

1.1 BRIEF DESCRIPTION OF HFIR

HFIR is a compact and versatile research reactor located at the Oak Ridge National Laboratory (ORNL). HFIR went critical in August of 1965 and achieved 100% power (100 MW_{th}) in September of 1966. The reactor was designed and built in response to needs for a very high flux isotope reactor for the purpose of producing transuranic isotopes such as ^{252}Cf. HFIR currently operates at 85 MW_{th} and provides cold and thermal neutron scattering, isotope production, materials irradiation, and neutron activation analysis capabilities. Spent fuel is also used for gamma irradiation after being discharged from the core and placed into a designated location in the spent fuel pool.

HFIR was designed to produce a large thermal flux to power ratio by including an over-moderated flux trap (FT) in the center of the reactor core and a large beryllium reflector on the outside of the core. On the outside of the FT are two concentric fuel annuli, an inner fuel element (IFE) and an outer fuel element (OFE). Two concentric poison bearing control elements (CEs) are situated outside of the fuel elements (FEs) for safety and regulation purposes and a large beryllium reflector encompasses the FEs and CEs. A mockup of HFIR is illustrated in Fig. 1.

This pressurized, beryllium reflected, light water-cooled and –moderated, flux-trap type reactor utilizes highly enriched uranium (HEU) fuel enriched to approximately 93 weight percent in $^{235}U/U$ in the form of U_3O_8 in an aluminum matrix. The IFE and OFE are constructed of 171 and 369 involute fuel plates, respectively, and the fuel is nonuniformly distributed along the arc of the involute and encapsulated within Al-6061 cladding. The total loading of a fresh HFIR core is about 9.4 kg of ^{235}U and a typical fuel cycle length ranges between 22 and 26 days.

Criticality cannot be achieved without the beryllium reflector in place because it enhances the neutron flux in the core by moderating the neutron energies and reflecting neutrons that leak from the core back into the core. The beryllium reflector houses numerous experiment facilities and is subdivided into three regions: a removable reflector (RB), a semi-permanent reflector (SPB), and a permanent reflector (PB). They are named based on the frequency in which they are replaced and they are replaced periodically due to swelling and cracking. The RB has an inner and outer radius of 24.45 cm and 30.25 cm, respectively, and is typically replaced after approximately 83,700 MWd (~40 full power cycles). The SPB has an inner and outer radius of 30.32 cm and 33.02 cm, respectively, and is typically replaced after approximately 167,400 MWd (~80 cycles). The PB has an inner and outer radius of 33.34 cm and 54.61 cm, respectively, and is typically replaced after approximately 279,000 MWd (~135 cycles).

1.2 NUCLEAR TRANSMUTATIONS DUE TO NEUTRON ACTIVATION

The main disadvantage of using beryllium in a neutron environment is the buildup of helium and hydrogen isotopes. ^{3}He is a neutron poison (absorber) that also causes swelling and mechanical property changes, ^{4}He is a gas that also leads to swelling, and tritium (^{3}H) is a radionuclide that has potential to be released. Beryllium reflectors must be replaced periodically due to the internal stresses that are caused by swelling generated by helium and hydrogen gases [1].

When ^{9}Be is irradiated by neutrons above ~1 MeV, it undergoes (n,α) and (n,2n) reactions [1]. ^{9}Be can also undergo (n,γ) reactions when irradiated by thermal neutrons. The reaction that causes poisoning is the (n,α) reaction that leads to the buildup of ^{6}Li and ^{3}He, two isotopes with large thermal absorption cross sections. Thus, a negative reactivity is introduced in the reflector, which has to be accounted in order to accurately predict the startup positions of control elements. The nuclear reactions that govern the transmutations and lead to poisoning are defined in Equations 1 through 7.

$$ {}^{9}_{4}\mathrm{Be} + {}^{1}_{0}n\left(1 - 2\ \mathrm{MeV}\right) \xrightarrow{20\ mb} {}^{6}_{2}\mathrm{He} + {}^{4}_{2}\mathrm{He} \qquad \text{(Equation 1)} $$

$$ {}^{9}_{4}\mathrm{Be} + {}^{1}_{0}n\left(2 - 4\ \mathrm{MeV}\right) \xrightarrow{85\ mb} {}^{6}_{2}\mathrm{He} + {}^{4}_{2}\mathrm{He} \qquad \text{(Equation 2)} $$

$$^{9}_{4}\mathrm{Be} + ^{1}_{0}n\left(4 - 10\ \mathrm{MeV}\right) \xrightarrow{10\ mb} ^{6}_{2}\mathrm{He} + ^{4}_{2}\mathrm{He} \qquad \text{(Equation 3)}$$

$$^{6}_{2}\mathrm{He} \xrightarrow{0.8\ s} ^{6}_{3}\mathrm{Li} + \beta^{-} \qquad \text{(Equation 4)}$$

$$^{6}_{3}\mathrm{Li} + ^{1}_{0}n(\mathrm{thermal}) \xrightarrow{950\ b} ^{4}_{2}\mathrm{He} + ^{3}_{1}\mathrm{H} \qquad \text{(Equation 5)}$$

$$^{3}_{1}\mathrm{H} \xrightarrow{12.33\ yr} ^{3}_{2}\mathrm{He} + \beta^{-} \qquad \text{(Equation 6)}$$

$$^{3}_{2}\mathrm{He} + ^{1}_{0}n(\mathrm{thermal}) \xrightarrow{5327\ b} ^{3}_{1}\mathrm{H} + ^{1}_{1}\mathrm{H} \qquad \text{(Equation 7)}$$

^{6}He is the result of the ^{9}Be(n,α) reaction and decays quickly into ^{6}Li. The relatively large thermal cross section of ^{6}Li, along with the abundance of thermal neutrons in the beryllium reflector lead to a ^{6}Li(n,α) reaction, which produces ^{3}H. During irradiation, ^{3}He is produced by the decay of ^{3}H and is depleted according to the ^{3}He(n,p) nuclear reaction. However, in a no flux environment (i.e. an outage) the ^{3}He concentration increases since there is no destruction mechanism. The transmutation path leading to neutron poisons in the beryllium reflector is shown in Fig. 2.

Other transmutation concerns are those leading to radioactive isotopes. Natural uranium is an impurity present in beryllium and since a high neutron flux exists in the beryllium reflector, the production of transuranic nuclides (Z > 92) and fission products (^{137}Cs, ^{90}Sr, ^{85}Kr, and etc.) is a concern. Natural uranium consists of ^{234}U (0.005 wt. %), ^{235}U (0.711 wt. %), and ^{238}U (99.284 wt. %) [2]. The uranium located in the beryllium reflector can transmute into heavier isotopes via neutron absorption and most of these heavy isotopes decay by emitting alpha and beta radiation. Thus, there is a potential for the HFIR reflectors to become transuranic waste. The primary nuclear reaction chain leading to transuranic isotopes from the uranium impurity is shown in Fig. 3 [3].

In addition, aluminum parts including, but not limited to experiment position liners, flanges, locking plates, and screws are incorporated in and disposed with the beryllium reflector regions. Neutron activation of aluminum as well as the impurities associated with both beryllium and aluminum (magnesium, silicon, iron, titanium, nickel, tin, vanadium, sodium, chlorine, and etc.) produces radioactive isotopes such as ^{55}Fe, ^{60}Co, ^{14}C, and ^{63}Ni.

1.3 HFIR STARTUP PROCEDURE

HFIR fuel cycle 407 ended on January 13, 2006 and cycle 408 began on May 13, 2007. The 484 day outage was due to the conversion of horizontal beam tube number four from a 30-m small angle neutron scattering facility to a high performance hydrogen cold source. The estimated symmetrical critical control element position (ESCCEP) for the startup of cycle 408 was 18.656 inches [4]. During the startup of cycle 408 the difference between the estimated and actual symmetrical positions

exceeded $1.55 requiring a pause in reactor startup. If the reactivity difference between actual and calculated startup equals or exceeds $1.55, approximately equal to 0.5 inches in control element position at startup, the startup procedure is suspended and a technical evaluation is performed to resolve the discrepancy prior to a restart. Following the pause in the approach to critical, the ESCCEP was reexamined and it was determined that the reactivity attributed to shutdown ^{3}He poison was too large using the current methodology.

The actual symmetrical critical control element position (ASCCEP) for the startup of cycle 408 was 18.090 inches, a difference of 0.566 inches from the estimated startup position. The incorrect prediction of the ASCCEP for cycle 408 due to an incorrect prediction in the ^{3}He poisoning effect is the motivation behind developing a better ^{3}He poisoning correlation. ^{3}He buildup effects have been studied for other beryllium reflected reactors such as the MARIA reactor at the Institute of Atomic Energy in Poland [5] and the Belgium Reactor 2 (BR2) located at SCK-CEN in Belgium [6].

The current methodology for calculating the change in symmetrical critical control element position with respect to a reference cycle, typically the cycle prior to the current cycle (ΔR), associated with ^{3}He buildup is shown in Equation 8 where $T_{d,C}$ is the decay time on the reflector for the current cycle (outage time between the shutdown of previous cycle and the startup of the current cycle) and $T_{d,R}$ is the decay time on the reflector for the previous cycle. The value 0.0047 is a multiplicative factor used to convert from decay time to change in control element position and was obtained for the reactor when it operated at 100 MW_{th} and is based on measurements obtained for typical outages during the late 1960s (a couple days to a month). If the current decay time is greater than the reference decay time the ΔR value is positive and the control elements are withdrawn (due to a larger, ^{3}He induced, negative reactivity) and vice versa.

$$\Delta R \text{ (inches)} = 0.0047 \text{ (inches/day)} \left[T_{d,C} \text{ (days)} - T_{d,R}\text{(days)}\right] \qquad \text{(Equation 8)}$$

1.4 BERYLLIUM WASTE CONSIDERATIONS

It is desired to move beryllium reflectors that have been discharged from the reactor out of their interim storage location in the spent fuel pool to make room for anticipated operational and maintenance activities. Once the Be reflectors are declared as waste, they must be characterized to ensure safe interim storage and to facilitate eventual disposal. As part of the characterization process, the isotopic inventory of radiological constituents in the reflectors must be determined. The curie inventory of identified isotopes is compared against the acceptance criteria of potential waste storage and disposal sites to determine whether the waste may be received and stored at the sites. The inventory also defines the nuclear hazard category which is used in defining hazard controls in

associated safety documentation for transportation and storage.

The reflectors are currently designated as “waste with no identified path to disposal” under DOE Order 435.1 [7 and 8], which specifies requirements for managing radioactive waste generated by DOE activities. DOE Order 435.1 requires that efforts be made to define an acceptable disposal option for waste that has been designated as having no identified disposal path. The reflectors were originally classified as having no identified disposal path based on a conservative characterization that indicated the curie inventory of selected isotopes would exceed the waste acceptance criteria for the Nevada Test Site, the expected disposal location. Since that time, studies of the beryllium reflector utilized in the Advanced Test Reactor (ATR) located at the Idaho National Laboratory (INL) have shown that reflectors used in the ATR will exceed the threshold for transuranic (TRU) waste because of trace uranium impurities in the beryllium [3]. Previous analyses have indicated that the HFIR reflectors may also satisfy the definition of TRU waste [9] because of similar levels of trace uranium initially in the beryllium base metal. Studies reported here were intended to confirm or refute the TRU waste characterization.

The current specification for the HFIR beryllium reflectors requires the uranium content to be less than 0.0011 weight percent (11 micrograms of uranium per gram of reflector), but previous HFIR reflectors have been fabricated with a greater uranium content (as much as 0.0044 weight percent). The Nevada Test Site Waste Acceptance Criteria [10] defines TRU waste as radioactive waste containing more than 100 nCi (3700 Bq) of alpha-emitting transuranic nuclides ($Z > 92$) with half-lives greater than 20 years per gram of waste. Therefore, the following nuclides are considered when making the TRU waste determination: ^{237}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{242}Pu, ^{244}Pu, ^{241}Am, ^{242m}Am, ^{243}Am, ^{243}Cm, ^{245}Cm, ^{246}Cm, ^{247}Cm, ^{248}Cm, ^{250}Cm, ^{247}Bk, ^{249}Cf, and ^{251}Cf. Using this criterion, the presence and quantity of applicable transuranic nuclides in the HFIR reflectors was evaluated at discharge and as a function of decay time for making the TRU waste determination.

In order to move HFIR reflectors from their current location to a new storage site, the degree of nuclear hazard associated with the reflectors must be determined in accordance with DOE-STD-1027 [11]. The post-irradiation isotopic inventory and curie levels in the reflector are used to determine the nuclear hazard category. This standard defines three hazard categories based on isotopic thresholds using a sum-of-the-fractions approach. The threshold for hazard category 3 was designed to include facilities and activities that under postulated accident conditions may have no more than localized radiological consequences. Postulated accidents involving a hazard category 2 inventory have the potential for significant on-site radiological consequences and accidents involving a hazard category 1 inventory can result in significant off-site consequences.

2 CALCULATIONAL METHODS

Three "sets" of calculations were performed: two sets as described below and in Sections 2.1 and 2.2, and one set using the CSAS and MCNP codes principally for quality assurance purposes. That is, the latter set of calculations was performed to extend and verify calculations from the initial set of TRITON/KENO calculations.

Version 6.0 of the SCALE package [2] was the primary code utilized for these studies. Three-dimensional KENO V.a and KENO-VI models of the HFIR [12] were used for multi-material depletion calculations. TRITON was used to couple the Monte Carlo transport code KENO to the point depletion code ORIGEN-S.

The KENO input was set up such that the RB was subdivided into 3 radial regions, the SPB was modeled as one radial region, and the PB was subdivided into 7 radial regions in order to utilize a radially dependent flux distribution since the neutron flux decreases with increasing penetration of the reflectors. An isometric view of the KENO model is shown in Fig. 4. The development of the HFIR KENO model and a more in depth description of this analysis are documented in Ref. 12.

For a few cases, in order to extend the TRITON results with larger numbers of histories and different random number sequences, the TRITON results were appended through the use of the SCALE CSAS module. For all SCALE calculations, the 238-group ENDF/B-VII cross section library was used.

As a quality assurance check of the TRITON-related model development, "stand-alone" ORIGEN calculations were performed. A benchmarked three-dimensional MCNP5 [13] model [14] along with continuous energy ENDF/B-VI.8 data was used to calculate the three-group fluxes required in standalone ORIGEN-S calculations. The time-dependent MCNP5 inputs used were generated by ALEPH [15], a “generic” Monte Carlo depletion tool that couples MCNP and ORIGEN 2.2. The KENO models used in these studies were developed based on the benchmarked/validated HFIR cycle 400 MCNP model and were benchmarked against this MCNP model in Refs. 12, 16, and 17 for various parameters such as flux magnitude, flux spectra, k_{eff}, and etc.

2.1 METHOD FOR DETERMINING STARTUP REACTIVITY EFFECTS

A typical HFIR fuel cycle consists of 25 days of irradiation followed by 25 days of outage time between cycles. For this study, 20 typical cycles were modeled with each cycle consisting of a 25 day irradiation period at a constant power of 85 MW_{th} (~8423 MW/MTIHM) and 25 days of down time, t_d. Another 25 day irradiation step was simulated after the 20 cycles and following the end of the 21st irradiation step multiple decay steps (3, 6, 12, 24, 48, 96, 192, 384, 492, 611, and 730 days) were defined. The first 20 cycles were modeled to allow for poison buildup and were chosen such

that equilibrium ^{6}Li would be reached.

The compositions of the reflector regions at beginning-of-cycle (BOC) 21 and following each of the decay steps (i.e. BOC 22 reflector compositions) were extracted and inserted into CSAS5 inputs, which were used for static k_{eff} calculations. The BOC 21 k_{eff} was calculated for the "reference" case and the BOC 22 k_{eff} values (based on the composition following the decay steps) were calculated in order to determine a correlation attributed to shutdown poisons as a function of t_d. Although the effective multiplication factors could have been taken out of the TRITON output, it was desired to increase the number of histories to obtain better statistics. Ten million histories per transport calculation were simulated in the TRITON/KENO run since numerous transport calculations were performed, but 50 million histories were simulated in the CSAS5/KENO runs. The geometry and material compositions, except for the reflector's composition, were the same for both the CSAS5 and TRITON inputs. Test cases utilizing up to 50 million histories per step were run in TRITON and no significant compositional or flux differences in the reflector regions were observed.

The k_{eff} values were extracted from the CSAS5 outputs and were used to calculate the $\Delta k_{eff}/k_{eff}$ (%) values (reactivity effect of ^{3}He as a function of t_d) using Equation 9. These values were then converted into $\Delta\rho$ (\$) values (change in reactivity) by dividing by 0.00762, the delayed neutron fraction (β_{eff}) associated with HFIR [18]. The corresponding ΔR values were obtained by utilizing the "typical" startup position control element worth of \$3.10/in.

$$reactivity = \frac{k_{eff}^{current} - k_{eff}^{reference}}{k_{eff}^{current} \times k_{eff}^{reference}} \qquad \text{(Equation 9)}$$

2.2 METHOD FOR DETERMINING WASTE CLASSIFICATIONS

Activation calculations were performed on the HFIR beryllium reflector regions in order to characterize them so HFIR staff could assess the viability of moving them out of the spent fuel pool. These studies were performed and documented in Refs. 16 and 17. Ref. 16 documents calculations performed to determine whether the permanent beryllium reflector number 3 (PB3) currently exceeds the TRU waste threshold and whether typical RB and SPB reflectors are discharged as or subsequently become TRU waste. Ref. 17 documents calculations performed to determine the total curie inventory in PB3 and representative RB and SPB reflectors at discharge and during decay.

At the time the calculations in Ref. 16 were performed, the goal was to calculate the TRU inventory. The beryllium reflectors were modeled as solid beryllium cylinders containing uranium and impurities based on neutron activation analyses (NAA) on three in-vessel non-reflector beryllium components. The uranium concentration in PB3 was known to be 0.0044 weight percent, which was used along with the NAA results for the other beryllium impurities. The PB3 design file [19] was

located by HFIR staff and the material composition contained therein was used in Ref. 17 to calculate the post-irradiation curie contributing inventory. The uranium concentration listed in Ref. 19 is 0.0044 weight percent, which was the value used in both Refs. 16 and 17.

The TRU inventory for PB3 and representative RB and SPB reflectors was initially calculated utilizing TRITON/KENO V.a. A separate calculation was performed for each of the three reflector regions. The KENO radial flux results in the reflector were compared to benchmarked ALEPH/MCNP [20 and 21] time-dependent results and were shown to be slightly larger and therefore slightly conservative since the reflector regions were depleted by flux (Fig. 5). The KENO fluxes are slightly greater because the fuel in the TRITON model is not depleted and poison is uniformly distributed in the control element channel. With the resources available, it was not feasible to deplete the fuel and simulate control element movement for 138 cycles. Comparisons over single cycles showed that the perturbations in flux magnitude and spectra are small when compared to time-dependent ALEPH/MCNP calculations where both fuel depletion and control element movement are modeled (Fig. 5).

PB3 was inserted into HFIR between cycles 244 and 245 (cycle 245 began on Jan. 1, 1984) and was removed following the end of cycle 382 (Oct. 1, 2000). HFIR experienced an outage for 3.5 years between cycles 287 and 288 while reactor vessel embrittlement issues were analyzed. The outcome of the outage, which occurred between November 14, 1986 and May 18, 1990, was a power decrease from 100 MW_{th} to 85 MW_{th}. Thus, the calculation performed in order to characterize PB3 utilized 4 steps: (1) irradiate at 100 MW for 926.5 days (92,646 MWd), (2) decay for 1280 days, (3) irradiate at 85 MW for 2,204 days (187,341 MWd), and (4) post-discharge decay [9 and 22].

A typical life of an RB is 83.7 GWd, which corresponds to approximately 1,000 days of irradiation at 85 MW_{th} and a typical life of an SPB is 167 GWd, which corresponds to approximately 2,000 days of irradiation at 85 MW_{th}. The RB and SPB reflectors were irradiated for 1,200 and 2,400 days, respectively, at 85 MW_{th} to account for those reflectors that accumulate greater than typical exposures and in order to obtain slightly conservative results. The RB and SPB were modeled with the same material composition as PB3 since they are all manufactured by the same company and should therefore be similar in composition.

For all calculations, the experiment facilities in the reflectors were not explicitly modeled. Thus, it is assumed that the reflector regions are concentric cylinders composed of beryllium and its impurities. This assumption is valid since these calculations are based on a per mass basis, i.e. the results of these analyses are given in radioactivity per mass of waste. Shutdown periods between cycles were not modeled, except for the 3.5 year shutdown between cycles 287 and 288 (applicable for PB3 model only). The difference in continual irradiation versus operating cycles (~25 days of

irradiation, ~25 days of outage, . . .) adds conservatism in that actual decay periods between operating cycles are not modeled. Thus, the curie levels calculated in the reflector by not modeling outages are greater than the curie levels that would be calculated if the outages were modeled.

Control element withdrawal and fuel burnup was not simulated and the reactor power was kept at a constant value of 85 MW_{th} (100 MW_{th} for the first ~3 years of PB3 analysis). The power distribution within the core could affect local TRU isotopic concentrations but the total inventory of TRU products in the reflector will be largely unaffected. Only the core leakage spectra and magnitude are important in these calculations of TRU products, not the details of what is happening inside the fueled region. Therefore, each of the reflector regions was modeled as one axial zone. However, they were modeled as multiple radial regions, such that a different flux is utilized in each separate region.

Calculations were performed in Ref. 17 to determine the total curie inventory in PB3, an RB, and an SPB at discharge and during decay. The design file for PB3 [19], which lists the chemical composition of the reflector (beryllium and impurities) as determined by chemical and neutron activation analyses, was utilized for determining the initial beryllium nuclide inventory. The Al-6061 parts (experiment position liners, locking plates, and etc.) and the 321 stainless steel (SST) O-rings were included in PB3 by distributing their constituents uniformly throughout the beryllium reflector. The 321 SST composition was retrieved from Ref. 9, which lists the AISI composition limits. The chemical composition of the Al-6061 was chosen based on the most conservative mixture determined in Ref. 23, a report in which a beryllium experiment facility composed of six different compositions (6 different calculations) was analyzed. TRITON was again used to couple KENO and ORIGEN-S, but for these studies KENO-VI was utilized. A HFIR KENO-VI model was constructed after performing the TRU studies [16] solely because the geometry package in KENO-VI is more flexible than that in KENO V.a. The same irradiation history as described in previous paragraphs was used to deplete PB3.

Due to 1) the desire by management to compare the current results with previous similar studies [9] utilizing standalone ORIGEN-S calculations, and 2) the computational time required to run these TRITON cases, it was decided to develop standalone ORIGEN-S inputs to deplete PB3 and compare to the TRITON results. The input generated in Ref. 9 was modified to include the more accurate PB3 material composition and irradiation history. Two inputs were constructed: a best estimate input and a conservative input. The cycle 400 MCNP inputs generated by ALEPH [20] were used to calculate the cycle-averaged three-group fluxes required by ORIGEN-S. The average thermal ($E < 0.625$ eV), resonance (0.625 eV $< E <$ 1 MeV), and fast ($E >$ 1 MeV) neutron fluxes were calculated and supplied to ORIGEN-S.

The best estimate input utilized the cycle-averaged fluxes along with the irradiation history described in previous paragraphs. The conservative case utilized a larger thermal and fast flux since the thermal flux is significant for producing non-tritium isotopes and the fast flux is significant for producing tritium. The conservative case did not include the 3.5 year outage and was irradiated for a conservative time period. The flux spectra and depletion history utilized in both PB3 ORIGEN-S inputs are listed in Table I.

The PB3 TRU waste inventory for both TRITON calculations was shown to be in good agreement, but both of the standalone ORIGEN-S calculations underpredicted (in comparison to TRITON) the TRU inventory. However, the ORIGEN-S calculations overpredicted (in comparison to TRITON) the light element curie inventory in PB3.

The last analysis in this study included determining the appropriate nuclear hazard category of PB3 and representative RB and SPB reflectors. Conservative standalone ORIGEN-S calculations were used to determine the nuclide inventory in the RB and SPB reflectors since it was shown that standalone ORIGEN-S conservatively calculates the light element inventory in comparison to TRITON (using the methods described in previous paragraphs), TRU analyses were already performed on representative RB and SPB reflectors, and the fact that TRITON is much more computationally intensive than ORIGEN-S.

The volumes of the RB and SPB reflectors' beryllium and aluminum regions were calculated slightly conservatively and the chemical compositions used were those previously described. Since the flux in the beryllium reflectors is greatest at EOC when the control elements are fully withdrawn, the EOC cycle 400 MCNP input generated by ALEPH [20] was utilized to calculate the EOC three-group fluxes for input into ORIGEN-S. The same irradiation histories applied in the TRU calculations for the RB and SPB reflectors were used in the ORIGEN-S calculations. The flux spectra/magnitude and depletion history utilized in the RB and SPB reflector's inputs are listed in Table II.

3 RESULTS AND DISCUSSIONS

3.1 STARTUP REACTIVITY EFFECTS

The beginning composition of the reflector regions was 100% ^{9}Be and equilibrium ^{6}Li [0.90 g (1.29 μg/g Be): 0.67 g (4.47 μg/g Be) in RB and SPB, 0.23 g (0.42 μg/g Be) in PB] was achieved after approximately 5 cycles (Fig. 6). The results shown in Fig. 6 reveal that the ^{3}He inventory never reaches equilibrium, it decreases during irradiation and increases during outages. Approximately 1.79 milligrams of ^{3}He (2.55 ng/g Be) is produced in a typical cycle [1.40 mg (9.34 ng/g Be) in RB and

SPB, 0.39 mg (0.71 ng/g Be) in PB]. Tritium and ^{4}He were shown to increase in concentration at a rate of approximately 0.38 grams (0.55 μg/g Be) per typical cycle [0.30 g (2.03 μg/g Be) in RB and SPB, 0.08 g (0.14 μg/g Be) in PB] and 3.78 grams (5.39 μg/g Be) per typical cycle [3.00 g (20.03 μg/g Be) in RB and SPB, and 0.77 g (1.41 μg/g Be) in PB], respectively.

Effective multiplication factors were calculated for the reference case (BOC 21 k_{eff}) and the cases corresponding to the multiple decay steps (BOC 22 k_{eff}). Since the ^{3}He concentration increases as a function of decay time between cycles, t_d, the k_{eff} values decreased as a function of t_d. All of the k_{eff} values were inserted into a spreadsheet and the $\Delta k_{eff}/k_{eff}$ (%) values (reactivity effect of ^{3}He as a function of t_d) were calculated and converted into $\Delta\rho$ ($) (change in reactivity) by dividing by the delayed neutron fraction (β_{eff} = 0.00762). The $\Delta\rho$'s were then converted into change in control element positions, ΔR's, using the conversion factor $3.10/inch thereby determining the change in symmetrical critical control element position associated with ^{3}He worth with respect to the reference case. The actual differential rod worth (DRW) is dependent upon the control element position, but a value of $3.10/inc corresponds to "typical" startup positions (17.5 to 18 inches withdrawn).

The purpose of this study is to determine the "added" reactivity poisoning effect of ^{3}He that builds in following shutdown. Therefore, the $\Delta\rho$ curve was shifted such that the $\Delta\rho$ value at $t_d = 0$ is zero. The curve based on a zero $\Delta\rho$ value at zero shutdown time is compared to the curve used in the current ESCCEP calculation in Fig. 7.

The proposed methodology for calculating the ΔR associated with ^{3}He buildup is shown in Equation 10 where $T_{d,C}$ is the decay time on the reflector for the current cycle in days (outage time between the shutdown of previous cycle and the startup of the current cycle) and $T_{d,R}$ is the decay time on the reflector for the previous cycle in days. A second order polynomial based on the curve shown in Fig. 7 was used to convert from decay time to ΔR rather than a linear (first order) conversion, which is used in the current methodology. If the current decay time is greater than the reference decay time the ΔR value is positive and the control elements are withdrawn (due to a larger negative reactivity) and vice versa.

$$\Delta R \text{ (inches)} = 7.569\text{x}10^{-3} (T_{d,C} - T_{d,R}) - 3.477\text{x}10^{-6} [(T_{d,C})^2 - (T_{d,R})^2] \qquad \text{(Equation 10)}$$

ESCCEP calculations for HFIR cycles 383 – 421 [12] were utilized to compare the current and proposed methodologies (Eq. 10). For this study, it was assumed that uncertainty only exists in the ^{3}He worth (i.e. reactivity worth associated with fuel, cylinder/plate burnup, cylinder/plate ^{182}Ta worths, and etc. remained unchanged from the original calculations). The ESCCEP calculated with both the current and proposed methodologies were compared to the ASCCEPs and the differences are

shown in Fig. 8.

On average the proposed methodology better estimates the symmetrical critical control element positions. The average absolute difference between the actual and estimated positions for the current and proposed methodologies for the 39 cycles is 0.174 and 0.154 inches, respectively. The proposed methodology better predicts the ^{3}He effect for longer outages, but little difference is observed for shorter outages. The lines in Fig. 8 that extend at a delta of 0.5 inches were included because a difference of ~0.5 inches will cause the reactor to be shutdown and a technical evaluation to be performed to resolve the discrepancy prior to startup. The difference of -0.56 inches calculated for cycle 408 using the current methodology was the motivation of this study. A more involved calculation was used for cycle 383 (t_d = 430 days), which produced an ASCCEP – ESCCEP value of -0.180 inch. However, if the current methodology would have been utilized, a difference of -0.858 inches would have been obtained. Thus, the current methodology overpredicts the effects attributed to shutdown ^{3}He and the proposed methodology mitigates the overprediction.

3.2 WASTE CLASSIFICATION

Permanent beryllium reflector number 3 along with representative RB and SPB reflectors were depleted to determine whether they are discharged as- or following decay would become TRU waste. If the beryllium reflectors are produced according to the current HFIR specification, discharged reflectors would not be TRU waste, but the RB (reflector closest to the core) would become TRU waste approximately 40 years after discharge. The current HFIR specification calls for the uranium content in the beryllium reflectors to be less than or equal to 11 micrograms per gram of reflector (0.0011 wt. %), but most previous reflectors have contained a greater quantity of uranium than the current specification outlines. The uranium content in PB3 was determined to be 44 micrograms per gram of reflector (0.0044 wt. %). Through calculations, it was concluded that with an initial uranium concentration of 0.0044 weight percent, PB3 was discharged below the TRU waste threshold, but became TRU waste within three years after being discharged. The RB and SPB reflectors were shown to be discharged as TRU waste if their initial uranium content was greater than or equal to 0.0044 weight percent. The TRU waste inventories in nCi per gram of waste as calculated with TRITON for the three reflector regions are shown in Fig. 9.

The PB3 TRU waste inventories as calculated in TRITON and standalone ORIGEN-S are illustrated in Fig. 10. As observed in Fig. 10, the two ORIGEN-S calculations are in similar agreement but underestimate the total TRU inventory in comparison to the TRITON calculations. The two TRITON calculations are in good agreement, but the results using a more accurate material composition and the KENO-VI geometry predict a slightly greater TRU waste curie inventory. Input

differences that caused these small inventory discrepancies are discussed in Section 2.2. All four calculations support the conclusion that PB3 currently exceeds the TRU waste limit. Note that PB3 was discharged in October of 2000.

The current (June 2010) radionuclide curie levels of the major curie contributing nuclides in PB3 and as calculated in TRITON and ORIGEN-S are shown in Fig. 11. The bar plot shows that the ORIGEN-S calculations estimated larger inventories of light elements in comparison to TRITON and TRITON calculated larger inventories of fission products in comparison to ORIGEN-S. The total PB3 radioactivity as calculated in TRITON, a best estimate ORIGEN-S calculation, and a conservative ORIGEN-S calculation as of June 2010 are 68 kCi, 101 kCi, and 161 kCi, respectively. Tritium, a beta emitter, contributes approximately 99% of the total radioactivity (Ci) while ^{60}Co and ^{137}Cs (sources leading to high energy gamma rays) contribute approximately 0.35% and 0.01% - 0.04%, respectively.

The primary radionuclides in PB3 following discharge are shown in Table III. The table lists nuclides present in quantities greater than 1 Ci at 9.67 years after discharge (i.e. June 2010) sorted in descending order. The values for the 15 nuclides in Table III are the largest values calculated in either TRITON or standalone ORIGEN-S. The radioactivity listed for ^{137}Cs, ^{137m}Ba, ^{90}Y, ^{90}Sr, and ^{241}Pu were produced by the TRITON calculation and the rest were calculated by ORIGEN-S. After comparing all of these values to the hazard category thresholds [11] using the sum-of-the-fractions approach, it is concluded that PB3 can be classified as nuclear hazard category 3.

The total radioactivity (Ci) of the RB and SPB beryllium and aluminum regions is shown in Fig. 12. The curie level of the beryllium regions dominates that of the aluminum regions due to the high levels of tritium, which contributes more than 99% of the total RB and SPB curie inventories after a short decay time (after the short-lived radioisotopes are removed). The primary radionuclides in the RB and SPB following discharge are listed in Tables IV and V, respectively. Both of these tables are sorted by the largest curie contributor to the smallest at a time 10 years after discharge for nuclides at levels greater than 1 curie. The primary curie contributing radioisotopes present in the aluminum regions (^{55}Fe, ^{60}Co, and ^{63}Ni) are plotted as a function of time after discharge in Fig. 13. After comparing the curie inventory to the hazard category thresholds [11] using the sum-of-the-fractions approach, it is concluded that the representative RB and SPB can be classified as nuclear hazard category 3.

4 CONCLUSIONS

The TRITON sequence within the SCALE code package was used to develop a new methodology for predicting the ^{3}He worth in the High Flux Isotope Reactor's beryllium reflector as a function of

outage time between cycles. ^{3}He is produced and depleted during operation (neutron flux environment) and is produced during an outage (no neutron flux environment) due to the radioactive decay of tritium. A newly-derived correlation was developed and shown to improve the accuracy of predicting the symmetrical critical control element positions following long periods of down time between cycles. The newly-derived methodology is currently being incorporated in the HFIR Estimated Symmetrical Critical Control Element Position procedure. The explanation for the observed inadequacy of the current procedure for predicting the poisoning effect following long shutdowns (several months) is due to the buildup of ^{3}He in the reflector as a function of decay time not corresponding to a linear decrease in reactivity. The current procedure for estimating poisoning effects had assumed a linear relationship and an operating power of 100 MW_{th}. While not the focus of this study, the results derived serve to validate the SCALE system and associated cross section libraries for predicting the in-growth of nuclides during irradiation of beryllium.

The results leading to the new reflector reactivity prediction methodology also provided insight into the production rate of other significant nuclides in the beryllium reflector regions. ^{6}Li was shown to reach an equilibrium concentration of approximately 0.90 grams (1.29 μg/g Be) [0.67 g (4.47 μg/g Be) in RB and SPB, 0.23 g (0.42 μg/g Be) in PB] after four to five full power cycles. ^{3}He was shown to never achieve equilibrium and is produced at a rate of approximately 1.79 milligrams (2.55 ng/g Be) per typical cycle [1.40 mg (9.34 ng/g Be) in RB and SPB, 0.39 mg (0.71 ng/g Be) in PB]. Tritium, a radiolytic gas, is produced at a rate of approximately 0.38 grams (0.55 μg/g Be) per typical cycle [0.30 g (2.03 μg/g Be) in RB and SPB, 0.08 g (0.14 μg/g Be) in PB]. Tritium is the major curie contributor in spent reflectors and although it is tightly bound in the beryllium lattice it can be released if the beryllium begins to corrode [3]. ^{4}He is produced at a rate of approximately 3.78 grams (5.39 μg/g Be) per typical cycle [3.00 g (20.03 μg/g Be) in RB and SPB, and 0.77 g (1.41 μg/g Be) in PB] and is a gas that causes the beryllium reflector regions to swell and crack and therefore dictates the life span of the reflector regions.

The TRITON sequence was also utilized to characterize, for waste disposal operations, HFIR's beryllium reflector regions. If the reflector regions are manufactured to the current HFIR specification of less than or equal to 11 micrograms of uranium per gram of reflector then the reflectors would be discharged below the transuranic waste threshold with the possibility of the RB exceeding the threshold after approximately 40 years of decay. If the initial uranium concentration is greater than or equal to 44 micrograms per gram of reflector then PB3 became transuranic waste approximately two to three years after being discharged and representative RB and SPB reflectors are discharged as transuranic waste. The maximum initial uranium concentration that could be

accommodated without exceeding the transuranic waste threshold was not determined, but it appears to be close to the 11 microgram per gram limit.

Therefore, the amount of uranium impurity in the fresh beryllium reflectors is a concern because the initial amount of uranium dictates whether or not the reflectors become transuranic waste. Classification of the reflectors as transuranic waste also categorizes them as waste with no path to disposal since the isotopic content exceeds the acceptance criteria for the Nevada Test Site [10] and non-defense transuranic waste is not currently accepted at the Waste Isolation Pilot Plant in New Mexico [24]. In addition to the transuranic waste determination, other factors such as the ^{14}C concentrations being greater than the Class C upper activity limit of 80 Ci/m^3 in activated metal [25] contribute to the designation of no path to disposal.

The total current (June 2010) curie inventory of PB3 as calculated in TRITON is approximately 67.6 kCi and 21 different radionuclides with curie levels greater than 0.1 Ci contribute to this radioactivity. The predominant radionuclide is tritium at 66.8 kCi (~6.91 grams). The predominant gamma emitter is ^{60}Co at 221 Ci (~0.20 grams). Other contributing nuclides include ^{55}Fe, ^{14}C, and ^{63}Ni at 386, 89, and 42 Ci, respectively. After 10 years of decay, the total radioactivity of a representative RB and SPB reflector as calculated in standalone ORIGEN-S is 861 kCi and 623 kCi, respectively, and over 99% of the radioactivity is contributed by tritium. The major gamma emitter, ^{60}Co, is present at curie levels of 117 and 125 Ci in the RB and SPB, respectively, after 10 years of decay.

According to the radionuclide threshold limits defined in DOE-STD-1027 [11], PB3 is currently nuclear hazard category 3 and representative RB and SPB reflectors were also shown to be hazard category 3. The nuclear hazard category and the specific isotopic inventories can be used to determine whether the reflectors fit within the authorized limits specified in the safety basis for the proposed temporary storage location. If that is found to be the case, the hazard category and isotopic inventories will be used to develop a transportation safety document for moving the reflectors from the spent fuel pool to the storage location.

The standalone ORIGEN-S calculations were shown to overpredict the tritium inventory and underpredict the transuranic waste inventory in comparison to the TRITON calculations. It is therefore shown that more details need to be inserted into standalone ORIGEN-S calculations in order to properly predict the total nuclide inventory in the beryllium reflector regions. The best estimate ORIGEN-S calculations utilized the cycle-averaged thermal, resonance, and fast neutron fluxes in the permanent beryllium reflector as calculated with benchmarked MCNP calculations. Thus, it is shown here that modeling the permanent reflector as a point is not sufficient for this application and therefore must be depleted via a radially varying neutron flux.

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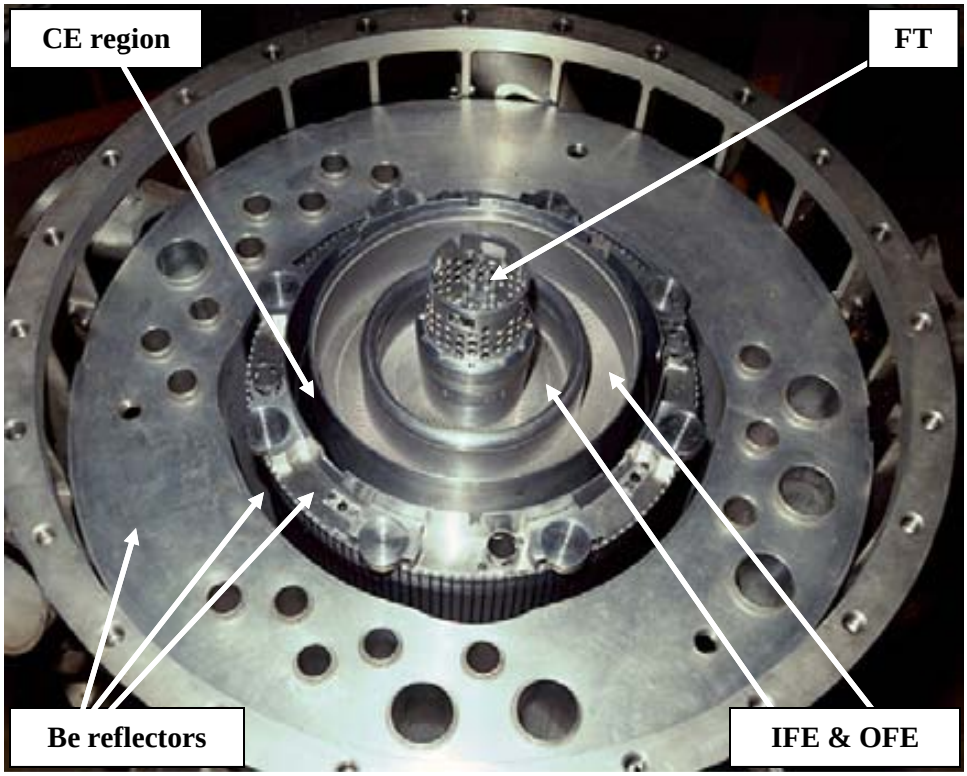

Fig. 1. Mockup of the High Flux Isotope Reactor.

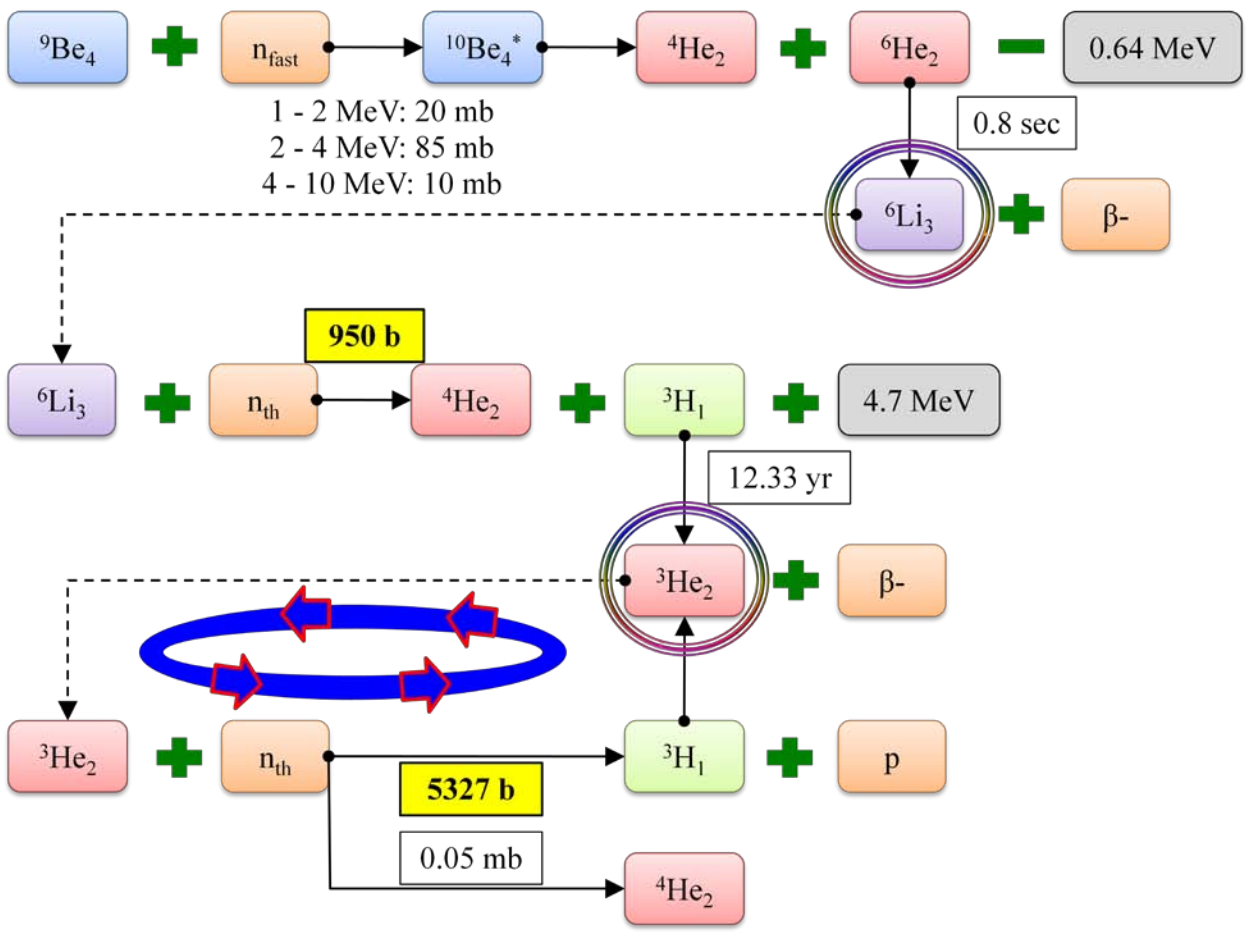

Fig. 2. Transmutation path leading to poison and tritium buildup in the beryllium reflector.

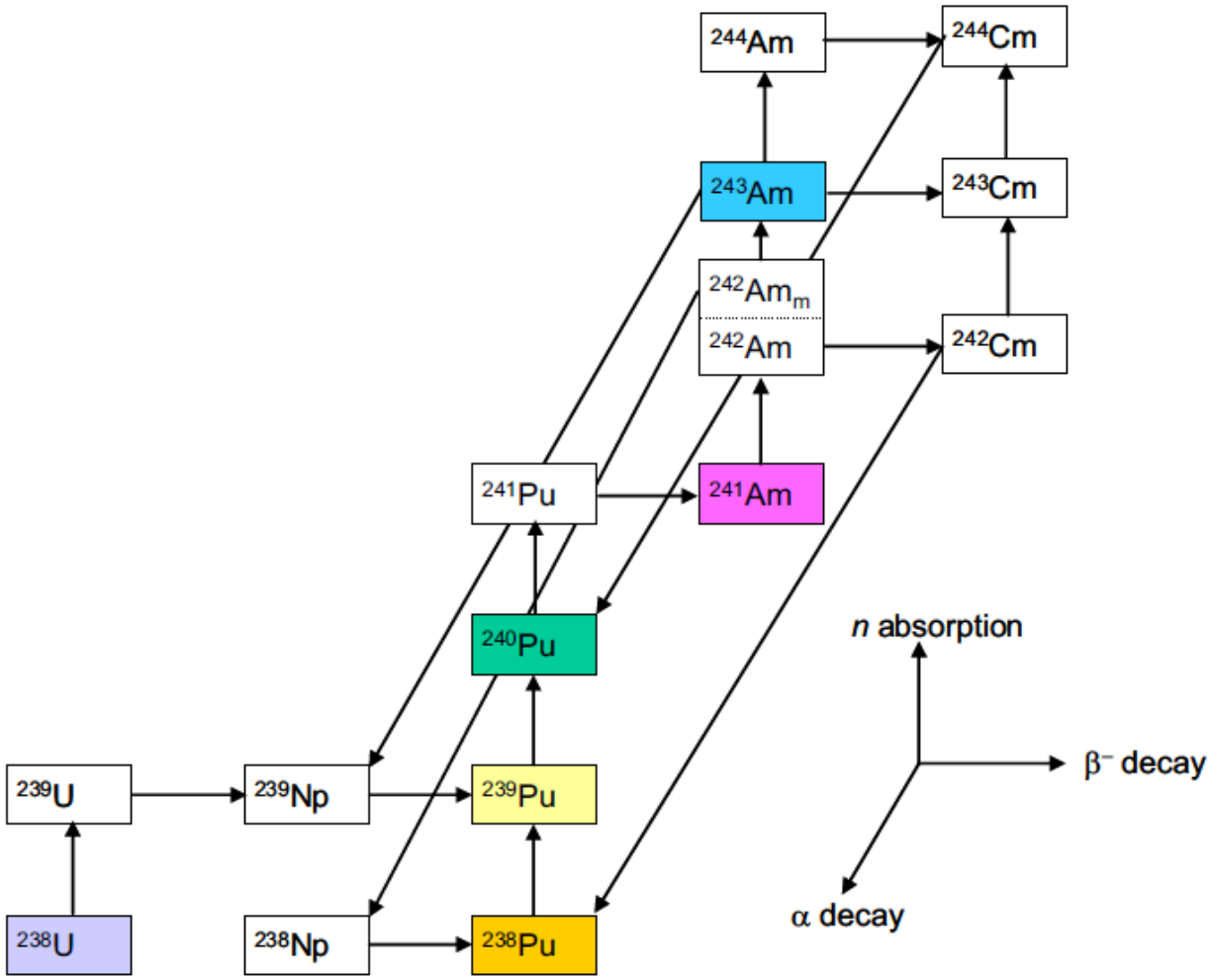

Fig. 3. Transmutation path for transuranic isotope production [Ref. 3].

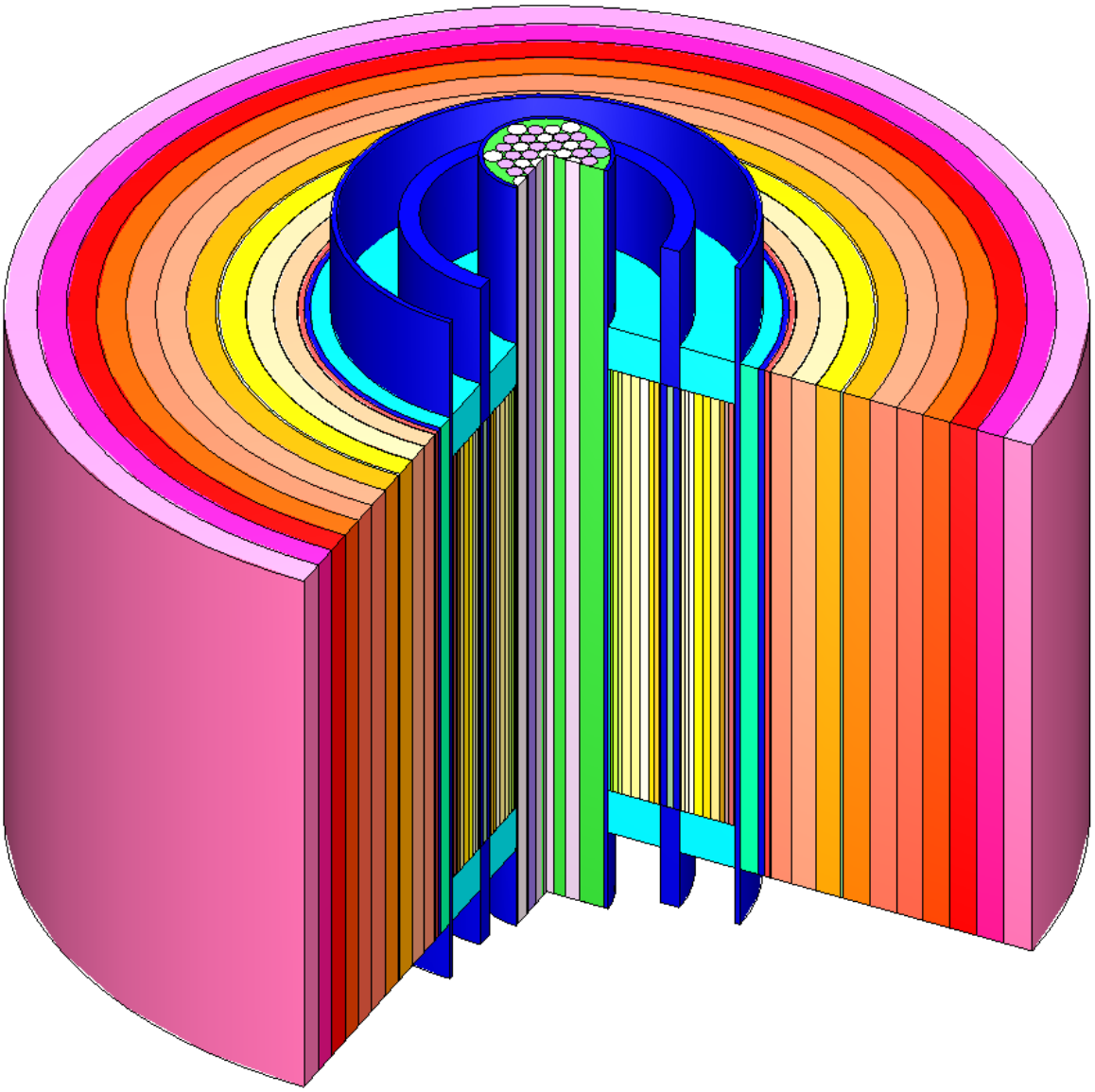

Fig. 4. Isometric view of the HFIR KENO model.

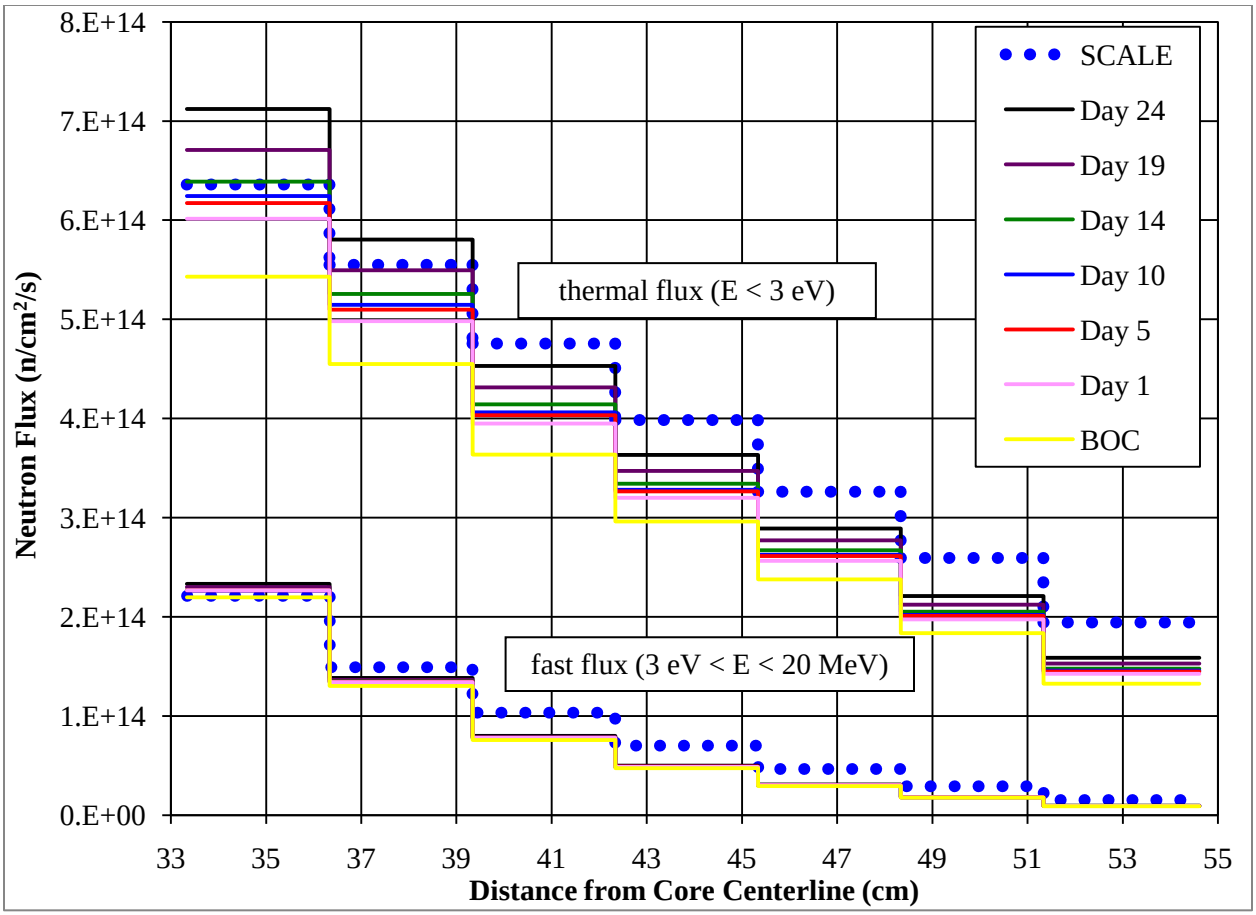

Fig. 5. SCALE and time-dependent ALEPH/MCNP flux comparisons.

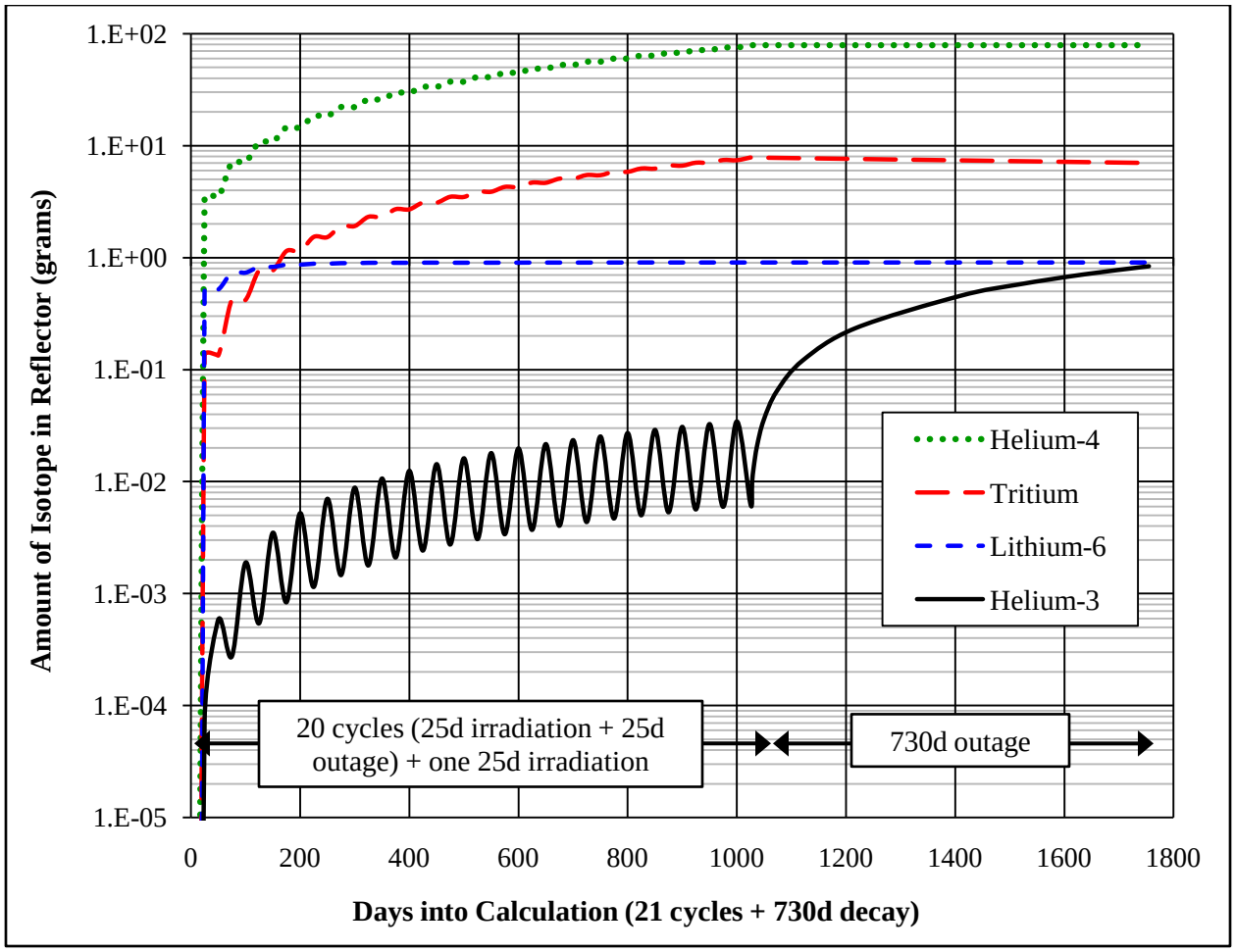

Fig. 6. Poison and gas buildup in the beryllium reflector resulting from neutron activation.

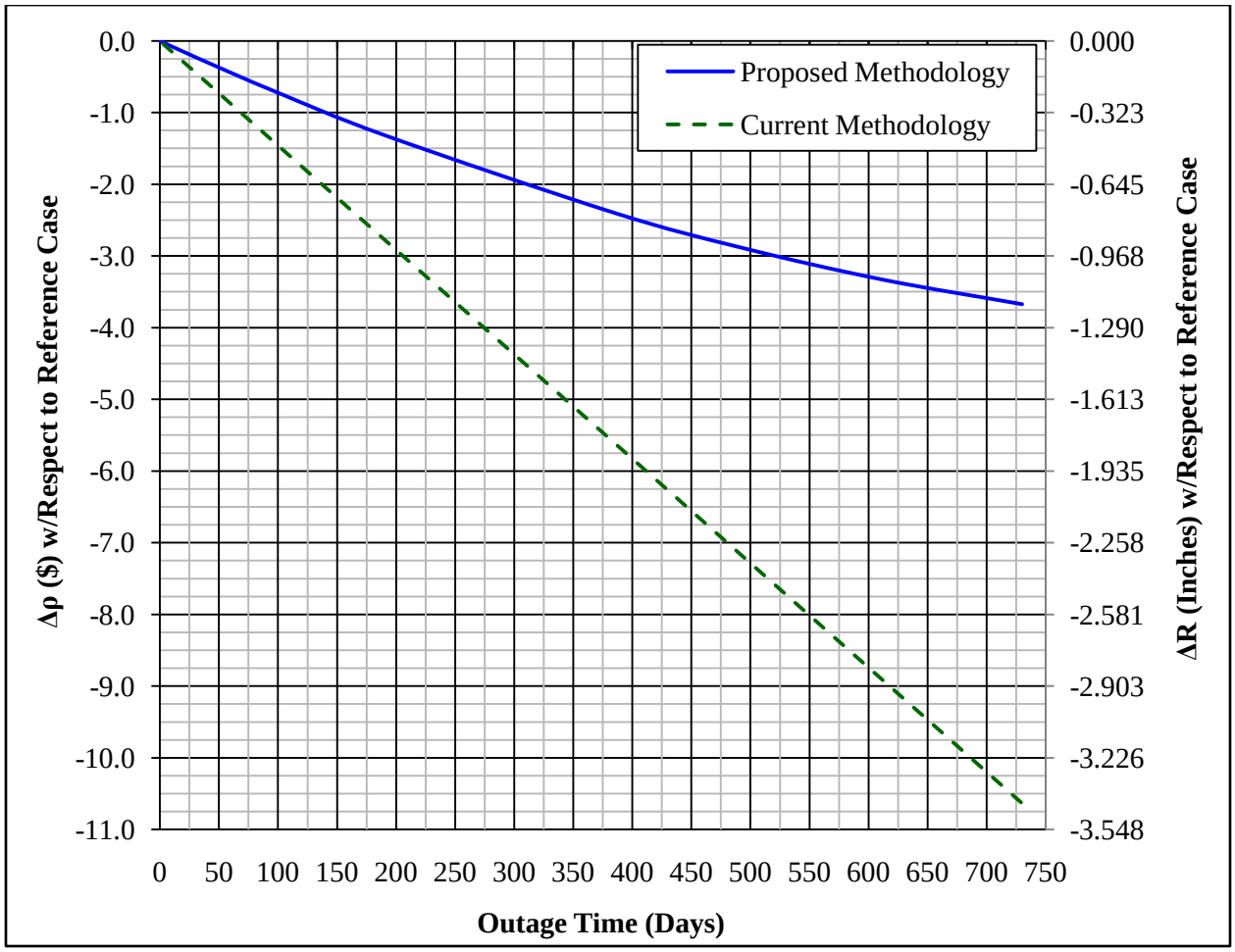

Fig. 7. Comparison of current procedure and newly derived ^{3}He poison effect estimates.

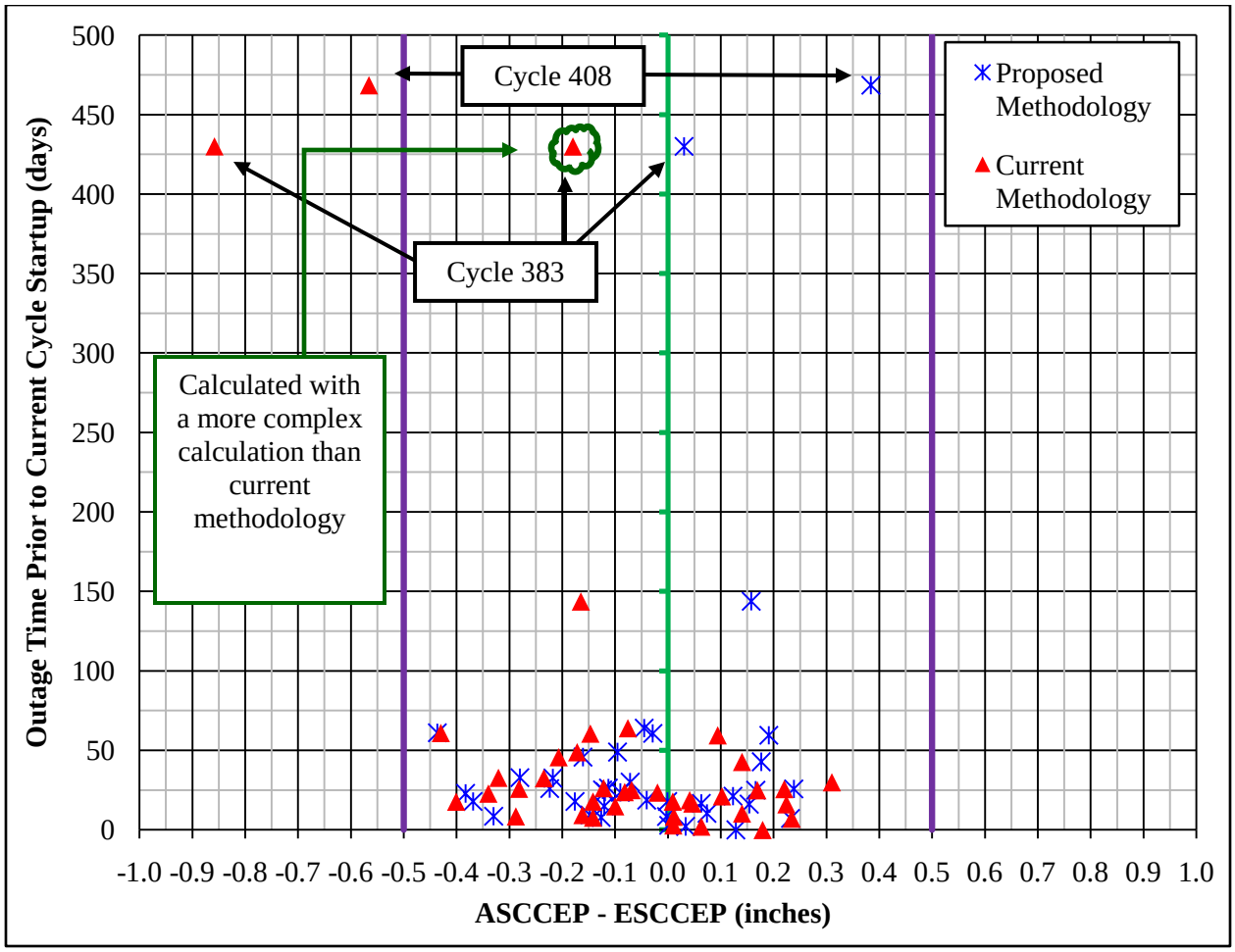

Fig. 8. Comparison of current and proposed ESCCEP methodologies against ASCCEP data.

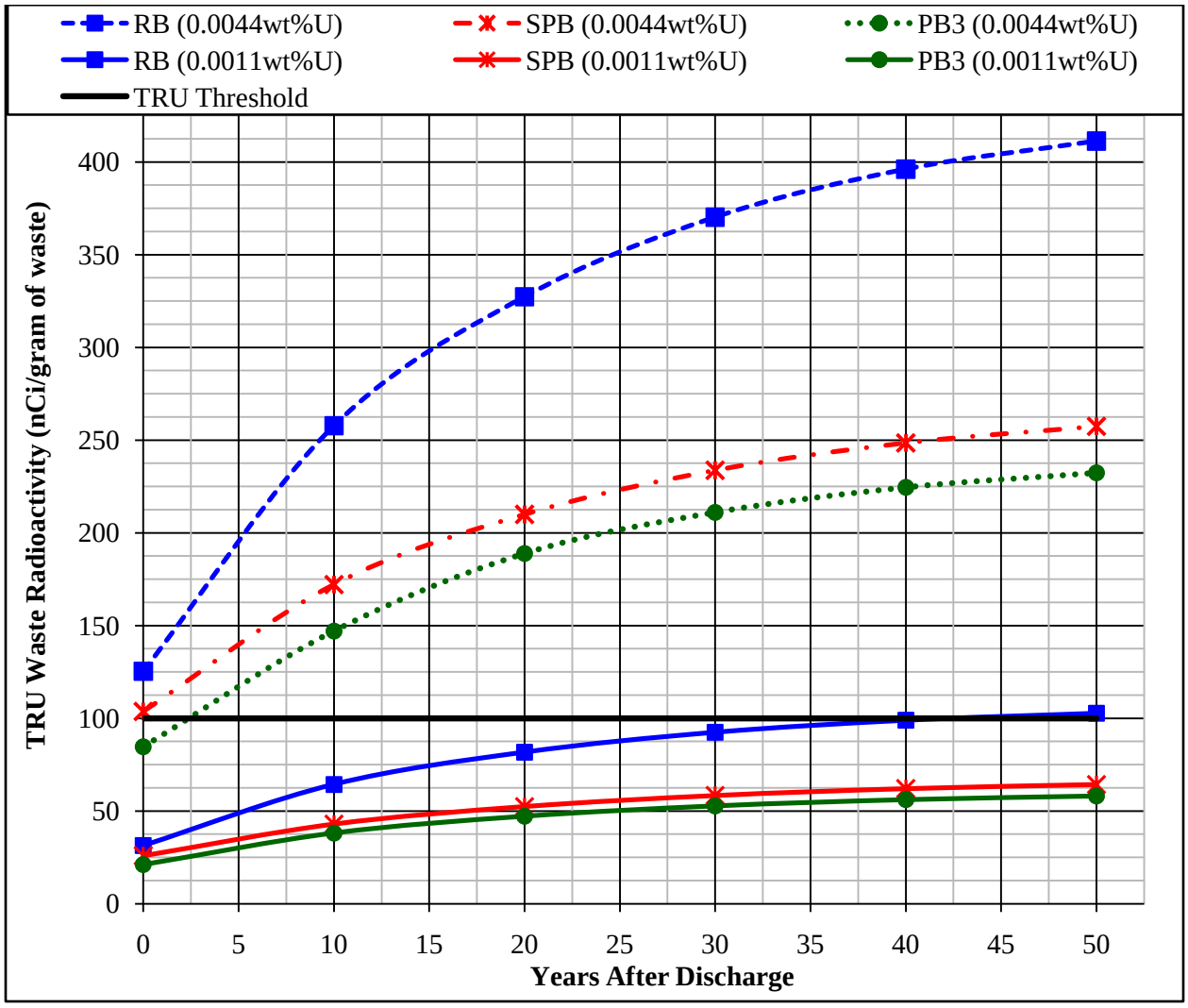

Fig. 9. TRU waste inventory in beryllium reflector regions following discharge.

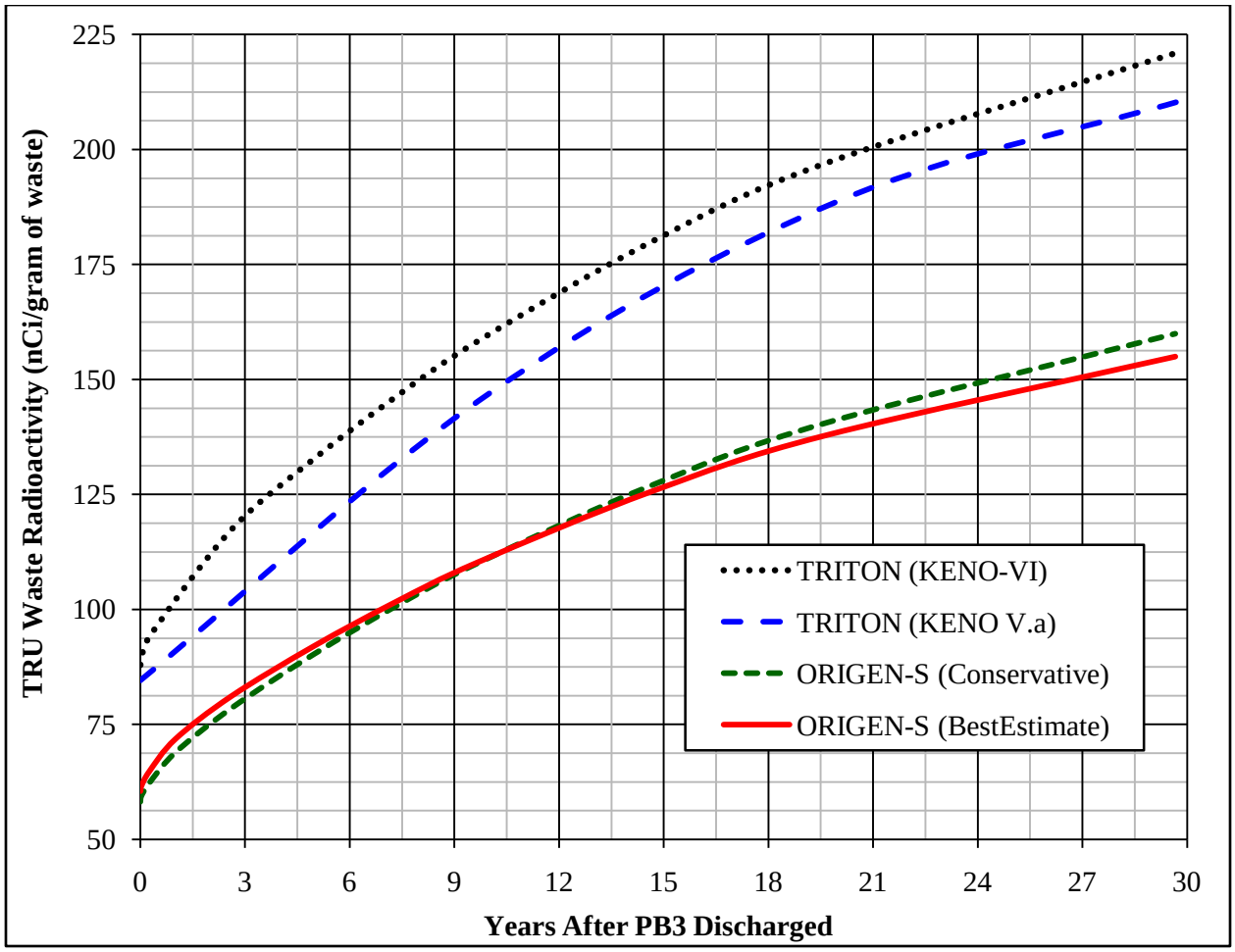

Fig. 10. TRU waste inventory in PB3 following discharge.

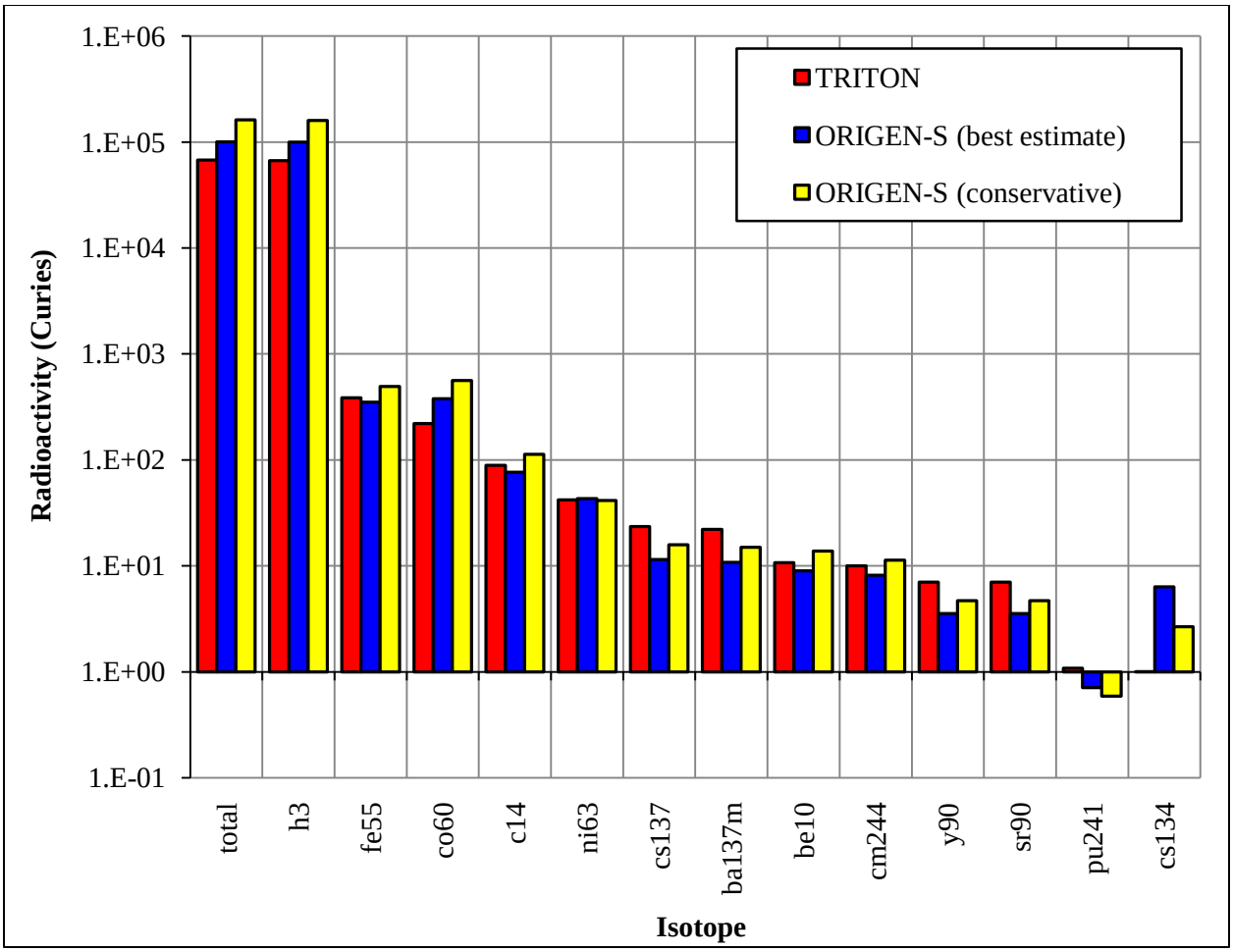

Fig. 11. Current (June 2010) radionuclide curie levels in PB3 as calculated in TRITON (left), ORIGEN-S best estimate (middle), and ORIGEN-S conservative (right).

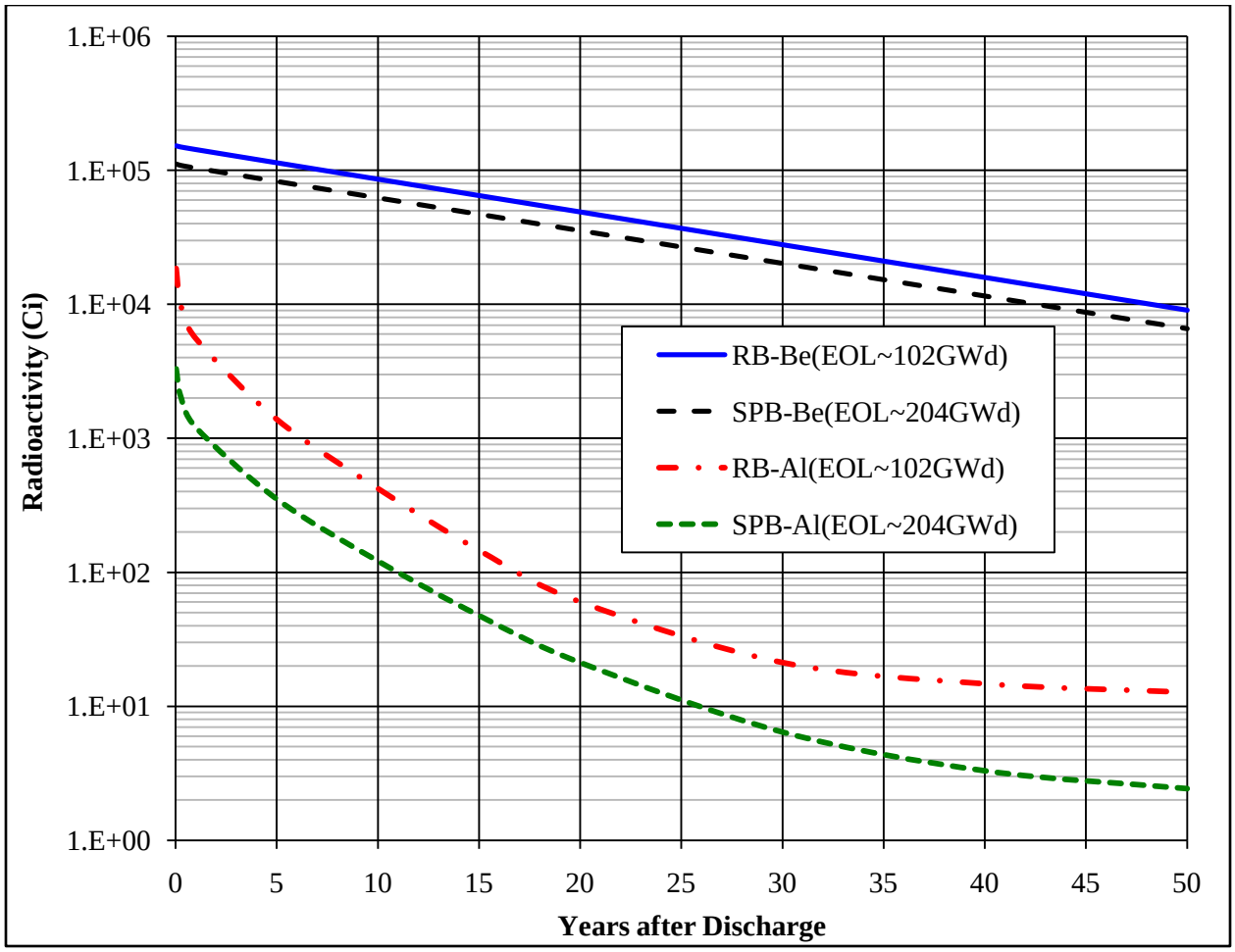

Fig. 12. Total radioactivity in RB and SPB beryllium and aluminum regions.

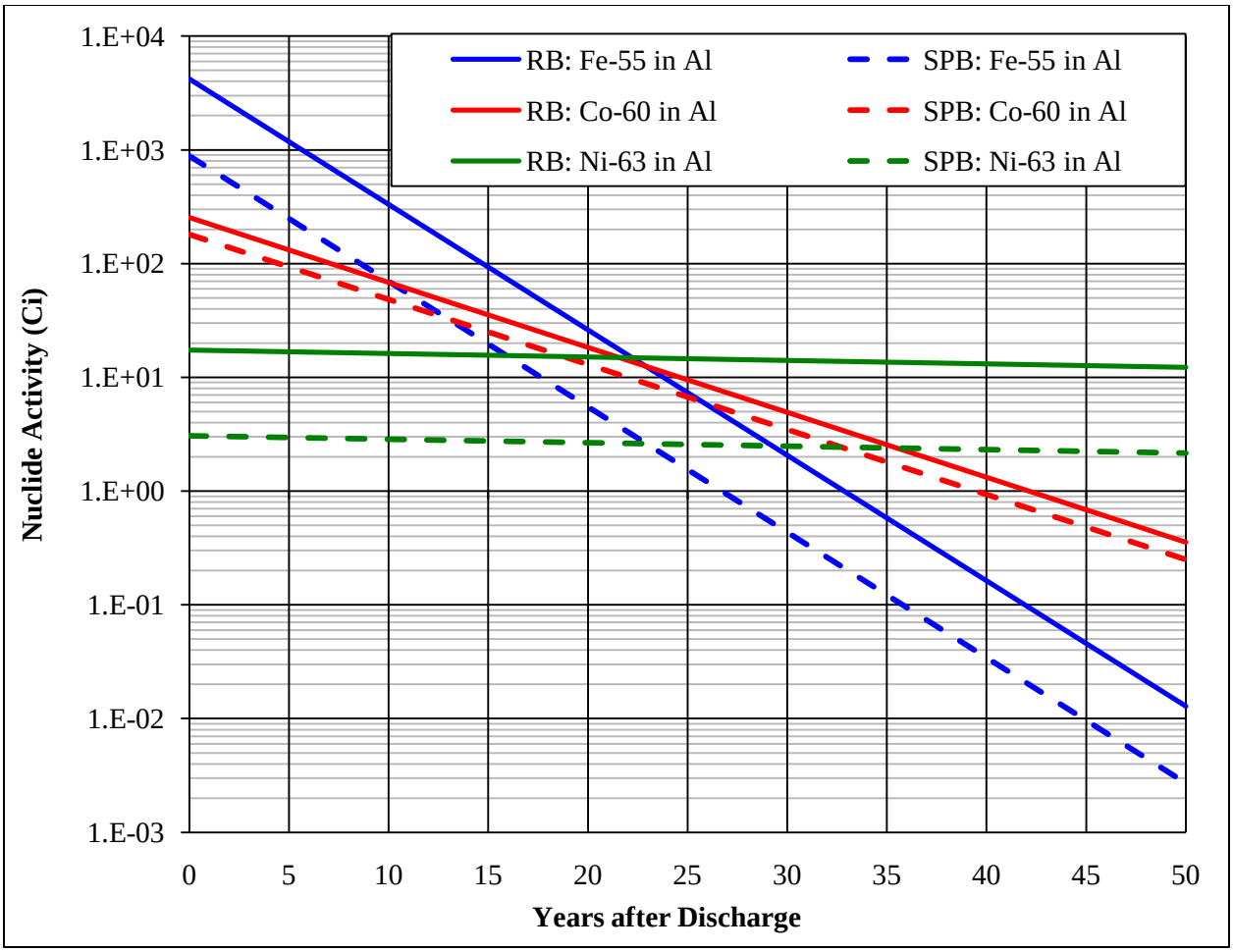

Fig. 13. Major RB and SPB aluminum region radionuclides.

Table I. Irradiation history input for PB3 ORIGEN-S calculations.

ORIGEN-S case	Step	Days	$\phi_{thermal}$ (n/cm^2/s)	$\phi_{resonance}/\phi_{thermal}$	$\phi_{fast}/\phi_{thermal}$
conservative	1	3294	5.00E+14	0.03	0.022
	2	10835	0	0	0
best estimate	1	914	3.82E+14	0.23	0.021
	2	1280	0	0	0
	3	2220	3.25E+14	0.23	0.021
	4	10835	0	0	0

Table II. Irradiation history input for RB and SPB ORIGEN-S calculations.

Reflector Region	Step	Days	$\phi_{thermal}$ (n/cm^2/s)	$\phi_{resonance}/\phi_{thermal}$	$\phi_{fast}/\phi_{thermal}$
RB	1	1200	8.823E+14	1.152	0.234
	2	18262.5	0	0	0
SPB	1	2400	8.152E+14	0.498	0.058
	2	18262.5	0	0	0

Table III. Primary radionuclides (Ci) in PB3 following discharge (largest values from TRITON` or ORIGEN-S).

Nuclide	Years Following PB3 Discharge					
	0.00	**2.60**	**4.96**	**9.67**	**19.67**	**29.67**
^{3}H	2.76E+05	2.38E+05	2.09E+05	1.60E+05	9.12E+04	5.20E+04
^{60}Co	2.00E+03	1.42E+03	1.04E+03	5.62E+02	1.51E+02	4.05E+01
^{55}Fe	5.73E+03	2.96E+03	1.63E+03	4.93E+02	3.89E+01	3.07E+00
^{14}C	1.13E+02	1.13E+02	1.13E+02	1.13E+02	1.12E+02	1.12E+02
^{63}Ni	4.62E+01	4.54E+01	4.46E+01	4.32E+01	4.03E+01	3.76E+01
^{137}Cs	2.93E+01	2.76E+01	2.61E+01	2.34E+01	1.86E+01	1.48E+01
^{137m}Ba	2.79E+01	2.60E+01	2.47E+01	2.21E+01	1.76E+01	1.39E+01
^{204}Tl	1.26E+02	7.82E+01	5.08E+01	2.14E+01	3.42E+00	5.47E-01
^{10}Be	1.38E+01	1.38E+01	1.38E+01	1.38E+01	1.38E+01	1.38E+01
^{244}Cm	1.64E+01	1.49E+01	1.36E+01	1.13E+01	7.73E+00	5.27E+00
^{85}Kr	1.80E+01	1.52E+01	1.31E+01	9.62E+00	5.04E+00	2.64E+00
^{90}Y	1.40E+01	8.34E+00	7.87E+00	7.01E+00	5.48E+00	4.28E+00
^{90}Sr	8.89E+00	8.34E+00	7.87E+00	7.01E+00	5.48E+00	4.28E+00
^{134}Cs	1.63E+02	6.82E+01	3.09E+01	6.34E+00	2.20E-01	7.63E-03
^{241}Pu	1.72E+00	1.52E+00	1.36E+00	1.08E+00	6.66E-01	4.11E-01
Total	3.12E+06	2.43E+05	2.12E+05	1.61E+05	9.16E+04	5.22E+04

Table IV. Primary radionuclides (Ci) in representative RB following discharge (ORIGEN-S).

Nuclide	Years Following RB Discharge (EOL ~ 102 GWd)							
	0	1	5	10	20	30	40	50
^{3}H	1.50E+05	1.42E+05	1.13E+05	8.56E+04	4.88E+04	2.78E+04	1.59E+04	9.04E+03
^{55}Fe	4.59E+03	3.56E+03	1.29E+03	3.62E+02	2.86E+01	2.26E+00	1.78E-01	1.41E-02
^{60}Co	4.38E+02	3.84E+02	2.27E+02	1.17E+02	3.15E+01	8.46E+00	2.27E+00	6.09E-01
^{63}Ni	1.99E+01	1.97E+01	1.92E+01	1.85E+01	1.73E+01	1.61E+01	1.50E+01	1.40E+01
^{14}C	6.24E+00	6.24E+00	6.24E+00	6.24E+00	6.23E+00	6.22E+00	6.21E+00	6.21E+00
^{125}Sb	4.70E+01	3.64E+01	1.32E+01	3.71E+00	2.93E-01	2.31E-02	1.82E-03	1.44E-04
^{204}Tl	1.61E+01	1.34E+01	6.43E+00	2.57E+00	4.11E-01	6.57E-02	1.05E-02	1.68E-03
^{85}Kr	3.10E+00	2.90E+00	2.24E+00	1.62E+00	8.49E-01	4.45E-01	2.33E-01	1.22E-01
^{137}Cs	1.79E+00	1.75E+00	1.60E+00	1.42E+00	1.13E+00	8.98E-01	7.12E-01	5.66E-01
^{137m}Ba	1.99E+00	1.66E+00	1.51E+00	1.35E+00	1.07E+00	8.48E-01	6.73E-01	5.34E-01
Total	8.91E+06	1.48E+05	1.15E+05	8.61E+04	4.89E+04	2.79E+04	1.59E+04	9.06E+03

Table V. Primary radionuclides (Ci) in representative SPB following discharge (ORIGEN-S).

Nuclide	Years Following SPB Discharge (EOL ~ 204 GWd)							
	0	1	5	10	20	30	40	50
^{3}H	1.09E+05	1.03E+05	8.22E+04	6.20E+04	3.54E+04	2.02E+04	1.15E+04	6.55E+03
^{60}Co	4.67E+02	4.10E+02	2.42E+02	1.25E+02	3.37E+01	9.03E+00	2.42E+00	6.51E-01
^{55}Fe	1.51E+03	1.17E+03	4.24E+02	1.19E+02	9.40E+00	7.42E-01	5.86E-02	4.63E-03
^{14}C	1.67E+01	1.67E+01	1.67E+01	1.67E+01	1.66E+01	1.66E+01	1.66E+01	1.66E+01
^{63}Ni	6.84E+00	6.80E+00	6.61E+00	6.39E+00	5.96E+00	5.56E+00	5.19E+00	4.84E+00
^{137}Cs	3.62E+00	3.53E+00	3.22E+00	2.87E+00	2.28E+00	1.81E+00	1.44E+00	1.14E+00
^{137m}Ba	3.96E+00	3.34E+00	3.04E+00	2.71E+00	2.15E+00	1.71E+00	1.35E+00	1.08E+00
^{85}Kr	4.97E+00	4.66E+00	3.60E+00	2.60E+00	1.36E+00	7.14E-01	3.74E-01	1.96E-01
^{204}Tl	1.41E+01	1.17E+01	5.64E+00	2.26E+00	3.60E-01	5.76E-02	9.21E-03	1.47E-03
^{10}Be	2.08E+00	2.08E+00	2.08E+00	2.08E+00	2.08E+00	2.08E+00	2.08E+00	2.08E+00
^{244}Cm	1.91E+00	1.84E+00	1.58E+00	1.31E+00	8.90E-01	6.07E-01	4.14E-01	2.82E-01
Total	2.28E+06	1.05E+05	8.29E+04	6.23E+04	3.54E+04	2.02E+04	1.15E+04	6.57E+03