Transmutation of Light Water Reactor-Discharge-Transuranics in a High Temperature Gas Cooled Reactor

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Abstract

An assessment of the General Atomics proposed Accelerator Transmutation of Waste concept using the high temperature gas reactor technology has been performed. In this concept, transuranic materials that are extracted from light water reactor spent fuel are fabricated into oxide fuel and burned in a critical and sub-critical accelerator driven gas-cooled transmuter. The transmuter operates in the critical mode for three cycles and then operates in a subcritical accelerator-driven mode for a single cycle. Both thermal and fast spectrum transmutation zones are utilized. The thermal zone is fueled with the transuranic oxide materials in the form of coated particles, which are mixed with graphite powder, packed into cylindrical compacts, and loaded in hexagonal graphite blocks with cylindrical channels. The fast zone is fueled with transuranic oxide materials in the form of coated particles from the graphite blocks that have been irradiated in the thermal zone for three critical cycles and an additional accelerator-driven cycle without the graphite powder. This mode of operation is intended to achieve high plutonium consumption in the thermal zone, and to consume the minor actinides in the fast zone. The Monte Carlo code MONK has been used to accurately model the geometrical details of the design to precisely account for the physics, and to assess the thermal transmuter performance, which are the subject of this paper.

The paper describes the transmuter and the major design parameters that can be altered to optimize the system design. Also, the Monte Carlo models developed and used for the neutronics analysis are discussed. The results of the parametric studies for the fuel block and the whole transmuter to understand the physics and the design analyses are given. Special attention was given for the burn-up analyses to assess the transmutation potential of this concept.

I. Introduction

An assessment of an Accelerator Transmutation of Waste (ATW) concept that uses the high temperature gas reactor technology has been performed. The concept was proposed by General Atomics [1]. This concept uses recycled transuranic (TRU) materials extracted from spent fuel, in a critical and sub-critical accelerator driven gas-cooled transmuter. The spent fuel was discharged from light water reactors (LWR). In this concept, the transmuter operates in the critical mode for three cycles and then operates in a subcritical accelerator-driven mode for a single cycle. The transmuter contains both thermal and fast spectrum transmutation zones. The thermal zone is fueled with the TRU oxide materials in the form of coated particles, which are mixed with a graphite powder, packed into cylindrical compacts, and loaded in hexagonal graphite blocks with cylindrical channels. The fast zone is fueled with the same fuel particles extracted from the cylindrical compacts that has been irradiated in the thermal region for three critical cycles and an additional accelerator-driven cycle without the graphite powder. This fuel management scheme is intended to consume most of the plutonium isotopes in the thermal-spectrum zone and the minor actinides in the fast-spectrum zone.

In this work, a special attention was given for developing accurate detailed geometrical models of the fuel blocks and for assessing the corresponding heterogeneity effects. The block geometry has several levels of heterogeneity that require proper treatment to predict the system performance. Another geometrical heterogeneity is also present in the system because of the annular configuration that employs inner and outer reflector zones. The MONK Monte Carlo code [2] has been used to allow an accurate explicit geometrical modeling and to assess the system performance. Both JEF2.2 and ENDF/B-VI nuclear data libraries were used in two forms: quasi-continuous energy representations (13193 or 8220 groups) and conventional multi-group libraries (172 or 69 groups). Also, the integrated burn-up capability of the MONK code was used to determine the feasibility of achieving high TRU consumption levels.

The study analyzed the effect of the major design parameters on the system performance using the developed geometrical models to understand the physics characteristics of this configuration. The results from the fuel block and the whole transmuter parametric studies are presented including power distribution and the detailed burn-up analysis.

II. System Description

The transmuter consists of a steel vessel housing, containing an annular transmutation region operating with a thermal neutron spectrum. This annular region contains the "fresh" TRU separated from light water reactor spent fuel. The TRU materials are contained in spherical TRISO-coated particles. These spherical particles consist of a 200-μm diameter $TRUO_{1.7}$ core, called kernel, surrounded by layers of graphite buffer (thickness 100-μm) to absorb gaseous fission products, pyrolitic graphite (thickness 35-μm), silicon carbide (thickness 35-μm) to serve as a stable barrier and pressure vessel, and an outside layer of pyrolitic graphite (thickness 40-μm). These particles are mixed with graphite powder and packed into cylindrical compacts. The compacts are loaded into cylindrical channels within hexagonal graphite blocks. These blocks also have channels for helium coolant flow and channels for introducing erbium burnable poison into the system. The block dimension is 36 cm flat-to-flat, and it contains 202 TRU channels, 108 coolant channels and 14 burnable poison (BP) channels, all the channels are arranged on a 1.88-cm triangular pitch. The fuel blocks are loaded into the fifth, sixth, and seventh radial rings of a hexagonal configuration. Three rings of graphite reflector are arranged both inside and outside this thermal region. The innermost layer is filled with fast fuel assemblies, composed of the TRU particles that have undergone four years of burning in the thermal region. The vertical configuration comprises ten active blocks stacked vertically. At the center of the configuration is the location for a spallation target used during the period of subcritical operation.

The transmuter operates in the critical mode for approximately three years. In this mode, the critical thermal region drives the fission process and limited transmutation events are expected in the fast region. After these three years, the thermal region becomes subcritical and is driven by the spallation target neutrons during the fourth year of the cycle. The local multiplication of spallation neutrons in the fast region might produce a significant fast flux thus helping the transmutation of the minor actinides. The plant has four 600 MWth transmuters, sharing one 15-MW beam accelerator.

The transmuter is cooled by helium with an outlet temperature of 850 °C. The heated helium is used in a direct-cycle gas-turbine-generator system. The high operating temperatures and the characteristics of the direct Brayton power conversion system allow electric generation with a high net thermal efficiency of ~47%.

III. Block Models and Analyses

The transmuter design includes several levels of heterogeneity effects that require proper treatment to obtain accurate performance predictions. The transmuter geometry consists of hexagonal prismatic blocks of graphite containing parallel vertical holes, arranged in a triangular pitch. These holes contain fuel or BP compacts and some vacant holes for helium coolant flow paths. The fuel and BP compacts consist of multi-layer ceramic-coated particles dispersed in a graphite matrix. Significant neutronics heterogeneities are created by these small particles. Fuel block heterogeneity arises from the heterogeneous arrangement of fuel, BP, and coolant channels in the graphite block. Another geometrical heterogeneity is due to the annular configuration that employs inner and outer reflector zones.

An explicit detailed model for the fuel block was developed using the MONK computer code. MONK has the capability to explicitly model the geometry under consideration and to perform criticality and burnup analyses in an integrated manner. The particles are modeled as a hexagonally close-packed lattice of spheres. The lattice forms a regular octahedron with a cylindrical boundary to represent the compact. MONK criticality calculations were performed with quasi-continuous energy and multigroup data sets. The quasi-continuous energy data sets are processed in a fine energy mesh (13193 or 8220 groups). The multigroup libraries are processed in a much coarser set (172 or 69 groups). The burnup analyses use the coarser data sets. The nuclear data libraries are based on JEF version 2.2 or ENDF/B-VI.

Three MONK models were developed for the heterogeneity analyses. These are:

- An explicit block model with explicit representation of the multi-layers of the fuel and the BP particles inside the compacts,
- A block model with homogenized particles inside the compact, and
- A block model with homogenized compacts inside the block.

Figure 1 shows the explicit MONK block model and an enlarged section is shown in Figure 2. The explicit fuel particle model is shown in Figure 3.

The difference in the block k_∞ provides a measure for the heterogeneity effect. Table I summarizes the results, which were obtained with the use of the 13193-groups quasi-continuous energy and the 172-groups nuclear data libraries. The volume packing fractions for the fuel and the BP particles are 0.1238 and 0.1, respectively.

In this case, the fuel block is loaded with fuel compacts and helium coolant channels, without erbium burnable poison compacts. The results show a strong heterogeneity effect. The difference in the results between quasi-continuous energy libraries and the multigroup libraries is due to the difference in thermal treatment of the carbon nuclear data. The quasi-continuous energy libraries are lacking the $S(\alpha,\beta)$ treatment for carbon. The homogeneous models give inaccurate k_∞ values because these models significantly under predict the self-shielding of the strong absorption resonances in the plutonium isotopes, particularly Pu-240. This is caused by the fact that the fuel particle dimensions are relatively large compared to the mean free path of neutrons in the low-energy-lying resonances of these isotopes. Because of this effect, the inner zone of the particle is shielded from neutrons by the outer zone and simple homogenization does not account correctly for the self-shielding effect.

Similar analyses were performed for the BP particles as shown in Table II. In this case, the self-shielding change caused by the Er-167 absorption resonance explains the change in the block k_∞. The burnable poison heterogeneity effect is smaller than the corresponding value for the TRU materials because the block has only 14 BP compacts relative to 202 TRU compacts.

For both the fuel and BP compact cases, the heterogeneity effects were found to be dependent on the particle composition and the packing fraction. The difference in k_∞ between the homogeneous and explicit models decreases as the packing fraction increases for fixed particle size or as the fuel kernel radius decreases for fixed packing fraction.

One important feature of this transmuter is the net negative temperature coefficient. As the transmuter temperature increases, the neutron spectrum peak shifts toward the absorption resonance of erbium-167. This results in more neutron absorption in erbium-167. The analysis was performed in steps to define the contribution of each material to this effect using the explicit MONK model with the 172-groups nuclear data library. In this case, the packing fractions are 0.15 and 0.1 for the fuel and the burnable poison, respectively. The first case has all the materials at 293.16 K. In the second case, the fuel particle temperature was changed to the average operating temperature without changing the temperature of the other materials. The third case is similar to the second case with the graphite temperature of the compact changed to average operating temperature. The last case changed the graphite block temperature to the operating temperature. Table III results show the results for these cases. Heating the graphite material increases the neutron absorption in erbium-167, which results in a negative temperature coefficient. This enhances the safety performance of the system.

Several parametric studies were performed [3] to characterize the block reactivity. For example, Figure 4 shows the block k_∞ as a function of the fuel-particle packing fraction for three different BP packing fractions, at the cold condition. The variation of k_∞ versus the fuel packing fraction shows a peak at low fuel packing fraction. The shift in the neutron spectrum with the packing fraction is responsible for this trend. As the packing fraction decreases, the carbon-to-heavy-metal ratio increases and leads to an increase in the neutron thermalization causing the neutron spectrum to become softer. This result in fewer neutron captures, which enhances the neutron utilization. The improved utilization of neutrons increases Pu-239 fission rate, which enhances the k_∞ as the fuel packing fraction decreases. The packing fraction corresponding to the highest k_∞ differs for the three curves. As the BP loading increases, the spectrum hardens due to the relative neutron absorption increase in Er-167 and Pu-240. Also, less fuel material is required to achieve the same k_∞. The k_∞ increase with the fuel packing fraction, below the peak value of k_∞, is due to the concentration increase of the fissile elements causing more fission reactions.

The block power distribution was also calculated using the explicit MONK model at the cold state (293.16 K). The maximum power occurs in a TRU compact located close to the block boundary because of the extra (non-cell) graphite present in this zone, which causes a softer neutron spectrum. The peak to the average is 1.07.

The block power distribution has very little peaking due to the BP distribution in the block, which flattens the block power distribution.

Table I. TRU compact heterogeneity effect without burnable poison

Model	$k_\infty{}^1$	$\Delta k^1_\infty/k^1_\infty$, %	$k_\infty{}^2$	$\Delta k^2_\infty/k^2_\infty$, %
Explicit modeling	1.2764	--	1.2534	--
Homogenized particles	1.1101	-13.02	1.1004	-12.21
Homogenized compact	1.0928	-14.38	1.0847	-13.46

1 MONK analyses with the 13193-groups quasi-continuous energy nuclear data library
2 MONK analyses with the 172-groups nuclear data library

Table II. BP MONK heterogeneity effect with the 13193-groups quasi-continuous energy nuclear data library

Model	k_∞	$\Delta k_\infty/k_\infty$, %
Explicit modeling	1.1864	
Homogenized particles	1.1649	-1.81
Homogenized compact	1.1604	-2.19

Table III. Temperature effect on the lattice performance

Burnable poison model	K_∞	Relative difference%
Cold Conditions	1.1327	
Hot Fuel Particles	1.1112	-1.90
Hot Compact	1.0954	-3.29
Hot Block	1.0562	-6.75

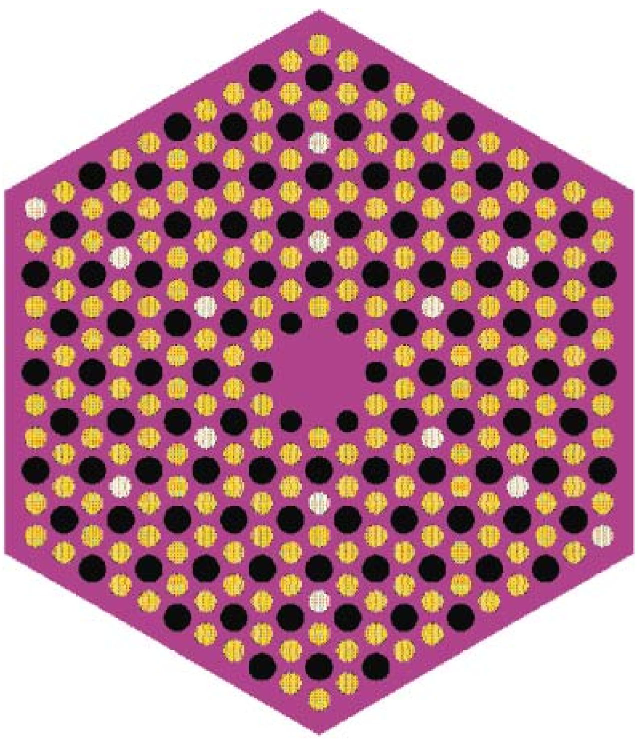

Figure 1. Thermal block model featuring the different channels

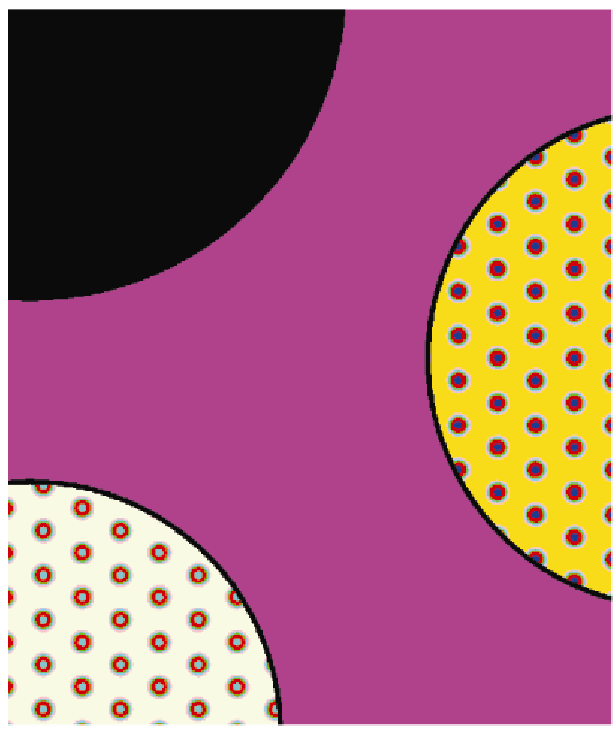

Figure 2. Enlarged block section featuring a section of the coolant channel (Top left), TRU compact (Right), and burnable poison compact (Bottom left)

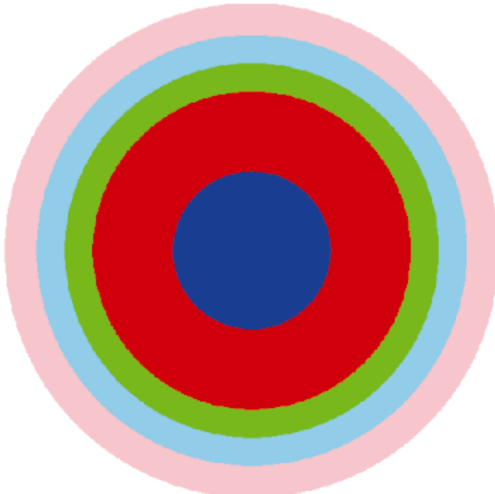

Figure 3. Fuel particle model

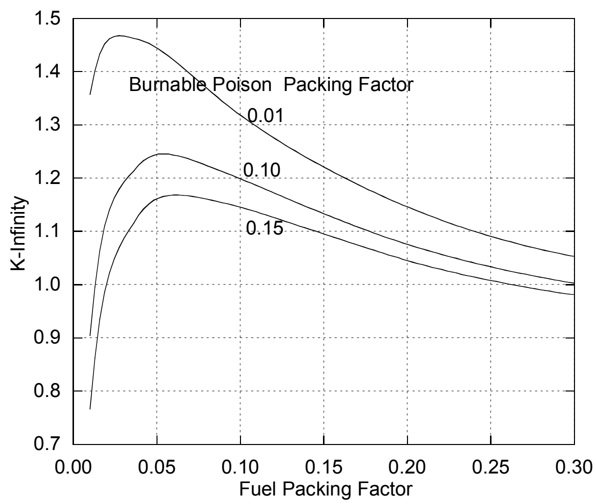

Figure 4. Block K_{∞} as a function of the fuel-packing factor for different BP packing factors

IV. Transmuter Model and analyses

A three-dimensional transmuter model was developed for performing the neutronics analyses with MONK code. The model has explicit representation for the fuel and the burnable poison particles including all the geometrical details. The fuel blocks are located in rings six to eight. Rings one to five and nine to eleven contain graphite reflector blocks. Axially, the whole length of the active core (793 cm), and additional lower and upper graphite reflector blocks are modeled. A vacuum boundary condition is used for all external surfaces. A cylindrical boundary is used for the radial reflector to match the actual configuration. A horizontal cross section of the model is shown in Figure 5.

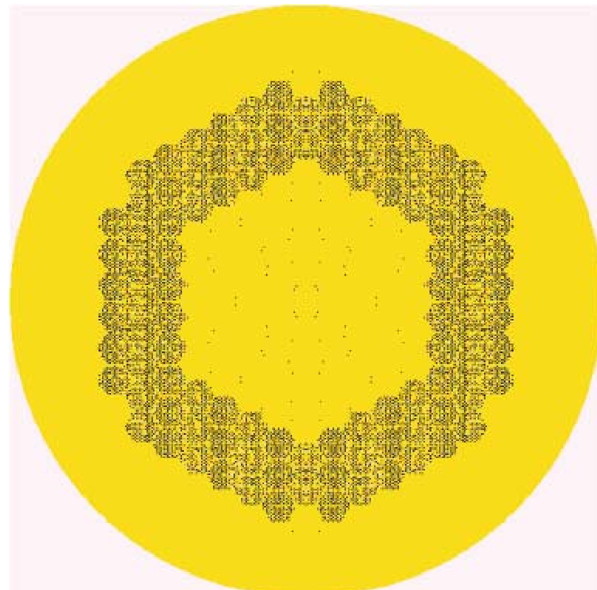

Figure 5. Horizontal cross section of the MONK transmuter model

The transmuter power distribution is calculated for fresh fuel blocks with a heavy metal loading of 787 kg and an Er-167 loading of 27.7 kg. The results show the peak power occurs in the sixth ring because of the central graphite reflector. The central graphite causes a softer neutron spectrum that enhances the fission rate. The peak to average is 1.22 as shown in Table IV.

Table IV. Normalized transmuter power distribution per fuel block shown for 60º section

R	1.039	0.980	1.005	1.005	0.980	1.039	R
0.895	0.830	0.845	0.836	0.845	0.830	0.895	
1.026	1.222	1.201	1.201	1.222	1.026		
R	R	R	R	R			
R	R	R	R				
R	R	R					
R	R						
R							

MONK burnup calculations were performed for the transmuter using the explicit geometrical model shown in Figure 5. The calculations were performed at the average operating temperatures of each material with the 172-groups nuclear cross section library. The fuel compact, the graphite block, and the reflector block temperatures are 1043.16, 993.16, and 993.16 K, respectively. The explicit representation of the geometry was maintained in the burnup calculations. A constant fission power of 600 MW was used in the calculations for 900 days. The packing factors for this configuration are 12.87 and 10% for the TRU and the BP, respectively.

The first step in this analysis was to define the appropriate time step (burnup interval) between subsequent flux calculations because of the large computer time required for each flux calculation. However, the use of a large time step reduces the accuracy of the results. A parametric study was performed to determine the effect of the time steps on the results. Several time steps were used as shown in Figure 6. The results show that the fresh transmuter has K-effective of 1.1005. K-effective drops to 1.0 after about 500 days. At 900 days, K-effective is very low for this configuration. The burnup parameters converge as the time step is reduced. The results from 12.5 and 25 days time steps are very close. About 44% of the TRU are burned in the first 500 days, and 80% are burned at 900 days. At K-effective of 0.9, the TRU burnup is 61%. Er-167 is consumed at much faster rate as shown in Figures 6 and 7. Only Er-167 is acting as burnable poison and it is converted to Er-168, as shown in Figure 7. Further investigations are required to define the optimum TRU and Er packing factors to achieve 900 days of operation, if it is required, with adequate reactivity and Er-167 concentration for negative temperature coefficient during the critical operating period.

The changes in the atomic concentrations of the different TRU isotopes as a function of the operating time are shown in Figures 8 and 9. During the critical operation, Pu-239 decreases linearly with the operating time while Pu-240 decreases slowly. During the subcritical operation, the remaining Pu-239 decreases slowly while Pu-240 decreases linearly with the operating time. Pu-241 increases to reach a peak value at about 380 days then it decreases linearly with the operating time. Pu-242 increases slowly during the operation. Pu-237 and Pu-238 decreases slowly during the operation.

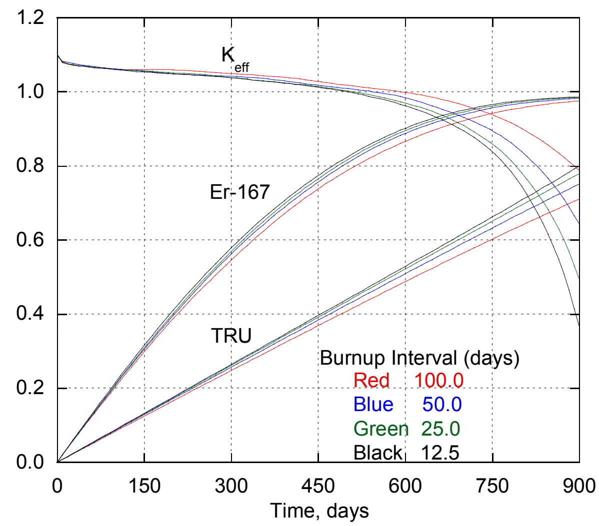

Figure 6. Main burnup parameters as a function of the operating time for different burnup intervals

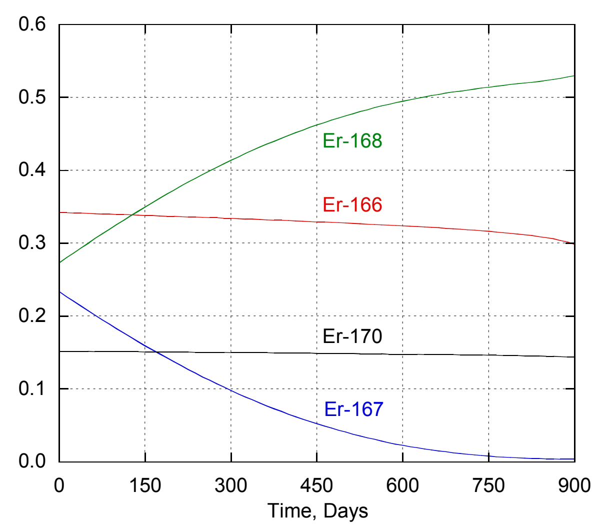

Figure 7. Relative atomic concentrations of the Erbium isotopes as a function of the operating time

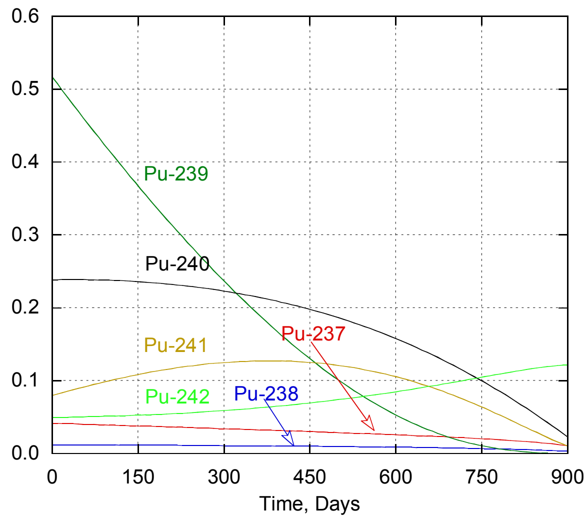

Figure 8. Relative atomic concentrations of the plutonium isotopes as a function of the operating time

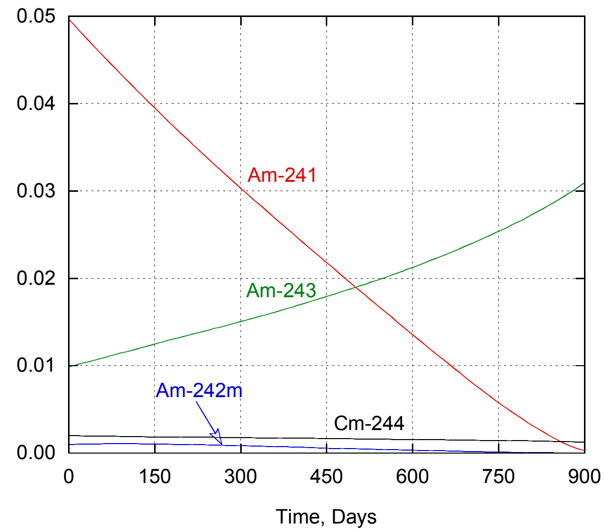

Figure 9. Relative atomic concentrations of the americium and curium-244 isotopes as a function of the operating time

Am-241 decreases linearly during the operation while Am-243 increases linearly. Am-242m and Cm-244 decrease slowly during the operation. Further analyses are required to define transmuter loading with TRU and BP materials and the fuel block parameters for optimum transmutation.

V. Conclusions

Several conclusions were obtained from this study. First, accurate geometrical models for Monte Carlo analyses were developed for the fuel block and the whole transmuter based on the high temperature gas reactor technology with TRU fuel in the form of coated particles. Second, these models were used successfully to perform detailed physics analyses including burnup. Third, the results show the need for using detailed geometrical models including explicit presentation for the multi-layers fuel particles because material homogenization introduces significant errors in the performance parameters. Fourth, the parametric studies show the potential for adjusting the block design to optimize the transmuter performance and the fuel cycle for achieving high transmutation rate.

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