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Reactor Physics Characterization of Transmutation Targeting Options in a Sodium Fast Reactor

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ABSTRACT

In sodium fast reactor designs, negative reactivity feedback is accomplished mainly through parasitic capture in U-238. However for an efficient minor actinide burning system, it is desirable to reduce or eliminate U-238 entirely to suppress further transuranic actinide generation. Consequently, reactivity feedback is accomplished by enhancing axial neutron streaming during a loss of coolant void situation. This is done by flattening "pancake" the active core geometry. Flattening the reactor also increases axial leakage which removes neutrons that could otherwise be used to destroy minor actinides. Therefore, it is important to tailor the neutron spectrum in the core for optimized feedback and minor actinide destruction simultaneously by using minor actinide and fission product targets.

1. INTRODUCTION

The radiotoxicity and heat load attributed to long lived fission product and minor actinide Spent Nuclear Fuel (SNF) isotopes are the limiting factors for repository capacity. Repository space is maximized when these isotopes are removed from the nuclear fuel cycle's waste stream through a process called transmutation. Long lived fission product isotope transmutation can be accomplished by adding neutrons to the long lived atomic nucleus until a less stable nucleus is created with a disproportionate neutron to proton ratio, which translates into a shorter half-life. Similarly, minor actinides (MA) may be transmuted to shorter half-life isotopes using "thermal spectrum" neutrons. Thermal spectrum reactors have a high MA neutron capture rate. Even so, neutron capture efficiently removes the primary SNF isotopes Np-237 and Am-241. However, the transmuted product actinides also have long half-lives. Thermal transmutation concentrates higher mass actinides. Many of these isotopes (Cm-245 and heavier), produce high gamma and neutron radiation fields when concentrated which presents a fuel handling issue. The higher mass actinide accumulation is less with "fast spectrum" neutrons because the relative fission-to-absorption cross section ratios are higher than in the thermal spectrum. This implicates Sodium Fast Reactors (SFR) as the means for MA destruction.

The two most basic sodium reactor control mechanisms are the Doppler feedback provided by fertile uranium in the fuel and blankets; and the increase in axial neutron streaming that occurs during coolant voiding. Generally for SFR with high MA loaded fuels, there is a tradeoff between the optimal Doppler and void coefficients and the attainable actinide destruction efficiency. This tradeoff stems from the fact that the mechanisms commonly used to remove neutrons from the reactor during transients also remove neutrons in steady state operation. For example, enhancing axial streaming through core geometry alterations can make the void coefficient more negative but the increased overall leakage removes nuetrons that could have lead to MA fission. Alternatively, using fertile U-238 to make the Doppler coefficient more

negative inherently produces further long-lived transuranic actinides by parasitic capture in the same cross section resonances that provide the beneficial feedback. Adding moderating materials to the core can increase the neutronic importance of these epithermal resonances by softening the neutron spectrum. However, softening the spectrum over the entire core also reduces the MA fission-to-capture ratio.

In all of these cases, more fissile plutonium is required to compensate for reactivity lost by the modifying strategy. When plutonium is involved in much larger concentrations than the other transuranics; the increase in plutonium destruction detracts from the MA destruction. This also lends to increased cycle reactivity swing.

2. METHOD OF CALCULATION

The, Los Alamos code package MONTEBURNS was used to model the transmutation and reactor physics characteristics. MONTEBURNS is a coupled Monte Carlo and fuel depletion code. It uses flux tallies generated in MCNP to produce single group fission and transmutation cross sections. These cross sections are used for neutron flux determination at a particular time step. These fluxes and cross sections are then fed into the Oak Ridge fuel depletion code ORIGEN2 for burnup and decay calculations to the next time step. Then MONTEBURNS feeds the updated isotopic composition back to MCNP and the process begins anew.

3. REFERENCE CORE DESIGN DESCRIPTION

It was desired that the reference core design have the size and thermal power of a commercial class fast reactor. Therefore, the S-PRISM core layout and thermal power rating was adopted. A 271 hexagonal fuel assembly design with a 12 cm pitch was used. The pin-cell within this assembly had a 1.26 pitch-to-diameter ratio with a 5.8 mm pin diameter and a 4.4 mm pellet diameter. The pellet-to-clad gap was sodium bonded. The fuel composition was metallic with a volume fraction composition of 15% TRU, 15% depleted uranium and 70% zirconium. The fuel's transuranic (TRU) isotopic vector is that for pressurized water reactor (PWR) spent nuclear fuel which was initially enriched to 5 w/o U-235/U-238, irradiated to 60 MWD/kg and cooled for 5 years. The reactor's fuel region was 100 cm tall with a 200 cm core diameter. The top 20 cm of the fuel pin was loaded with axial blanket compositions.

Due to computer memory and speed limitations, the fuel, bond, coolant and structure materials were smeared within the fuel assembly. Considering the long mean-free-path for fast neutrons (mfp>20cm), the spatial self-shielding was treated on the dimensional level of the fuel assembly. Also, since the reference core design has $1/6^{th}$ radial symmetry; reflective boundary conditions were used to model a $1/6^{th}$ slice of the entire core. All transuranic atoms with a concentration higher than 0.1% of the total fuel mass were tracked by the Monteburns code.

Because, this study is intended as a scoping calculation to explore target designs, only a single batch startup core was evaluated. Fuel shuffling and the equilibrium reactor fuel cycle scenario will be evaluated in a later publication. The active driver fuel used for this discussion has a low uranium concentration and therefore may be considered low fertile. The small uranium content limits the available cycle length required for completely destroying minor actinides in the

targets[1]. A discussion on the optimal uranium concentration to achieve a more appropriate cycle length follows in a later section.

4. VOID COEFFICIENT AND TECHNETIUM TARGETS

A SNF isotope that has very similar cross section resonances to U-238 is Tc-99 [1]. Tc-99 is a long lived SNF isotope with a reasonably high radiotoxicity. Tc-99 is also a UREX process waste stream which would require long term storage if not transmuted in a SFR. Tc-99 is a long lived fission product ($T_{1/2}$=2.111E5 yr) allowing its destruction in the reactor to be profitable with regards to repository storage. Therefore, Tc-99 may prove useful as an alternative SFR resonance absorber. As can be seen from Figure 1, the epithermal absorption resonances for both Tc-99 and U-238 are very similar in magnitude and spacing [2].

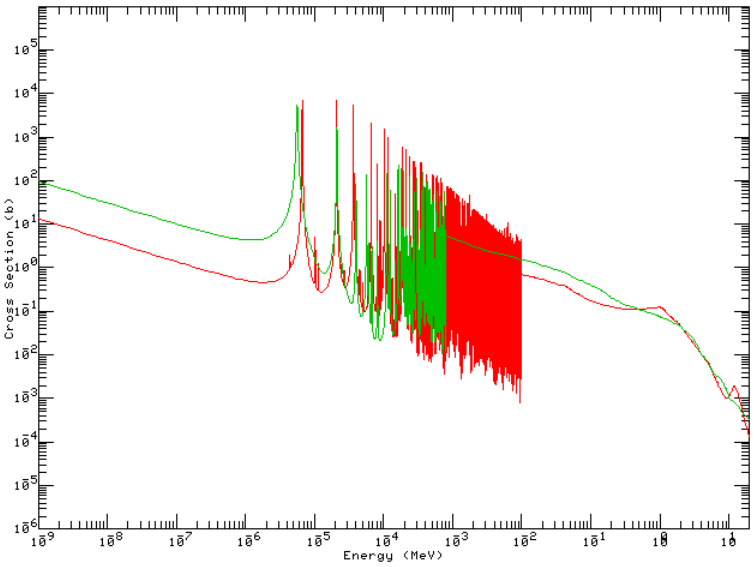

Figure 1: Comparison of U-238 (red) and Tc-99 (green) total absorption cross sections.

Because this isotope is not fertile, it may be used to make the void coefficient more negative without breeding further transuranics. This SFR Tc-99 application is analogous to using burnable poisons in light water rectors. The difference is that the important capture resonances are in the epithermal not thermal energies and the 1/v component is orders of magnitude less than the thermal absorbers B-10 or Gd-155.

Unlike most thermal burnable poison applications, homogeneous fuel compositions with thermal or epithermal absorbers can actually create undesirable reactivity feedback coefficients. This is because the Tc-99 resonances can self shield the minor actinides' capture resonances thus causing localized spectral hardening in the fuel [2][[3]. This hardening of the flux increases the minor actinides' fission-to-absorption ratio during a transient. The resulting increase in fast multiplication may supersede the parasitic capture in epithermal resonances. This energy self-

[1] The nature of the MCNP and ORIGEN coupling allows the fuel to be depleted beyond the available reactivity of a more realistic fuel irradiation time.

shielding effect may be avoided by incorporating the technetium loading in standalone target assemblies separate from the driver fuel. This separation increases the absorber cross section importance by forcing neutrons born in one driver assembly to cross the technetium target assembly before interacting with fuel in an adjacent assembly. This spatial self-shielding effect prevents driver assemblies that are separated by the technetium assembly from communicating with one another during spectrum hardening.

An internally blanketed core design similar to the S-PRISM design was considered. Figure 2 shows the core radial profile. Tc-99 targets are included in the internal radial blanket positions. The driver fuel was assumed to be composed of a uranium and molybdenum alloy. Technetium and molybdenum are neighboring transition metals on the periodic table. Also, Molybdenum's manufacturability and irradiation tolerance led to it's alloying in driver and blanket fuel for the Enrico Fermi Reactor [4]. Uranium volume fractions of 30, 60 and 90 v/o were considered. A comparison is made with these all uranium compositions and a 30Tc/70Mo, 90Tc/10Mo and a 15Tc/15U/70Mo alloy. Also considered was a hypothetical 15Tc/15U/65La's/5Mo alloy. The lanthanide isotopic vector was for the same discharged PWR fuels as the actinides. The void scenario was achieved by removing the coolant while leaving the bond sodium in the fuel/clad gap.

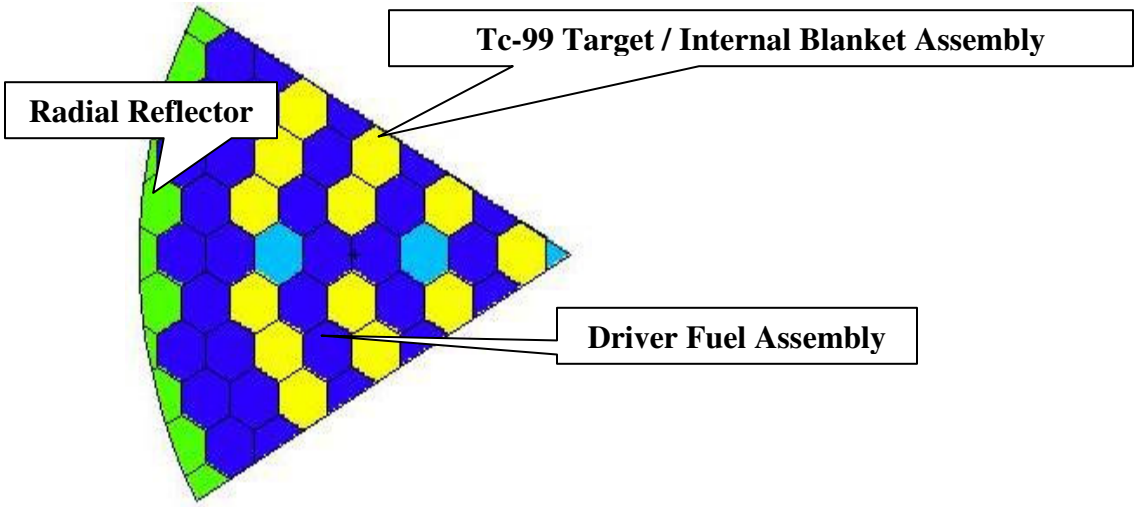

Figure 2: Driver fuel and Tc-99 target assembly radial core profile.

Figure 3 shows the neutron flux distribution change between void and non-void for the 30U/70Mo case. Notice the shift in the neutron spectrum to higher energies in the voided case. The reduced neutron down-scattering caused by the sodium void reduces the flux below 5 KeV.

In addition to the percent change in multiplication factor, a capture reaction rate spectrum was found for each case as a function of neutron incident energy. The incident energy reaction rate distribution was attained by using the cross section multiplier card in MCNP. The result is a continuous spectrum that weights the tallied neutron flux from Figure 3 with the continuous capture cross section plots from Figure 1. Figures 4a and 4b show the incident energy capture rate spectrums for the 30U/70Mo and 90U/10Mo against that for the 30Tc/70Mo and 90Tc/10Mo cases. These plots show that the sodium void has the strongest reduction on the

neutron capture rate at energies below 10 KeV. It is important to observe from Figure 1 that 10 KeV also marks the beginning of the U-238 unresolved resonance region. The loss of sodium reduces the level of down-scattering. Thus the epithermal flux and capture rate is reduced.

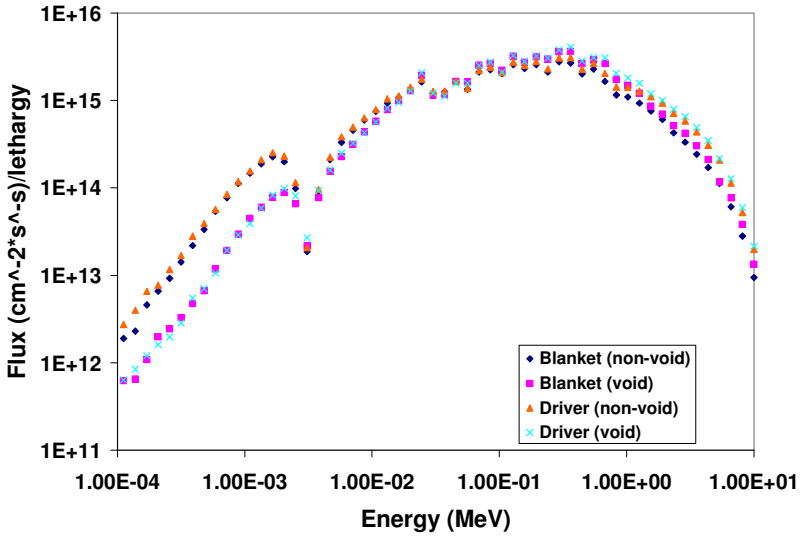

Figure 3: Flux comparison for 30U/70Mo blanet in the void vs non-void condition.

The centroid energy for the area under each curve in Figure 4 was found in order to quantify the level of spectrum hardening. Using this data, the percent change in centroid energy from non-void to void for each blanket composition was then calculated. Figure 5 shows the percent change in multiplication constant as a function of percent change in centroid energy.

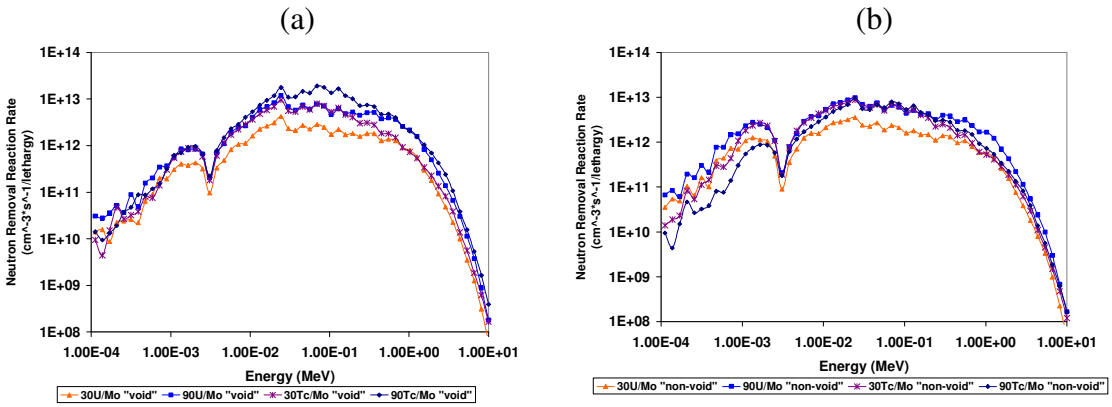

Figure 4: Neutron capture reaction rates as a function of incident neutron energy for: (a) voided core and (b) non-voided core.

As can be seen from Figure 5, increasing the absorber atom loading in the blanket has the most effect on reducing the percent increase in k-effective. This is because the resolved and un-resolved resonance capture rate is monotonically increased as the atom density is increased. The overall capture rate is increased until the loss of sodium's spectrum shift has little effect on the overall reactivity. This is the case for the 90Tc/10Mo and 90U/10 Mo cases. The sodium loss

creates a percent increase in the centroid energy. However, the percent increase is not as great for increasing absorber atom loading.

In the case of uranium, this is related to the U-238 resolved resonances starting at 10 KeV. The U-238 resolved resonance region ends an order of magnitude higher than that for Tc-99. Therefore, uranium is more visible than Tc-99 to neutrons between 10 KeV and 1 KeV. This is also the range where the loss of down-scattering in sodium has the largest change to the fast reactor's neutron spectrum (Figure 3). Since the blanket separates adjacent rows of driver, increasing the uranium loading also increases the resonance capture for neutrons crossing the blanket. However, as the uranium loading approaches 100% of the blanket assembly, the increase in spatial self shielding becomes less and less. This would explain the decreasing proximity for increasing uranium loading for the points in Figure 5.

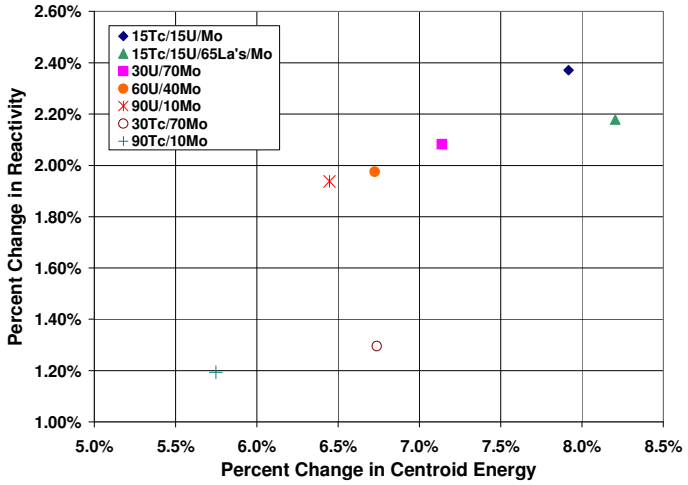

Figure 5: Percent increase in multiplication constant "k-effective" compared with percent change in capture rate centroid energy over a range of resonance absorber compositions.

The Tc-99 resolved resonance range ends at about 1 KeV. The fast reactor's neutron flux at this energy is only about 1E14 $cm^{-2}s^{-1}$ which is only 3% of the highest flux over the entire spectrum. Only about 5% of all the capture reactions occur below this energy. Conversely, about 25% of the capture reactions occur below 10 KeV. Therefore, Tc-99 is not as useful of a resonance absorber as U-238 in the metallic fueled fast reactor spectrum. However, the Tc-99 un-resolved resonance cross section is higher than U-238 as can be seen in Figure 1. Also the atom density for pure technetium is approximately 7E-2 atom/b*cm compared to 4.8E-2 atom/b*cm for metallic uranium. These two factors combined create the high capture rates above 10 KeV for the technetium loadings shown in Figure 4a and 4b.

Lack of resonances explains the small change in the 90Tc/10Mo capture spectrum shape (E<10 KeV) between Figure 4a and 4b. The 90U/10Mo capture rate decreases at E<10 KeV. This decrease is from the loss in down-scattering which shifts neutrons to energies higher than where the U-238 resonances can absorb. The same lack of down-scattering increases the 90Mo/10Mo capture rate more than for 90U/10Mo at E>10 KeV. This relative difference is from more neutrons being absorbed in Tc-99 un-resolved resonances. The smaller 90Tc/10Mo percent change in centroid energy in Figure 5 is a result of the capture rate always being centered in the

un-resolved resonance range despite loss of down-scattering. Since the technetium target's spatial self-shielding is centered in the un-resolved resonance region; an increase in neutron multiplication from absorptions in the driver fuel at these energies is minimized.

5. REACTIVITY INVESTMENT AND MINOR ACTINIDE TARGETS

The spent nuclear fuel isotopes Np-237 and Am-241 may also be advantageously used as a burnable reactivity control material. Americium loaded into a thermal spectrum fuel matrix, removes neutrons by absorption in broad thermal spectrum (0.3, 0.6, 1eV) cross section resonances. In addition to being a strong thermal neutron absorber, americium is also fertile in the sense that some fissile Am-242 and also plutonium isotopes are generated by its transmutation.

For every one in ten neutrons absorbed into Am-241, the long lived ($T_{1/2}$=141a) Am-242^m isotope is created (See Figure 6). The fission-to-absorption ratio for Am-242^m is quite high and fissions by this isotope early in fuel irradiation provides for unburned odd plutonium isotopes to remain until later in fuel life. The conserved plutonium allows for an extended reactivity limited burnup. Ninety percent of neutron absorptions in Am-241 lead to the Am-242 ground state. The short lived ($T_{1/2}$=16.2hr) Am-242 decays into Cm-242 by beta decay with a yield of 83%. Cm-242 subsequently decays with a half life of 163 days into Pu-238. The other 17% of Am-242 decays by electron capture into Pu-242. Pu-242's transmutation path leads to Cm-244 and higher mass actinides. The curium isotopes heavier than Cm-244 are significantly long lived. However, these isotopes also have a high fission to absorption ratio in a fast spectrum. When Np-237 absorbs a neutron it becomes the short lived ($T_{1/2}$=2.117d) Np-238 isotope which beta decays into Pu-238. Np-238, though short lived has a large fast fission to absorption ratio (fiss/abs>90%).

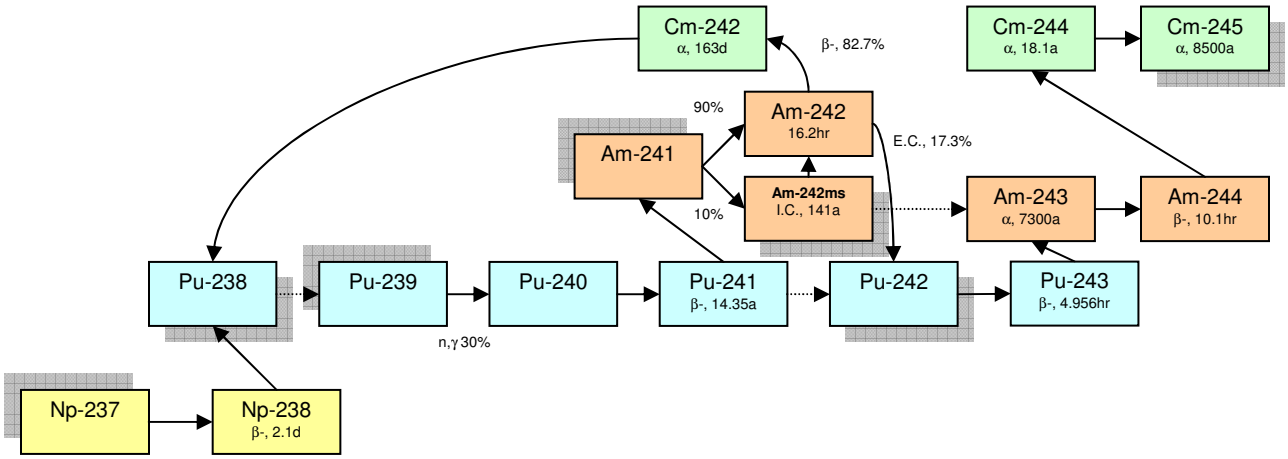

Figure 6: Important initial SNF isotopes (shadow above box) Np-237, Pu-239 and Am-241 and their transmutation endpoints Pu-238,242 Am-242^m and Cm-244 (shadow below box).

All of these transmutation products accumulate more readily in a thermal rather than a fast spectrum. This is because their thermal fission-to-absorption ratios are smaller in the thermal than the fast spectrum. But after successive neutron bombardment, product isotopes with a higher fast fission-to-absorption ratio than the initial material are generated. For example, in a thermal spectrum, the even numbered plutonium isotopes can not have a large fissile contribution

because their thermal cross section is too small to be useful. In a fast spectrum, the fission to capture ratio for Pu-238,242 is higher than for Np-237 and Am-241. Therefore, it is neutronically advantageous to transmute neptunium and americium into even plutonium isotopes. This "fast" fertile nature of neptunium and americium is useful as a fast reactor U-238 blanket replacement.

Thermalized MA targets in a sodium reactor core design can breed even isotope plutonium while simultaneously absorbing excess neutrons [5]. In a fast reactor, the breeding of transmuted isotopes can be accomplished with a minor actinide fast flux trap within the core. The goal of such a flux trap would be to recover the fast flux leakage and moderate it to a thermal spectrum before irradiating a minor actinide rich target. The resulting plutonium isotopes have higher "fast" fission-to-capture ratios than the initially loaded minor actinides. The relationship of this flux trap to the active driver is depicted by the reactor concept in Figure 7 dubbed "Heterogeneous Transmutation Fast Reactor".

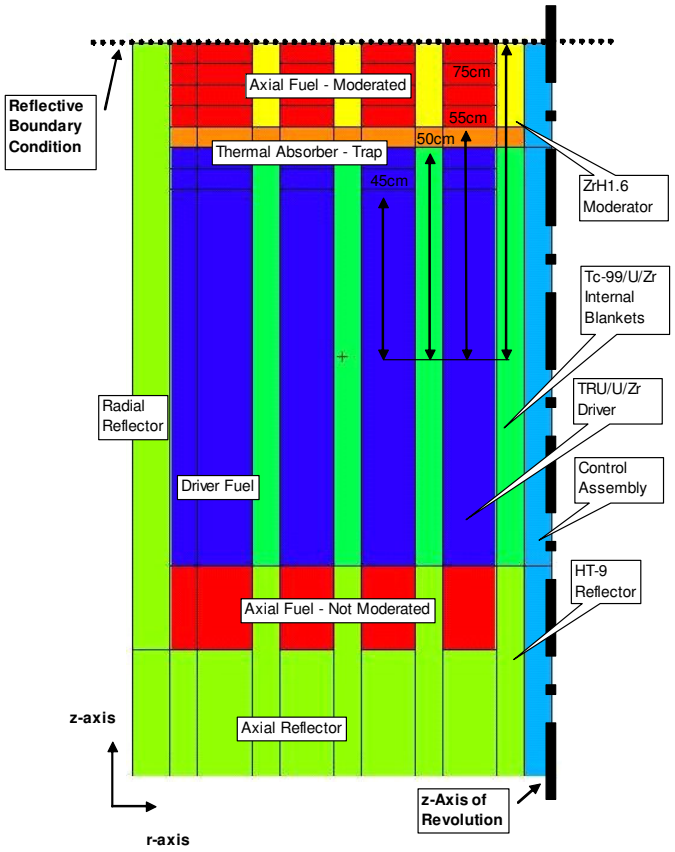

Figure 7: r-z reactor profile for SFR concept capitalizing on americium thermal transmutations in the upper axial fuel zone.
Note: The radial profile for this reactor is the same as Figure 2.

The principal design advantage of a fast flux trap is in its heterogeneity. The active driver core is allowed to be fueled with low minor actinide content fuel while the total minor actinide content of the overall core remains high. Also, the active driver portion of the core is allowed to have a high axial leakage which improves neutron streaming during the void scenario. This axial

leakage may be further increased by increasing the sodium volume percent in the core. Either way, active core neutrons leaked into axial targets during steady state operation are simply invested in transmutation processes as opposed to being lost.

To increase the proliferation resistance of the fuel, neptunium and plutonium are lumped together in the active driver fuel and not placed in targets. Americium and curium target rod bundles placed adjacent to $ZrH_{1.6}$ moderating rod bundles in the axial blanket location above the driver fuel are considered. The targets are exposed to fast flux from below and thermal flux from the side. This fuel configuration is different from earlier studies where the hydride material was used in the fuel matrix or placed in the fuel assembly. The intimate proximity of hydride to the hot fuel raised some concern over hydrogen dissociation from the matrix [6]. The proposed moderator design geographically separates the hydride material from the fission heat source. Figure 6 shows the upper portion of the internal radial blanket assemblies filled with $ZrH_{1.6}$. To prevent power peaking in the driver near the moderator, a thermal absorber such as cadmium, silver/indium or SNF lanthanides is used to separate the moderated upper axial zone from the driver.

This configuration demonstrates that fast neutrons leaked from the driver are invested in MA transmutation for even isotope plutonium breeding. Excess neutrons are shed from the active driver fuel by axial leakage for MA target bombardment. The investment in even plutonium isotope generation creates available reactivity to be used later in fuel life. This also has the advantage of removing excess reactivity early in fuel life. Figure 8 shows that the MA blanket has a similar effect on the reactivity curve as a purely depleted uranium blanket without moderation.

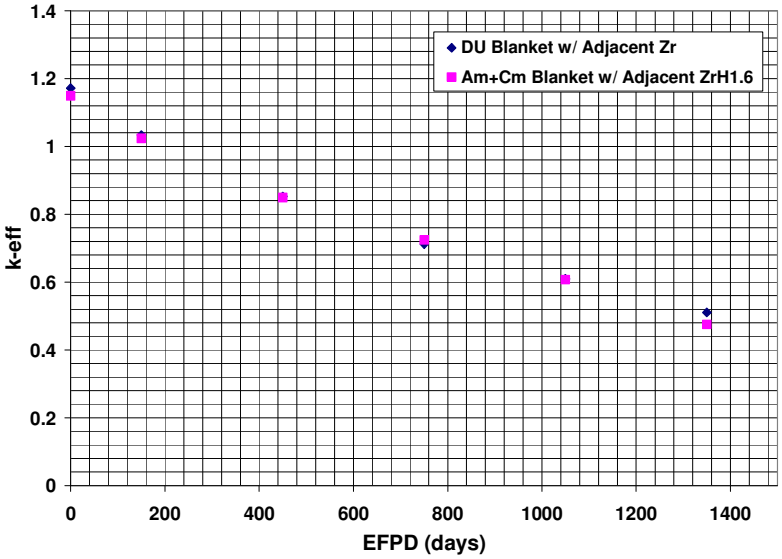

Figure 8: Startup core reactivity curve for a 15Am/15Zr verses a 40U/60Zr axial blanket. Note: The active driver fuel is a 15TRU/15U/70Zr composition.

The reactivity curves for the MA and DU blanketed cores lie on top of one another and have a reactivity limited irradiation time of 240 Effective Full Power Days (EFPD). Assuming the core is loaded with five batches, the total fuel irradiation time at equilibrium is 400 EFPD. Therefore,

the DU reactivity curve departure from the MA curve at 1360 EFPD in Figure 8 will not be reached.

The even isotope plutonium atoms have a higher fission-to-absorption ratio than americium. This allows fast neutrons streaming into the target to burn off the transmuted plutonium which reduces accumulation of Pu-238. Neutrons absorptions in Pu-238,242 that do not result in fast fission convert even into odd isotope plutonium and eventually curium where the fission-to-absorption ratio is even greater than the even plutonium isotopes. In this way a better MA incinerator is created by breeding increasingly more fissile material with every neutron capture. Unlike purely thermal or fast systems, isotopes are equally exposed to neutron fluxes at both energy ranges allowing capture and fission reactions to work together, transmuting and burning fertile MAs in situ. Figure 9 shows the incident energy reaction rate spectrum for fissions in Pu-238, Pu-239 and Am-241 and the incident energy capture rate spectrum for Am-241.

In the active core, the flux is entirely fast. Therefore, the incident energy for fission reactions is in the fast flux range. In the targets, thermal neutron capture in Am-241 is higher in the epithermal range resulting in a higher overall Pu-238 production rate than the active core. Blanket Pu-238 then has the opportunity to fission using incident neutrons mostly in the epithermal to fast range. Neutron absorption in Pu-238 that does not cause fission instead generates Pu-239. Pu-239 then has the opportunity to fission using incident neutrons from thermal, epithermal and fast.

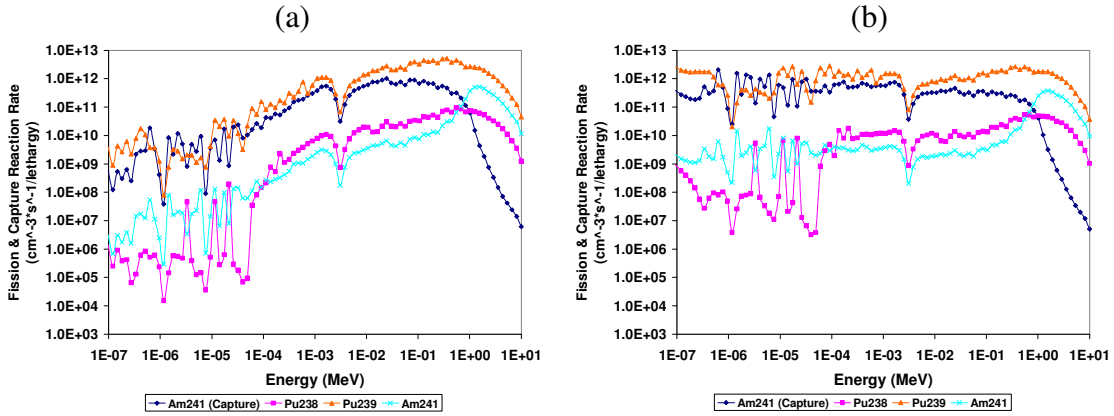

Figure 9: Incident energy reaction rate spectrum for: (a) active driver fuel (b) thermalized axial blanket

Based on fission-to-capture ratio calculations from the spectrums in Figure 9, one Pu-238 atom is produced for every 1.36 $_0$n capture in Am-241. This newly formed Pu-238 atom than fissions for every 3.04 neutron absorptions. This is a smaller neutron investment compared to Light Water Reactor Inert Matrix Fuel; which is 5.47 absorptions per fission. This reflects the bombardment of the targets by fast neutrons as well as thermal. An additional capture in Pu-238 produces Pu-239 which has a fission-to-absorption ratio in the targets of 0.66. The in situ breeding and burning of even plutonium isotopes can be seen in Figure 10.

It is important to note the similar amount of bred plutonium in both blanket designs. The DU blankets bred 1E4 grams of Pu-239 by 500 Effective Full Power Days (EFPD). This is nearly

the same amount of Pu-238 produced by 500 EFPD in the minor actinide target. Given a fuel irradiation time of 400-500 EFPD, the Am-241 in the targets may be decreased by roughly 50%. Higher americium destruction may be obtained if the irradiation time may be extended. Increasing the transuranic and/or heavy metal loading in the active core could increase the irradiation time by breeding replacement Pu-239. Increased breeding reduces the reactor's overall transuranic destruction. However, irradiating the targets to 1000 EFPD and beyond would maximize minor actinide destruction. Therefore, even though the reactor's total TRU destruction efficiency is reduced, the percent MA in the TRU mass is minimized.

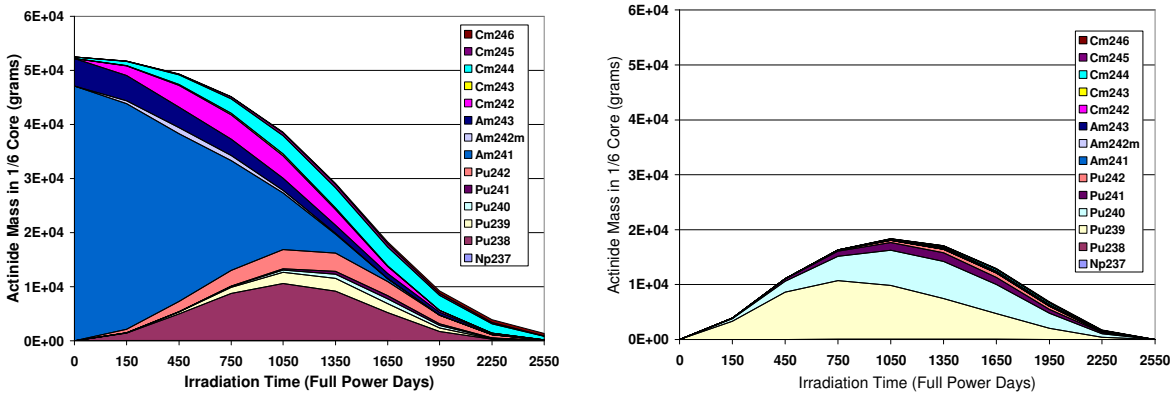

Figure 10: Am-241 destruction and even plutonium destruction in the thermalized axial blanket verses odd plutonium production in the depleted uranium non-thermalized blanket.

The MA reduction has a fuel handling advantage because the gamma and neutron dose per mass of TRU in fuel fabrication is linked to americium and curium isotopes heavier than Cm-244. It also has a repository benefit because the small amount of mass lost during reprocessing which is sent to the repository has a smaller Am-241 concentration. Am-241 is the primary heat load contributer for the first 1000 years in the repository. Because of its decay heat, Am-241 is the driving factor in the drift tunnel spacing. Reducing the americium concentration allows the drifts to be spaced closer together which increases the available storage space in the repository.

6. CONCLUSIONS

Transmutation scenarios involving MA axial targets exhibit promising reactivity suppression and feedback aspects by allowing for fast flux neutron streaming from active core regions. Simultaneously, neutrons are invested in blanket Am-241 which creates reactivity later in fuel life. This is essentially what U-238 does in both fast and thermal reactors. The use of a moderating material in a fast reactor infers that the driver fuel and thermalizing medium be spatially separated and neutronically decoupled using a strong neutron absorber. This prevents thermal neutrons from causing fissions in fissile driver fuel atoms, thus avoiding power peaking in the fast reactor system.

The current core design lacked the available reactivity to achieve a cycle length long enough to adequately destroy the targeted MA. This was due to too small of a heavy metal loading (both fissile and fertile) in the driver fuel. Future investigations will evaluate optimizing the cycle length to adequately destroy the targeted MA.

Also, the use of technetium is shown as a promising absorber material supplement for U-238. This is because the spatial self-shielding that Tc-99 targets provide prevents adjacent driver fuel assemblies from communicating with each other in the un-resolved resonance region. A high atom density and greater un-resolved resonance cross sections than U-238 allows absorbing more neutrons at higher energies. This characteristic reduces the spectrum hardening affect on MA multiplication as a result of a loss in sodium.

ACKNOWLEDGMENTS

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