## Minor Actinide Recycle in Sodium Cooled Fast Reactors Using Heterogeneous Targets

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#### **ABSTRACT**

This paper investigates the plausible design of transmutation target assemblies for minor actinides (MA) in Sodium Fast Reactors (SFR). A heterogeneous recycling strategy is investigated, whereby after each reactor pass, un-burned MAs from the targets are blended with MAs produced by the driver fuel and additional MAs from Spent Nuclear Fuel (SNF). A design iteration methodology was adopted for customizing the core design, target assembly design and matrix composition design. The overall design was constrained against allowable peak or maximum in-core performances. While respecting these criteria, the overall design was adjusted to reduce the total number of assemblies fabricated per refueling cycle. It was found that an inert metal-hydride MA-Zr-H<sub>x</sub> target matrix gave the highest transmutation efficiency, thus allowing for the least number of targets to be fabricated per reactor cycle.

#### 1. INTRODUCTION

There has been recent interest in burning the transuranic (TRU) waste component of Light Water Reactor (LWR) Spent Nuclear Fuel (SNF) in sodium cooled fast reactors (SFR). In addition to fissile plutonium, this would include the minor actinides (MA): neptunium, americium, curium, berkelium and californium. The associated gamma and neutron radioactivity, as well as heating, associated with decay of these MAs (except for neptunium) may significantly complicate fuel handling and fabrication if these actinides are distributed among all SFR fuel assemblies.

The TRU mass could be partitioned from the transuranic waste stream during reprocessing and discarded to a geologic repository after every fuel recycle. However, the rate of MA generation within the SFR core due to non fission transmutation of reactor grade plutonium is significant. This is particularly true of curium and higher mass isotopes<sup>1,2</sup>. Therefore, continuous expulsion of MA waste from the fuel cycle causes the SFR to become a net producer of additional MAs in addition to that produced by the

LWRs. As an alternative option, MAs could be partitioned from the transuranic waste stream and loaded in specialized irradiation vehicles (i.e., transmutation target assemblies). The objective of this study is to investigate a specialized target assembly design for irradiation of MAs in the SFR core separate from the driver fuel assemblies. The aim of these target assemblies is to reduce the volume of assemblies requiring more rigorous handling or a more complicated fabrication process.

The term "minor actinide" is a misnomer when one considers the waste impact of this material on the nuclear fuel cycle. It is well understood that many of the design issues associated with disposal of SNF are linked to the MAs<sup>3</sup>. Furthermore, several of the MA isotopes contribute to high specific alpha, photon and/or neutron emission when compared to plutonium isotopes. These heat (alpha, gamma) and radiation sources (gamma, neutron) can be constraining factors for fabricating MA-containing fuel<sup>4,5</sup>. If the SFR's fresh fuel driver assemblies all contain MA (i.e., homogeneous recycling) then the entire SFR fuel fabrication infrastructure would need to be designed to tolerate these heat loads and radiation fields to some degree. The transmutation target approach tailors a small part of the overall fuel fabrication infrastructure for the special handling requirements of the MAs (i.e., heterogeneous recycling). Most of the isotopes of americium, curium, berkelium and californium have considerably higher alpha, gamma and neutron emission rates than neptunium and plutonium. Therefore, these elements are selected for partitioning and transmutation in the target assemblies.

This paper summarizes a conceptual design study of transmutation target assembly deployment scenarios (i.e., core loading placement, recycling frequency, etc.) in SFRs. These designs span fertile uranium matrices, inert un-moderated matrices, and moderated hydride matrices for dispersing MAs within all pins of the fuel assembly design. Also specialized pin lattices incorporating pins of hydride materials and pins carrying MAs in a non hydride inert matrix were investigated. These assembly designs were matched with a heterogeneous core geometry (i.e., number of targets) and core residency time (i.e., cycles of target irradiation) that maximized transmutation efficiency while meeting the in-core performance constraints as closely as possible. For example, a core design with 18 targets and 10 cycles of irradiation will require 1.8 targets charged to the core per cycle, on average. Whereas, a 48 target core with an 18 cycle irradiation will require 2.7 targets charged per cycle, on average. A core and target design combination that allows for a small number of targets needing to be fabricated per cycle is considered attractive.

#### 2. HOMOGENEOUS VERSUS HETEROGENEOUS RECYCLE

These studies were performed using the Advanced Recycling Reactor (ARR) as the reference core and assembly design<sup>6</sup>. The same assembly design used by the driver fuel was adopted for the transmutation target assemblies. In the reference homogeneous core, all transuranics are recovered and distributed among the driver fuel assemblies, whereas in the heterogeneous cases, Np+Pu was recovered and returned to driver fuel while Am+Cm+Bk+Cf was recovered and put into the target assemblies. In both cases, an external supply of transuranics having an LWR SNF isotopic vector was added to the recovered material after each cycle to compensate for transuranics destroyed by fission. This external TRU supply corresponds to spent PWR fuel irradiated to 50 MWD/kg,

cooled for five years before reprocessing and stored for two years following separation prior to irradiation. The isotopic composition of the external TRU, Np+Pu and Am+Cm+Bk+Cf supply is given in **Table 1**.

In order to give a fair comparison between the heterogeneous target cases and the homogeneous reference case, the ratio of Am+Cm+Bk+Cf to Np+Pu in the external supply of transuranics was held constant and equal to the actual ratio found in LWR SNF. Because this ratio was always respected in conjunction with a constant thermal power rating, an equal comparison could be made between the reference homogeneous core and the heterogeneous designs. Target assemblies contain some Np+Pu due to decay of MAs after fabrication.

Homogeneous	Heterogeneous	Heterogeneous
Reference Driver	Case Driver Case Targe	
	Assemblies	Assemblies
5.5393E-02	5.8750E-02	1.7550E-03
2.5048E-02	2.6610E-02	2.5530E-05
4.6129E-01	4.9010E-01	1.0230E-04
2.2642E-01	2.4010E-01	7.8880E-03
9.3565E-02	9.9420E-02	9.9060E-07
7.0039E-02	7.4420E-02	3.1200E-06
4.2200E-02	1.0060E-02	5.5570E-01
9.9716E-05	0.0000E+00	1.6940E-03
1.9067E-02	0.0000E+00	3.2380E-01
2.9415E-07	0.0000E+00	4.9960E-06
5.1334E-05	0.0000E+00	8.7190E-04
5.9322E-03	0.0000E+00	1.0080E-01
3.8126E-04	0.0000E+00	6.4750E-03
4.9266E-05	0.0000E+00	8.3670E-04
	Reference Driver  5.5393E-02 2.5048E-02 4.6129E-01 2.2642E-01 9.3565E-02 7.0039E-02 4.2200E-02 9.9716E-05 1.9067E-02 2.9415E-07 5.1334E-05 5.9322E-03 3.8126E-04	Reference Driver         Case Driver Assemblies           5.5393E-02         5.8750E-02           2.5048E-02         2.6610E-02           4.6129E-01         4.9010E-01           2.2642E-01         2.4010E-01           9.3565E-02         9.9420E-02           7.0039E-02         7.4420E-02           4.2200E-02         1.0060E-02           9.9716E-05         0.0000E+00           1.9067E-02         0.0000E+00           2.9415E-07         0.0000E+00           5.1334E-05         0.0000E+00           5.9322E-03         0.0000E+00           3.8126E-04         0.0000E+00

 Table 1
 Isotopic Composition of External TRU Feeds.

#### 3. DESIGN ITERATION AND METHODOLOGY

A series of computer codes and analysis tools were utilized in order to determine the transmutation and irradiation performances of the various target designs. **Fig. 1**, describes the general outline for the design iteration between the core loading pattern, target pin-lattice and target matrix selection. The fundamental objective was to minimize the number of target fabrication per cycle while constraining in-core irradiation performances. These in-core performances were evaluated by using bounding fuel and material irradiation metrics based on values found in literature.

- 1. All driver and target assemblies in the core were held to as close to but restricted to be below a fast fluence limit of  $4\times10^{23}$  cm<sup>-2</sup> (E>0.1 MeV), which corresponds to the irradiation limit of the SFR grade ferritic/martensitic steel, HT-9<sup>7</sup>
- 2. The plenum pressure in the driver and target pins was not allowed to exceed 100 atm by the time they were withdrawn from the core at End-of-Life (EOL)<sup>8</sup>

- 3. The temperature of the inner cladding wall was not allowed to exceed 650°C to prevent or minimize the possibility of melting at the fuel to cladding interface due to eutectic phase formation<sup>9</sup>
- 4. All assemblies were limited to pellet centerline temperatures equal to or less than those of the homogeneous reference ARR. 10

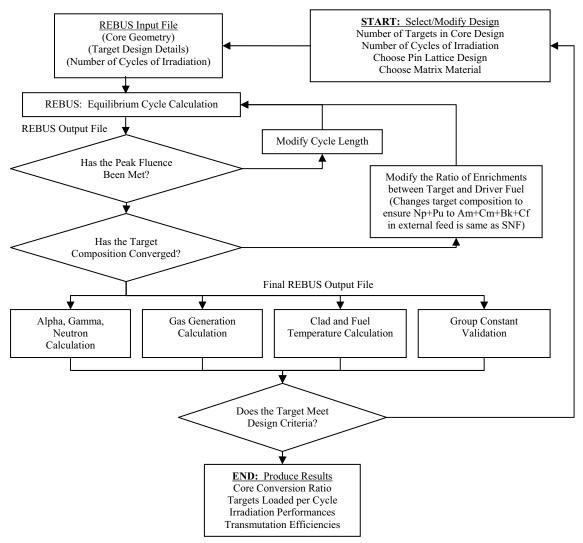


Fig. 1 Design and Analysis Methodology for Core Pattern, Pin Lattice and Pellet Matrix Iteration

#### 3.1 Description of Reactor Physics Tools

The fast reactor codes MC<sup>2</sup>-2, DIF3D and REBUS were used for the reactor physics and fuel cycle calculations<sup>11,12,13</sup>. The MC<sup>2</sup>-2 code was used to generate a 33 group cross section set for each driver fuel enrichment zone, the targets, reflectors and shields. Starting with an ultra fine group ENDF-V/B cross section library, MC<sup>2</sup>-2 creates a collapsed cross section set by performing a zero dimensional critical buckling search using the extended P1 method<sup>11</sup>. Using this collapsed cross section set, the DIF3D

diffusion code was used to solve the multi-group steady state neutron diffusion equation using a hexagonal-z nodal coordinate system<sup>12</sup>. In the nodal discretization, each hexagonal node in the lateral direction represents an assembly. REBUS uses DIF3D to perform a criticality search for the uncontrolled excess reactivity at each time step in its fuel depletion algorithm<sup>13</sup>. In this search, the fresh fuel transuranic enrichment is adjusted until the specified cycle length is achieved. For each enrichment adjustment, the fluxes from DIF3D are used to carry out the isotopic buildup/depletion process over the time of the irradiation cycle. REBUS also performs the in-core fuel management and out of core cooling, reprocessing and re fabricating calculations for each reactor cycle. These operations are carried out until the BOEC excess reactivity is found for the prescribed cycle length at equilibrium.

#### 3.2 Heterogeneous Core Design

This core is modified from the homogeneous-geometry ARR to accommodate a heterogeneous recycling scheme utilizing 18, 21 and 48 MA target assemblies located within the first row of reflector assemblies. **Fig. 2** shows the layout of the 1/3 symmetric core models used in this analysis.

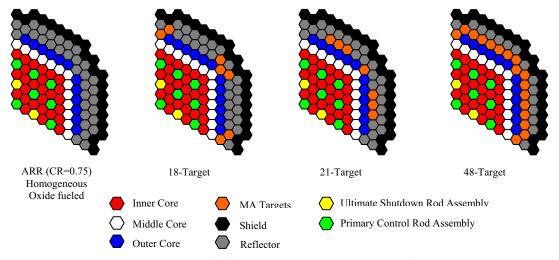


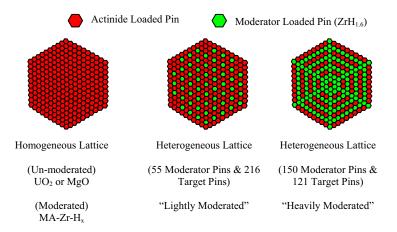
Fig. 2 Radially Heterogeneous ARR Designs

The decision to place the targets in the core periphery was based on the desire to minimize the overall safety implications of MA-bearing assemblies by keeping them out of the high-importance central region of the core. In particular, the concern lies with the threshold-fission characteristic of MAs causing the already positive void coefficient of the active core to be increased with the addition of MAs. The heterogeneous designs mitigate this by placing both un-moderated and moderated targets on the core periphery of the ARR. The high leakage occurring at the core periphery allows for the least amount of above-threshold fission multiplication from affecting the active core.

#### 3.3 Transmutation Target Design

The transmutation target assembly design consists of a 271 pin lattice with identical duct, pitch and pin dimensions to those of the driver fuel assemblies. The main

difference in the target assembly designs investigated is the selection of a homogeneous lattice versus a heterogeneous lattice of target pins. A homogeneous target pin lattice was considered with MA-O<sub>2</sub>/UO<sub>2</sub>, MA-O<sub>2</sub>/MgO or MA-ZrH<sub>x</sub> matrix compositions. The UO<sub>2</sub> and MgO matrix options draw upon a large irradiation experience database from various in-pile tests. Two heterogeneous lattices were investigated, one with 55 moderator pins, the other with 150 moderator pins (**Fig. 3**). These two configurations represent two possible scenarios of introducing increasing amounts of moderation to the target irradiation vehicles. The heterogeneous pin lattice offers the advantage of mechanical separation of the MA-carrying pins from the moderator pins. An alternative approach is to select a matrix material that is hydrogenous. Metal hydride reflectors and matrix materials have been developed for various special applications such as TRIGA reactor fuels.



**Fig. 3** Homogeneous and heterogeneous lattice geometries

#### 3.4 Partitioning Strategy

The separation and recycling strategy investigated in this work assumes the ability to partition uranium, Np+Pu, and MAs (Am+Cm+Bk+Cf) into three separate waste streams. The separation strategy is outlined in **Fig. 4**. The general philosophy of maintaining the MA inventory in transmutation targets is indicated by the hot cell and glove box images at the center of the figure. In each recycle, the Np+Pu produced by the targets is separated from the MAs and recycled into the next batch of driver fuel. The driver external makeup feed is comprised of LWR SNF Np+Pu and recovered uranium (uranium recovered from SNF). Also in each recycle, the Am+Cm+Bk+Cf produced by the driver fuel is separated from the Np+Pu and recycled into the next batch of targets. The transmutation target external makeup feed of MAs has the same isotopic vector as the Am+Cm+Bk+Cf corresponding to LWR SNF.

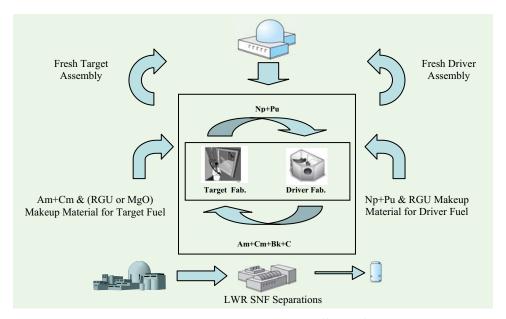


Fig. 4 Heterogeneous recycling scheme

#### 4. PERFORMANCE COMPARISON

The transmutation, fabrication, mass flow and irradiation performances of the targets are symbiotically related. In order to assess the general transmutation and in-core performances of each target matrix composition, a set of scoping calculations were performed. These scoping calculations examined transmutation and in-core performances for the 18-target and 48-target core geometries. For each matrix combination and core geometry combination, the number of target irradiations was held constant at 10 cycles. This is the approximate number of cycles to achieve the  $4\times10^{23}$  cm<sup>-2</sup> (E>0.1 MeV) fluence limit in the fast spectrum targets given the irradiation environment in the SFR. The softer neutron spectrum offered by the moderated targets can potentially extend the requisite in-core residency. This option is further explored in a later section. **Table 2** provides the target pellet makeup and MA enrichment values, defined as volume of MA phase per pellet volume.

<b>Table 2</b> Fuel pellet volumetric enrichment (MA Phase Vol./Pellet V	√ol.	.)
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18-Target Cases		48-Target Cases		
UO <sub>2</sub> -18,10	65	UO <sub>2</sub> -48,10	8	
MgO-18,10	31	MgO-48,10	8	
ZrH <sub>x</sub> (55/271)-18,10	25	ZrH <sub>x</sub> (55/271)-48,10	7	
$ZrH_x(150/271)-18,10$	37	$ZrH_x(150/271)-48,10$	13	
ZrH <sub>1.6</sub> -18,10	14	ZrH <sub>1.6</sub> -48,10	5	
ZrH <sub>2.7</sub> -18,10	14	ZrH <sub>2.7</sub> -48,10	6	
Optimized: Number of Targets/Number of Irradiation Cycles				
ZrH <sub>1.6</sub> -21,10	11	ZrH <sub>1.6</sub> -48,18	8	

<sup>\*</sup>case label format: "Target Matrix"(NP/271)-NA,NC

NP=Number of Target Pins, NA=Number of Target Assemblies, NC=Number of Cycles

#### 4.1 Transmutation Efficiency

Since the mass of MAs charged to the target assembly at Beginning of Life (BOL) is not the same in every design, it is useful to compare the MA depletion without units of initial mass. Instead, the depletion trend for each target design is normalized in terms of the MA destruction efficiency. The MA destruction efficiency ( $E_{MA}$ ) is defined as the fraction of the initial MA mass that is removed from the fuel cycle during the irradiation time by either a fission or a capture reaction. This same equation can be applied to the TRU destruction efficiency ( $E_{TRU}$ ).

$$1 - E_{MA} = \frac{M_f^{MA}}{M_i^{MA}} \quad \Rightarrow \quad E_{MA} = \frac{M_i^{MA} - M_f^{MA}}{M_i}$$

The choice of fertile versus inert matrix makes a significant difference on the net transuranic destruction capability of the target design. As can be seen from **Fig. 5**, if the transmutation efficiency for transuranics is plotted for various target designs, the targets can actually be a net producer of transuranics even though they are a net destroyer of MA. It is also important to note the increase in transmutation efficiency with moderated as opposed to non-moderated targets. Both  $UO_2$  and MgO target designs have  $E_{MA}$  less than any of the cases involving zirconium hydride.

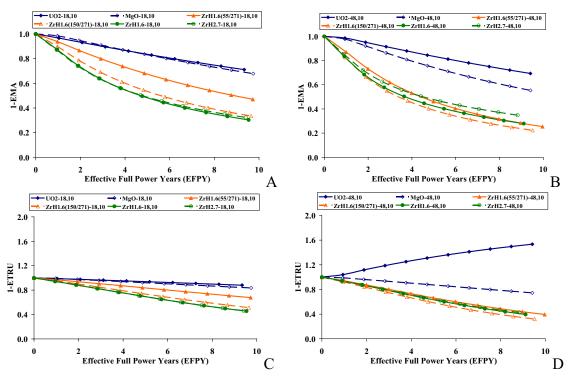


Fig. 5 Depletion behavior normalized to MA or TRU transmutation efficiency: A)  $E_{MA}$  for 18-targets, B)  $E_{MA}$  for 48-targets, C)  $E_{TRU}$  for 18-target, D)  $E_{TRU}$  for 48-targets. 10-cycle irradiation (no fluence optimization).

#### 4.2 Fuel Fabrication Attributes

Curium, particularly Cm-242 and Cm-244, is transmuted by neutron capture in the americium isotopes Am-241 and Am-243, respectively. Cm-242 is short-lived with a 163 day half-life for alpha decay ending in Pu-238. Cm-244 is longer-lived with an 18.1 year half life ending in Pu-240. Because of its longevity compared to its life in a reactor, Cm-244 can be transmuted by further neutron capture while in the core to higher mass actinides, ultimately building up Berkelium and Californium isotopes. It is the curium and higher mass actinides that yield significant decay heat, gamma ray and neutron emission rates. **Fig. 6.A** shows the mass specific heat and neutron emission rates. However, from a fuel handling perspective the heat and neutron emission rates per volume of pellet, shown in **Fig. 6.B** give a more representative measure of the radiation fields encountered.

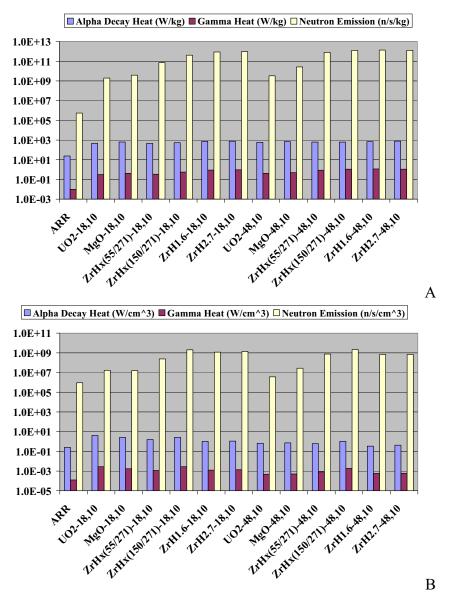


Fig. 6 Heat and dose source strengths: A) by heavy metal, B) by pellet volume

The targets' MA enrichment is limited by the available target pellet volume dictated by core design (i.e, target pellet volume/batch) and also the amount of MA mass being charged to a batch of targets. This mass of MA material is externally supplied from LWR SNF as well as internally supplied by recycled SFR driver fuel and targets. The target volume is affected by the target assembly and size of the target region within the core which is ultimately determined by the reactor designer. The amount of MA externally supplied by the LWR SNF recycler has been fixed to the externally supplied Np+Pu required in the driver fuel to achieve a desired cycle length. This quantity is relatively invariant on the target transmutation performance so long as the targets do not produce plutonium in quantities high enough to significantly raise the core conversion ratio. However, the amount of internally supplied MAs from driver fuel and target recycle is directly determined by the transmutation efficiency.

**Table 3** shows the rate of MA feed supplied to the targets from internal versus external recycling sources. As can be seen from the table, adoption of heterogeneous recycling can actually increase the MA throughput of the SFR recycling infrastructure over that of homogeneous recycling in an ARR. This is undesirable because at best the heterogeneous recycle approach is intended to decrease the MA inventory. It should be noted that the moderated target designs did decrease the ratio of MA throughput from SFR recycle relative to the external feed from SNF, thus indicating that moderated inert targets can decrease the MA inventory of the SFR's closed fuel cycle.

Table 3	MA throughput from both LWR SNF and SFR recycled mass flows.

	MA Feed from	MA Feed from	MA Feed from
Case*	Driver Recycle	Target Recycle	SNF Recycle
	(kg/MWY)	(kg/MWY)	(kg/MWY)
ARR	54.1	n/a	9.2
UO <sub>2</sub> -18,10	19.0	87.4	21.2
MgO-18,10	15.6	46.1	5.5
ZrH <sub>x</sub> (55/271)-18,10	16.7	21.1	5.6
$ZrH_x(150/271)-18,10$	16.7	12.4	5.9
ZrH <sub>1.6</sub> -18,10	17.0	11.0	6.1
ZrH <sub>2.7</sub> -18,10	17.3	12.2	6.2
UO <sub>2</sub> -48,10	11.6	32.7	2.3
MgO-48,10	15.0	26.5	4.9
$ZrH_x(55/271)-48,10$	16.8	8.4	5.9
ZrH <sub>x</sub> (150/271)-48,10	18.0	7.7	6.6
ZrH <sub>1.6</sub> -48,10	18.9	11.0	7.0
ZrH <sub>2.7</sub> -48,10	19.3	15.4	7.1

<sup>\*</sup>Core conversion ratio was allowed to fluctuate depending on target TRU destruction

#### 4.3 In-Core Performance

From **Table 4**, it can be seen that for all target designs considered, the peak linear heat generation rate (LHGR) and temperatures in the targets were calculated to be well below that of the driver fuel, as can be noted by observing the peak temperatures of the homogeneous ARR case. This can be explained by the: 1) placement of targets in the

core periphery where the flux is lowest 2) the MA enrichment, and corresponding power density, had to be reduced to ensure the gas production fell within the EOL plenum pressure criterion.

 Table 4
 Calculated peak thermal performances.

Case	Peak LHGR (kW/m)	Peak Centerline Temp. (°C)	Peak Pellet/Clad Temp. (°C)
ARR	26.9	2025	819
UO <sub>2</sub> -18,10	10.8	637	427
MgO-18,10	5.8	512	395
ZrH <sub>x</sub> (55/271)-18,10	8.8	592	415
$ZrH_x(150/271)-18,10$	19.7	949	491
ZrH <sub>1.6</sub> -18,10	10.2	615	427
ZrH <sub>2.7</sub> -18,10	11.2	640	436
UO2-48,10	6.0	827	648
MgO-48,10	2.3	416	371
$ZrH_x(55/271)-48,10$	4.8	477	377
$ZrH_x(150/271)-48,10$	11.6	664	437
ZrH <sub>1.6</sub> -48,10	6.1	511	400
ZrH <sub>2.7</sub> -48,10	6.4	519	402

From **Table 5**, it can be seen that in general, the un-moderated target designs fall near or just over the fast fluence limit imposed for the target structural materials. However, for the moderated targets the peak fluence after ten cycles of irradiation is much less than the  $4\times10^{23}$  cm<sup>-2</sup> (E>0.1 MeV) fluence limit imposed on the target assembly design envelope.

**Table 5** Calculated gas pressure and fast fluence performances.

Case	MA Enrichment (v/o)	EOL Gas Plenum Pres. (atm.)	Peak Fast Fluence (10 <sup>23</sup> cm <sup>-2</sup> )
ARR	n/a	60	3.99
UO <sub>2</sub> -18,10	65	147	4.12
MgO-18,10	31	103	4.06
ZrH <sub>x</sub> (55/271)-18,10	25	131	3.71
$ZrH_x(150/271)-18,10$	37	242	3.21
ZrH <sub>1.6</sub> -18,10	14	112	2.79
ZrH <sub>2.7</sub> -18,10	14	117	2.48
UO2-48,10	8	38	3.66
MgO-48,10	8	35	3.66
$ZrH_x(55/271)-48,10$	7	51	3.39
ZrH <sub>x</sub> (150/271)-48,10	13	99	2.66
ZrH <sub>1.6</sub> -48,10	5	47	2.17
ZrH <sub>2.7</sub> -48,10	6	49	1.92

Also, it can be seen that most of the 18-target designs fall within 30 atm of the target design pressure limit of 100 atm. The exception to this rule is the "heavily moderated" heterogeneous cases where the pressure is calculated to by 2.5 times the constraint. This is attributed to the fact that in these cases, the number of pins containing MA has been reduced and with it the plenum space available. Also, the pressure in the 18-target UO<sub>2</sub> case is nearly 50 atm greater than the 100 atm constraint. This is because the target pellet in this design is 65% MA thus lending to a larger alpha decay and fission density in the target pellet. Of the 18-target cases, the MgO matrix case most closely meets the limiting pressure and fluence criteria without going over. This case (18 targets, 10 cycle irradiation) gives the least number of targets needing fabrication per batch while still meeting the in-core performance criteria. Though this design minimizes the number of assemblies requiring fabrication per cycle, the MA throughput during recycle is high compared to the moderated target cases (Table 3). All of the 48-target cases were well under the pressure constraint. This is because the MAs are distributed over a larger number of assemblies giving much more available plenum space while at the same time reducing the target power density.

#### 5. FINAL DESIGN MORPHOLOGY

Of all of the scoping calculation designs, the metal hydride matrix cases had the highest transmutation efficiency (**Fig. 5**). Because the moderation was provided without displacing any target pins containing MA, the plenum space above every pin in the assembly was available for gas storage, thus the pressures were not excessive. Also, the softer neutron spectrum allowed all of the moderated target designs to be below the fast fluence limit by a large margin. From **Fig. 5** it is apparent that a high hydrogen stoichiometry does not greatly improve transmutation efficiency. Therefore, a zirconium hydride stoichiometry of 1.6 hydrogen atoms per one zirconium atoms was selected. It should be noted that pure ZrH<sub>1.6</sub> exists as a delta phase which retains composition for temperatures up to  $1000^{\circ}$ C. This precludes hydride formation with the metallic MAs present in the composition.

From **Table 5** it can be seen that the 18-target  $ZrH_{1.6}$  matrix overshot the EOL pressure criteria while the 48-target  $ZrH_{1.6}$  matrix grossly undershot the fluence criteria. A compromise design was found that gives an EOL plenum pressure of 99 atm which is just under the plenum pressure constraint. To do this, more targets were added to the core design (21-targets) while maintaining the same number of irradiation cycles (10 cycles). This had the effect of decreasing fission density while increasing the available plenum space to the targets. This design still avoided reaching the fluence limit easily.

Because of the softer spectrum brought about by the local moderation, the target assemblies could be irradiated for much longer than 10 cycles. The number of cycles had to be increased to 18 in order to irradiate the targets to the fluence limit. This is more realistic than the 10 cycle scenario because the fluence limit is intended to provide the reasonable metallurgical limit of the cladding and structural materials. Increasing the number of irradiation cycles also effectively increases the number of batches in the target region. Therefore, the number of total targets in the core design had to be increased to allow space in the core for these added batches. Similar to the increase from 18 to 21

targets, the increase to 48-targets is necessary from the standpoint of ensuring sufficient plenum space to hold the helium and fission gas produced by the high burnup of the long irradiation.

The  $ZrH_{1.6}$ -21,10 case provides an enrichment of 11% based on only the plenum pressure criteria. The  $ZrH_{1.6}$ -48,18 case provides a lower enrichment limit of 8%. The long irradiation time of the  $ZrH_{1.6}$ -48,18 case produces the smallest recycled target mass stream. However, the  $ZrH_{1.6}$ -21,10 case has the lowest number of targets needing fabricated per batch at 2.1 assembly per batch, as opposed to 2.7 targets per batch for the  $ZrH_{1.6}$ -48,18 case. An overview of these two optimized cases is provided in **Table 6**.

**Table 6** Overview of metal hydride targets with optimized in-core residency and batch fraction to be within gas pressure and fast fluence limits

batch fraction to be within gas pressure and fast fluence limits					
		ZrH <sub>1.6</sub> -21,10		ZrH <sub>1.6</sub> -48,18	
Conversion Ratio	0	.66	0.64		
Cycle Length (Days)	343		334		
Number of Targets in Core		21	4	8	
Number of Cycles of Target Irradiation		10	1	8	
Avg. Number of Targets per Batch	2	2.1	2.	.7	
MA Transmutation Efficiency: $E_{MA}$	,	76	8	4	
TRU Transmutation Efficiency: E <sub>TRU</sub>	(	66	7	9	
Heat and Neutron Emission at Disc	charge (b	y heavy m	etal mass)		
Alpha Decay Heat (W/kg)	1	17	6	9	
Gamma Energy (W/kg)	0.19		0.11		
Neutron Emission (n/s/kg)	2E11 1E11		11		
Heat and Neutron Emission at Da	ischarge	(by pellet	volume)		
Alpha Decay Heat (W/cm^3) 0.15 0.07				07	
Gamma Energy (W/cm^3)	2.5E-04 1.1E-4		E <b>-4</b>		
Neutron Emission (n/s/cm^3)	3E8 1		1E	E8	
Feed Stream A	Analysis				
MA Feed from Driver Recycle (kg/MWY)	0.0182		0.0	0.0187	
MA Feed from Target Recycle (kg/MWY)	0.0086		0.0053		
MA Feed from SNF Recycle (kg/MWY)	0.0066		0.00	070	
In-Core Performance					
	Driver	Target	Driver	Target	
Peak Fuel Centerline Temp (°C)	2142	640	2208	521	
Peak Inner Cladding Temp (°C)	435	430	556	398	
Peak LHGR (kW/m)	27	12	28	7	
Peak Fluence (10 <sup>23</sup> cm <sup>-2</sup> )	3.88	2.48	3.89	3.92	
Peak EOL Pressure (atm)	53	99	52	90	

#### 6. SUMMARY OF OPTION SPACE

This work investigated a large array of core design, lattice and matrix options. The use of fertile targets was found to have approximately the same MA transmutation efficiency as non-fertile targets but reduced the net transuranic burning performance of

the core design due to plutonium production from capture in U-238. This was true even for a small number of high-MA enrichment targets such as in the 18-target case. Nonfertile matrix targets do not have this disadvantage. Both fertile and non-fertile matrices are limited by a fast fluence limit dictated by the maximum acceptable cladding and structural material damage. Moderated targets decrease the local fast flux in the target region, thus allowing for longer irradiation times enabling higher burnup. There is also a spectral advantage to moderated targets in that generally, neutron cross-section increases with decreasing energy.

Moderation decreases neutron energies, increasing the reaction rates experienced in the targets. In particular, the neutron capture cross-sections of the long-lived MAs, Am-241, 243 and Cm-244 are two orders of magnitude greater just below 1 MeV than just above 1 MeV. The inverse is true of the fission cross-sections for these isotopes known as the fission threshold shared by all even-neutron numbered actinides. Reduction of neutron energies to just below 1 MeV or lower, gives an immediate jump in transmutation efficiency as was apparent when comparing fast versus thermal options in **Fig. 5**. Taking full advantage of the longer irradiation times and the higher reaction rates allowed by moderated targets allows for the maximum EOL MA and TRU transmutation efficiencies. A comparison of depletion trends by isotope for fertile, non-fertile, moderated and moderated with extended irradiation times is given in **Fig. 7**.

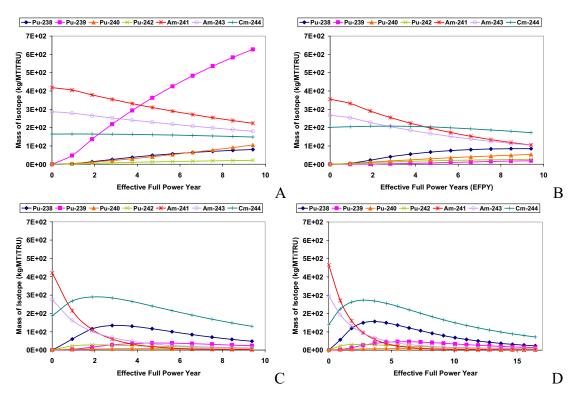


Fig. 7 Depletion curves for: A) 48-targets and 10 cycles with UO<sub>2</sub> matrix, B) 48-targets and 10 cycles with MgO matrix, C) 21-targets and 10 cycles with ZrH<sub>1.6</sub> matrix, D) 48-targets and 18 cycles with ZrH<sub>1.6</sub> matrix

#### 7. CONCLUSIONS

The evaluation of heterogeneous recycle of MA targets has revealed that the use of fertile-free moderated targets, such as actinide alloyed metal hydrides, allows for the largest amount of MA waste destruction. From a continuous recycle standpoint for these targets, the high transmutation efficiency for this matrix concept equates into the smallest amount of MA mass circulating through the fuel cycle. This is the most promising option for decreasing the size of the fuel recycling and manufacture infrastructure for MAs, because the more MA mass that is destroyed during irradiation, the less MA mass that reenters the target recycling infrastructure. A MA-ZrH $_{1.6}$  (8 v/o enrichment) matrix target irradiated for 18 cycles requires a total of 48-targets in the core to accommodate a batch size of 2.7 targets per reload. Of all the metal hydride matrix targets, this design came closest to the maximum pressure and fast fluence criteria.

It should be noted that though metal hydride targets decrease the amount of MA mass circulating through the fuel cycle, this design does not give the smallest number of targets fabricated per reactor cycle. In fact, an MAO<sub>2</sub>-MgO (31 v/o enrichment) with 18 targets and a ten cycle irradiation gives 1.8 targets per reload. Of the un-moderated targets, this design came closest to the maximum pressure and fast fluence criteria. However, because the transmutation efficiency for this design was so low, the MA throughput for reprocessing spent targets is quite high. This high MA reprocessing throughput will likely cause a quicker degradation and/or higher volume of solvent if using aqueous reprocessing. It may also make likely that a situation where high mass inventories of processed MAs waiting for target fabrication to occur at the recycling center. A moderated target design avoids this by storing MA inventories in the reactor for much a much longer number of cycles (~18 EFPY) than in un-moderated targets (~10 EFPY). Also because the MA transmutation efficiency is much higher than for un-moderated targets, less MA recycling throughput is required.

Transmutation targets with moderating matrix are a fairly new concept. Successful synthesis of curium hydride and americium hydride has been reported<sup>14</sup> and there exists a large body of knowledge and experience associated with the U-ZrH<sub>2</sub> TRIGA fuel<sup>15</sup>. These processes draw upon metal fuel casting similar to that performed for metallic SFR fuels with an added hydration step. Because literature confirms synthesis of americium and curium hydrides and due to the promising waste transmutation attributes, metal hydride matrix targets deserve further consideration.

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#### **NOMENCLATURE**

ARR Advanced Recycle Reactor

**BOEC Beginning of Equilibrium Cycle** 

BOL Beginning of Life

E<sub>MA</sub> MA Transmutation Efficiency

**EOEC** End of Equilibrium Cycle

EOL End of Life

E<sub>TRU</sub> Transuranic Transmutation Efficiency

MA Minor Actinide

SFR Sodium cooled Fast Reactor

SNF Spent Nuclear Fuel

TRU Transuranic

#### REFERENCES

- 1. R. FERRER, S. BAYS, M. POPE, Evaluation of Homogeneous Options: Effects of Minor Actinide Exclusion from Single and Double Tier Recycle in Sodium Fast Reactors, INL/EXT-08-14034 (2008).
- L. BUIRON, F. VARAINE, D. LORENZO, H. PALANCHER, B. VALENTIN, "Minor Actinides Transmutation in SFR Depleted Uranium Radial Blanket, Neutronic and Thermal Hydraulic Evaluation," Proc. of Global'07 – Advanced Nuclear Fuel Cycles and Systems, Boise, Idaho, September 9-13, 2007, (2007).
- 3. R. WIGELAND, T. BAUER, "Repository Benefits of AFCI Options", ANL-AFCI-129 (2004).
- 4. M. SALVATORES, C. PRUNIER, Y. GUERIN, A. ZAETTA, I SLESSAREV, "The "SPIN" Program at CEA", *Proc. of Global'93*, Seattle, Washington, September 12-17, 1993, (1993).
- 5. S. PILLON, J. SOMERS, S. GRANDJEAN, J. LACQUEMENT, "Aspects of Fabrication of Curium Based Fuels and Targets," *Journal of Nuclear Materials*, **320**, pp. 36-43, (2003).
- 6. E. HOFFMAN, W. YANG, R. HILL, "Preliminary Core Design Studies for the Advanced Burner Reactor Over a Range of Conversion Ratios," ANL-AFCI-177 (2006).
- 7. D. GELLES, Microstructural Development in Reduced Activation Ferritic Alloys Irradiated to 200 dpa at 420 °C, *Journal of Nuclear Materials*, **212**, pp. 714-719, (1994).
- 8. A. WALTER, A. REYNOLDS, *Fast Breeder Reactors*, Pergamon Press Inc., Maxwell House, Fairview Park, Elmsford, New York, United States, (1981).
- 9. R. PAHL, C. LAHM, S. HAYES, Performance of HT9 Clad Metallic Fuel at High Temperature, *Journal of Nuclear Materials*, **204**, pp. 141-147, (1993).

- 10. J. CARBAJO, G. YODER, S. POPOV, V. IVANOV, A Review of the Thermophysical Properties of MOX and UO2 Fuels, *Journal of Nuclear Materials*, **299**, pp. 181-198, (2001).
- 11. H. HENRYSON II, B. TOPPEL, and C. STENBERG, "MC<sup>2</sup>-2: A Code to Calculate Fast Neutron, Spectra and Multi Group Cross Sections," ANL-8144, (1976).
- 12. K. DERSTINE, DIF3D: "A Code to Solve One, Two, and Three Dimensional Finite Difference Diffusion Theory Problems," ANL-82-64, (1984)
- 13. B. TOPPEL, "A User's Guide to the REBUS-3 Fuel Cycle Analysis Capability," ANL 83 2, (1983)
- 14. W. OLSON AND R. MULFORD, "The Americium Hydrogen System", *The Journal of Physical Chemistry*, **70**, pp. 234-237, (1966)
- 15. M. SIMNAD, "The U-ZrHx Alloy its Properties and Use in TRIGA Fuel," *Nuclear Engineering and Design*, **64**, pp. 403-422, (1981)