THORIUM-BASED TRANSMUTER FUELS FOR USE IN LIGHT WATER REACTORS

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ABSTRACT

Advanced fuels for thermal reactors will be needed in the future for several reasons: reduced production of plutonium and minor actinides (MA, i.e., neptunium, americium, curium, and the higher elements), achieving higher burnups, and a more robust fuel-form/waste-form for temporary storage awaiting reprocessing or for holding the fission products and actinides in a permanent repository. Thorium-based fuels reduce the total amount of plutonium produced and produce a mixture of plutonium isotopes high in ^{238}Pu. Because of the high decay heat and spontaneous neutron generation by ^{238}Pu, this isotope helps to provide a higher measure of intrinsic proliferation resistance. Thoria as a fuel-form and as a waste-form is difficult to dissolve, thus making the diversion of spent fuel for weapons' production purposes more difficult.

This analysis has been conducted to determine if ThO_2-UO_2 fuels can be used to further increase the proliferation resistance of LWR fuels, while reducing the plutonium and MA inventory. Some of the proliferation concern in the world today stems from plutonium that has already been separated from spent fuel. Currently separated plutonium is being incorporated in UO_2-PuO_2 mixed oxide (MOX) fuel. However, because MOX fuel contains ~90 wt % ^{238}U, substantial amounts of ^{239}Pu are produced in the MOX fuel and net plutonium burnup rates are only 30-50% per cycle. The incorporation of plutonium into a ThO_2 matrix will consume already-separated plutonium without breeding additional ^{239}Pu. The MAs would be included in the ThO_2 to further reduce the overall long-term radiotoxicity of the fuel cycle. In the work presented here, ~11% of the fuel pins in a typical PWR are replaced by the thorium-based fuel pins, where the first cycle shows a small net increase of plutonium. Further optimization should show a net decrease in the total core inventory of plutonium with modest loadings of the thorium fuel.

1. INTRODUCTION

Thoria fuels appear promising as a matrix for plutonium and the minor actinides during mono-recycling in light water reactors. The goals of this recycling strategy are to reduce overall inventories of plutonium, to render the resulting spent fuel as proliferation-resistant as possible through the accumulation of ^{238}Pu, ^{240}Pu, ^{242}Pu and ^{232}U, to consume the minor actinides, and to produce a very robust waste form. A schematic of the mono-recycling strategy is shown in Figure 1. In this fuel cycle the LWR fuel assembly consists of 89% standard UO_2 fuel rods with a ^{235}U enrichment of 4.95 wt %. The plutonium and minor actinides produced in earlier standard rods is separated and placed in thoria-urania pins occupying 11% of the positions in the fuel assembly. Thus the goal of the mono-recycling strategy or "twice through fuel cycle" is to transmute the great majority of the long lived actinides in existing LWRs and to discharge a fuel form that is a very robust waste form and whose isotopic content is very proliferation resistant.

In this analysis, we have used a model, shown in Figure 2, consisting of nine fuel pins, eight of which are standard UO_2 pins and one of which is a $[Th\text{-}U\text{-}Pu\text{-}MA]O_2$ transmuter pin. The plutonium and minor actinides are derived from UO_2 fuel irradiated to 45 MW-d/kg and reprocessed 30 years after discharge. In the various cases analyzed, the center pellet contained 0-10 wt% recovered uranium and 6 - 15 wt% Pu+MA. The recovered uranium (98.5 wt% ^{238}U) was included to denature the ^{233}U below the 12 wt% limit for low enrichment uranium (LEU) [1]. For these analyses, the burnup code MOCUP [2], which uses the Monte Carlo transport code MCNP [3] and the exponential matrix generation and depletion code ORIGEN2 [4], was used. MOCUP was used with 60 day time steps and tracked the generation and depletion of 50 fission products and 38 actinides. The model has white reflecting boundaries to simulate an infinite array. The eight outer pins are modeled as eight individual fuel zones and the center pin is divided into twenty equal-volume zones.

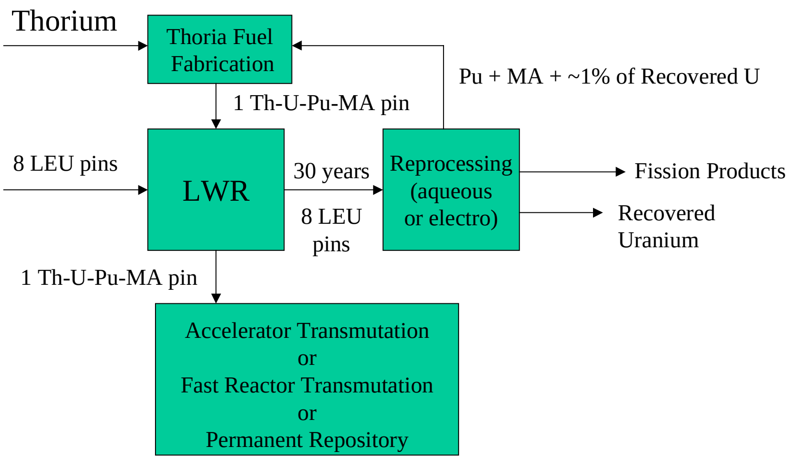
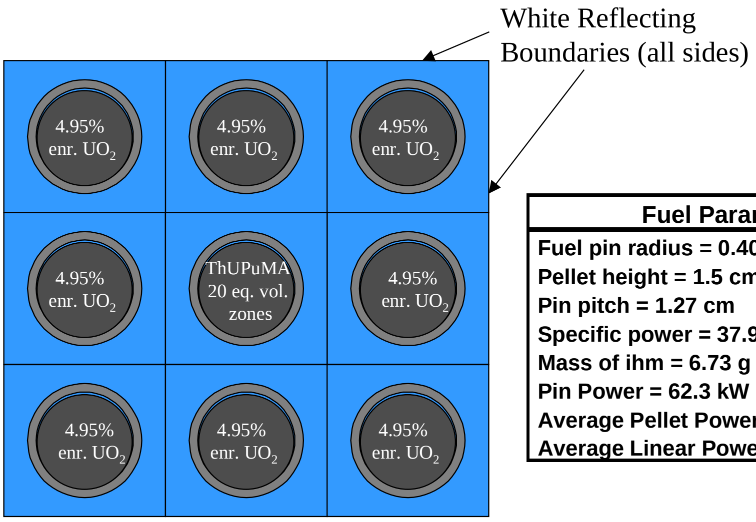


Figure 1 Mono-recycling strategy for Light Water Reactors



Fuel Parameters
Fuel pin radius = 0.4096 cm
Pellet height = 1.5 cm
Pin pitch = 1.27 cm
Specific power = 37.9 W/g ihm
Mass of ihm = 6.73 g
Pin Power = 62.3 kW
Average Pellet Power = 256 W
Average Linear Power = 171 W/cm

Figure 2 Nine-pellet model and fuel parameters

The isotopic constituents of the fresh UO_2 fuel and of the recovered uranium and plutonium plus minor actinides are shown in Table 1.

We analyzed ten cases using the nine-pellet model show above. The constituents of the center pellet were varied to determine the most effective combination for extended burnup, proliferation resistance and consumption of the minor actinides. In some cases the center pellet was irradiated for the entire lifetime of the surrounding UO_2 pellets, then removed from the first assembly and placed in a second, fresh UO_2 assembly. The isotopic concentrations in the center pellet were tracked both spatially and temporally through the entire irradiation. In addition, isotopic ratios that are important to proliferation resistance have been tracked both spatially and temporally for each of the ten cases.

2. ISOTOPIC INVENTORY

The variation of the ^{239}Pu concentration in the center pin is shown in Figure 3. Note that the ^{239}Pu, initially uniform across the diameter, is burned out on the periphery first because of the well-moderated flux near the coolant. After a burnup of 52.4 MW-d/kg (red curve), the center concentration is about 25% of the initial concentration and the peripheral concentration is about 10% of the BOL concentration. 52.4 MW-d/kg is the reactivity-limited burnup using 4.95% enrichment in the eight outer pins. Further irradiation of the center pin in a fresh assembly reduces the ^{239}Pu content, but only marginally. The concentrations at 52.4 MW-d/kg are shown twice in Figure 3, at the end of the first LEU cycle and at the beginning of the second.

Table 1 Isotopic Input

	Fresh Fuel	Recovered U
U-234	0.00%	0.027%
U-235	4.95%	0.908%
U-236	0.00%	0.578%
U-237	0.00%	0
U-238	95.05%	98.487%

The uranium is included in the center pin only as a diluent for the ^{233}U to approach the LEU limit, 12% ^{233}U/total U.)

45 MWd/kg ihm UO2 fuel, 30 years after discharge		
	Fraction of Pu+MA	**Elemental Fraction**
Np-236	0.00%	0.00%
Np-237	6.03%	100.00%
Np-238	0.00%	0.00%
Pu-237	0.00%	0.00%
Pu-238	1.77%	2.16%
Pu-239	49.00%	59.99%
Pu-240	21.71%	26.59%
Pu-241	3.29%	4.03%
Pu-242	5.90%	7.23%
Pu-243	0.00%	0.00%
Am-241	10.79%	89.06%
Am-242m	0.01%	0.07%
Am-243	1.32%	10.87%
Cm-242	0.00%	0.01%
Cm-243	0.00%	1.75%
Cm-244	0.15%	84.74%
Cm-245	0.02%	11.89%
Cm-246	0.00%	1.59%
Cm-247	0.00%	0.02%
Cm-248	0.00%	0.00%

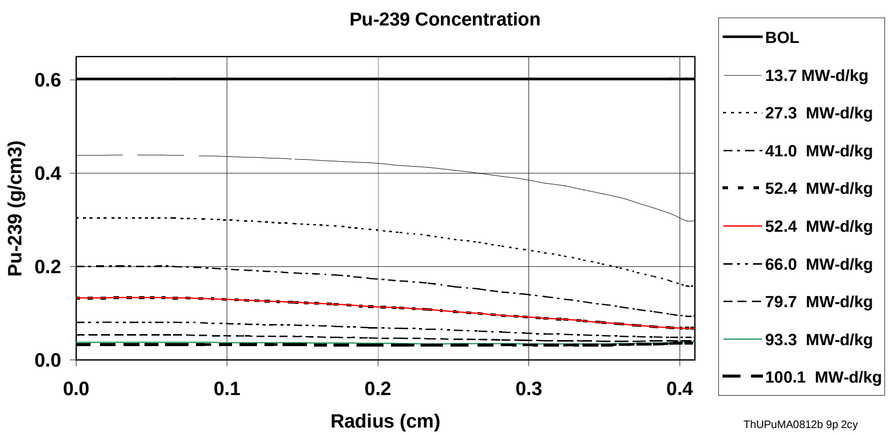


Figure 3 Pu-239 Concentration in center pin during irradiation

3. PROLIFERATIONS RESISTANCE

The various isotopes of plutonium are compared in Table 2. The high spontaneous neutron generation and high decay heat of ^{238}Pu would make any plutonium separated from the center pin mixture very difficult to use for weapons purposes.

Table 2 Properties of dominant plutonium and americium isotopes.

Isotope	Halflife	Bare Crit	Spontaneous Fission Neutrons	Decay Heat
	years	kg, α–phase	neutrons/gm-s	Watts/kg
Pu-238	87.7	10	2600	560
Pu-239	24,100	10	0.022	1.9
Pu-240	6,560	40	910	6.8
Pu-241	14.4	10	0.049	4.2
Pu-242	376,000	100	1700	0.1
Am-241	430	100	1.2	114

An example of the inventory variations of the ^{233}U/total U, ^{238}Pu/total Pu and ^{239}Pu/total Pu for the center pellet, which at BOL contained 10 wt% recovered uranium and 15 wt% Pu+MA, is shown in Figure 4. Note that the ^{239}Pu as a fraction of total plutonium decreased from 70% to 20%. At the same time, the ^{238}Pu fraction of the total plutonium increased from about 2% to 20%. Thus, any plutonium extracted from the discharged fuel would have high decay heat and spontaneous neutron production, and thus would not be attractive for use in weapons. In addition, ^{233}U as a fraction of the total uranium content remains below 12%, thus making the uranium very resistant to weapons use.

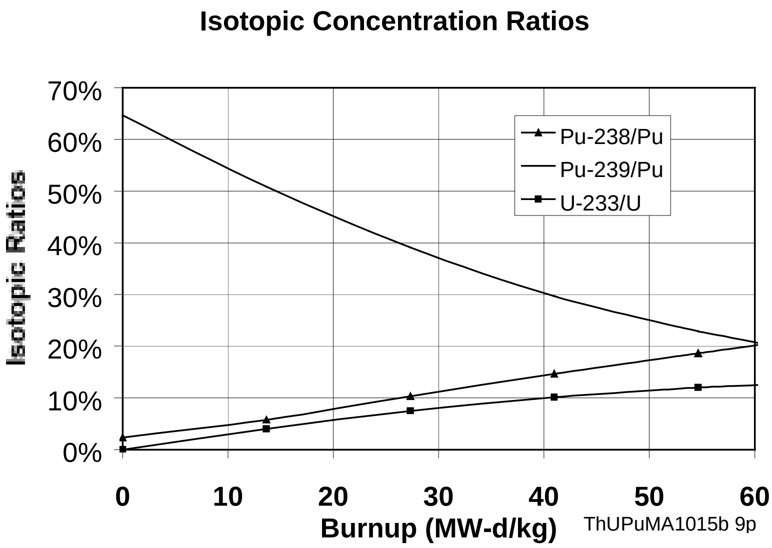


Figure 4. Isotopic Concentration Ratios of Importance to Proliferation Resistance

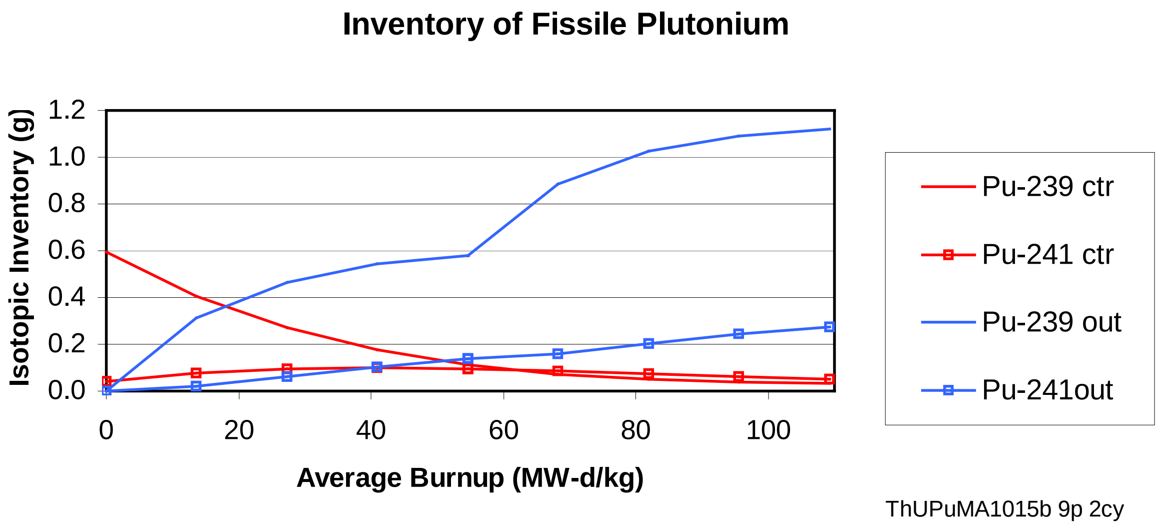


Figure 5 Inventory of Fissile Plutonium Isotopes in the Nine Pellet Model

4. OVERALL PLUTONIUM INVENTORIES

The inventories of ^{239}Pu and ^{241}Pu in the eight outer pellets and in the center pellet are shown in Figure 5. The inventories of the center pellet are shown in red and the total inventory for the eight outer pellets are shown in blue. After a burnup of 52.4 MW-d/kg the center [Th-U-Pu-MA]O_2 pin is placed in a fresh assembly. The inventories of ^{239}Pu and ^{241}Pu in the first set of eight LEU pins is added to the plutonium generated in the fresh LEU outer pins, resulting in the double-growth in the ^{239}Pu curve for the outer pins.

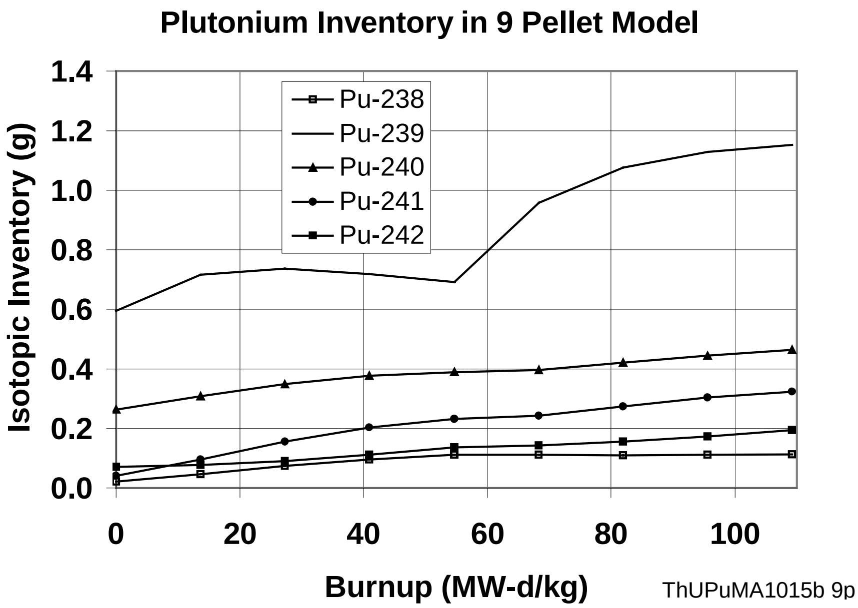


Figure 6 Overall Plutonium Inventory in the 9 Pellet Model

We have tracked the total inventory of plutonium isotopes for both the center transmuter pellet and the outer LEU pellets in Figure 6. Note that the total inventory of ^{239}Pu is about 0.6 gm at BOL and about 0.7 gm at 52.4 MW-d/kg. At BOL the ^{239}Pu is entirely contained in the transmuter pin, while at 52.4 MW-d/kg, about 0.15 g is left in the transmuter pellets and 0.55 g has been generated in the outer LEU pellets. Thus the center pellet is consuming ^{239}Pu at nearly the same rate that it is being generated in the outer pins. This reduction is similar to the CORAIL concept [5], where the overall amount of plutonium is kept constant throughout the fuel cycle (i.e., a net-zero production) using a 31% loading of MOX pins. Note, however, that only 11% of the pins contain plutonium and MAs in the concept presented here, thus allowing for much simpler core design changes based on reactivity coefficients and hot channel concerns.

5. FISSION HEATING PROFILE

Finally, we have tracked the fission heating in the transmuter pellet from BOL to 109 MW-d/kg. The results are shown in Figure 7. In standard LEU pellets the fission heating is fairly uniform at BOL and becomes peaked as ^{239}Pu is produced at the periphery at high burnup. The opposite occurs in the transmuter pellets. The ^{239}Pu is initially uniformly distributed in the center pellet. However, because of the strong fission resonance at 0.3 MeV, the peripheral ^{239}Pu is more rapidly consumed in fission and the heat profile flattens with high burnup.

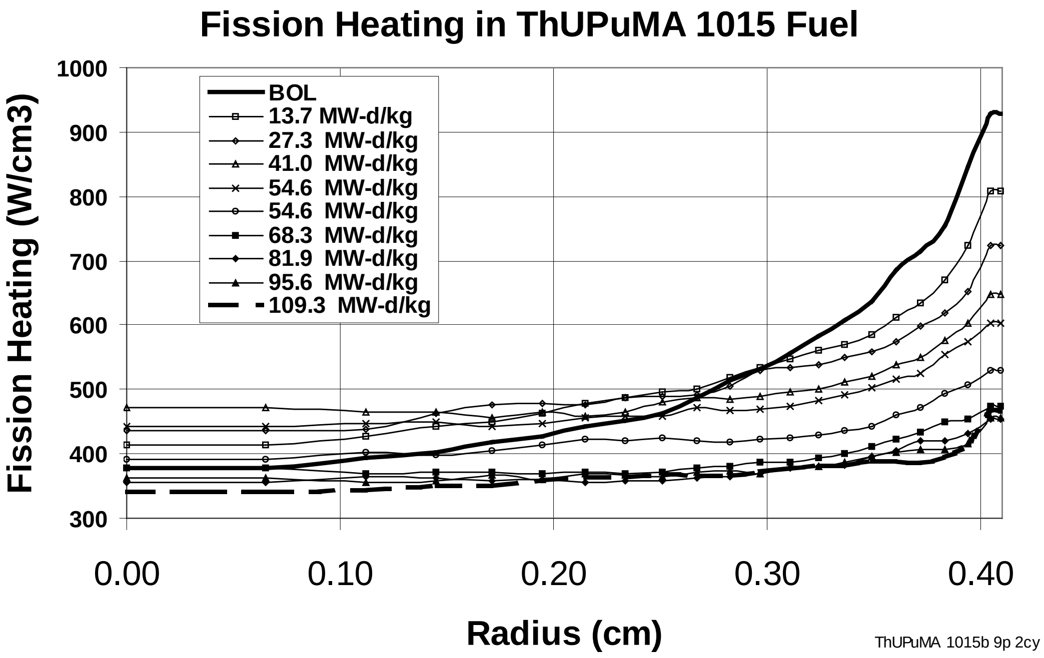


Figure 7 Fission Heating Profile in Transmuter Pellet

6. CONCLUSIONS

Use of thorium-based fuels continues to be promising in the development of proliferation resistant fuel forms. In particular, the intrinsic resistance of plutonium mixtures with high ^{238}Pu is enhanced through high decay heat and spontaneous neutron production.

Use of thorium-uranium fuels, and perhaps fuels with non-fertile matrices, shows promise as LWR transmuter fuels that would significantly reduce the amount of transuranics (TRU) going to a permanent repository or to long-term interim storage. Furthermore, these transmuter fuels used in a mono recycling or "twice through fuel cycle" significantly decrease the volume of spent fuel going to a repository while greatly increasing the proliferation resistance and waste form durability of the resulting fuel. Work is continuing in the analysis of non-fertile and thorium-based LWR transmuter fuels.

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