

QUANTITATIVE COMPARISONS OF ACTINIDE PARTITIONING-TRANSMUTATION IN LIGHT WATER REACTORS AND FAST REACTORS

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Abstract

Previous partitioning-transmutation (P-T) studies at the Oak Ridge National Laboratory showed that partitioning of actinides from light water reactor (LWR) spent fuel and subsequent transmutation in LWRs can contribute significantly to the extension of repository lifetime for well over 100 years. At some time in the future, fast reactors (FRs) are expected to be added to the U.S. fleet of commercial reactors, especially when the cost of natural uranium becomes excessive. Interim use of FRs as actinide burners is also planned. Thus, a study was initiated to quantitatively compare the P-T performance of the systems using LWRs, FRs, or combinations of the two. For the LWR systems, the previous studies showed that a major effect on the pathway of actinide transmutation is based on the length of decay time for ^{241}Pu (half-life—14.3 years). For long-aged fuel (>30 years), the transmutation pathway is predominantly through ^{241}Am to ^{238}Pu ; whereas, for short-aged fuel (<10 years), the pathway is predominantly through ^{242}Pu to ^{243}Am and curium isotopes. In comparison, for the FR systems, the same effects occur but at a much slower rate, primarily because of a much slower rate of conversion of ^{240}Pu to ^{241}Pu . Thus, the rates of production of nuclides heavier than ^{241}Pu are slowed significantly. Another significant effect observed is that the minor actinides (Np, Am, Cm) created when low-enriched uranium is irradiated in LWRs are not destroyed as fast in subsequent irradiation in FRs as in LWRs.

INTRODUCTION

In previous studies, multicycle partitioning-transmutation (P-T) in existing and future thermal spectrum, light water reactors (LWRs) was evaluated to determine the approach to equilibrium mass levels of transuranic (TRU) actinides [1]. The use of compact advanced burner reactors (ABRs), using fast spectrum irradiation at low conversion ratios (<0.5), is now planned. Previous studies indicated that the ABRs would partially consume the TRU actinides and that larger portions of fission products and lesser amounts of heavier actinide isotopes would be created [2]. However, no direct comparison of the approaches to equilibrium mass levels of the TRU actinides has been published heretofore. Therefore, this study was initiated to provide a direct comparison.

IRRADIATION CONFIGURATION

Fast Reactor Model

The lattice representing the seven-ring, compact, sodium-cooled, low-conversion ratio fast reactor design described by Argonne National Laboratory [3] was modeled using the HELIOS code, 112 neutron groups, and the fast reactor cross section library. In the reactor lattice model, each of the fuel assemblies contained 271 fuel rod positions. The active fuel height was 113 cm. The seven-batch compact core contained a total of 102 driver fuel assemblies and was operated at a power of 840 MWth, using a capacity factor of 85%. Total irradiation time was ~ 3 years. Three fuel rod diameters of 0.67, 0.62, and 0.59 cm were used to provide conversion ratios of 0.50, 0.35, and 0.25, respectively.

Light Water Reactor Model

In all cases (for both Am-Cm and Pu-Np), the transmutations were driven by enriched ^{235}U drivers. The enrichment of the ^{235}U in the driver fuel rods and in the Am-Cm target diluent was kept below the currently approved limit for commercial enrichment (5.0% ^{235}U). By using enriched ^{235}U drivers, the fuel/target rod loadings could be kept constant during the multiple P-T cycles. In most of the calculations made in this study, the Am-Cm “target” rods consisted of a loading of 10.0 wt % Am-Cm in a matrix of UO_2 containing 5.0 wt % ^{235}U . Each fuel assembly consisted of 48 target rods inserted into a standard 17×17 pressurized water-reactor (PWR) fuel rod configuration, together with 216 standard “driver rods” containing UO_2 fuel enriched to 5.0 wt % ^{235}U .

Similarly, the Pu-Np rods consisted of a loading of 9.28 wt % plutonium plus neptunium both in the oxide form and in a matrix of depleted UO_2 . Each fuel assembly consisted of 104 mixed oxide (MOX) rods, together with 160 standard “driver rods” containing UO_2 fuel enriched to 3.5 wt. % ^{235}U .

The fuel assemblies were irradiated for three reactor cycles of 18 months each in a 3400-MWth core, which contained 193 fuel assemblies. Detailed multi-dimensional neutronics calculations were performed with the HELIOS code using 45 neutron groups.

PARTITIONING-TRANSMUTATION SCENARIOS EVALUATED

Figures 1 and 2 show the P-T scenarios evaluated. The TRU actinide feed material for both cases was obtained from LWR UO_2 spent fuel that had been irradiated for ~ 45 GWd/MT and had decayed for 30 years. A constant electric power production of 100 GWe was assumed for the entire system for both P-T scenarios. Initially, the power production was all from LWRs, and it resulted in ~ 2000 MT/year of spent fuel containing ~ 23 MT/year of TRU actinides. Recycled fuel displaced a proportionate amount of the LWR UO_2 fuel in the overall system as multiple cycles occurred.

For the fast reactor scenario (Fig. 1), a homogenous core with uranium-TRU-zirconium metal fuel was used. Conversion ratios of 0.50 and 0.25 were evaluated. The thermal reactor scenario (Fig. 2) was

the same as used in previous studies—a heterogeneous core design with U-Pu-Np MOX fuel and U-Am-Cm targets [1].

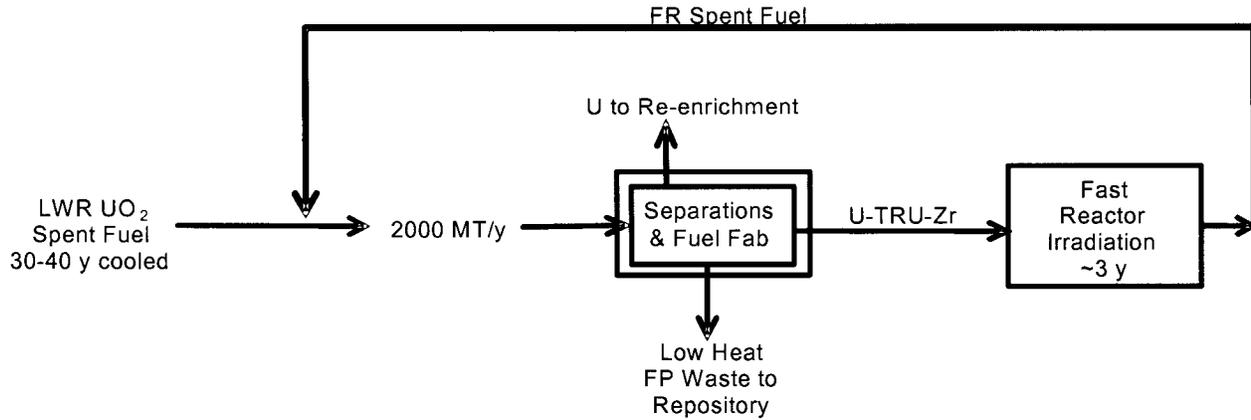


Fig. 1. Fast burner reactor-transmutation scenario.

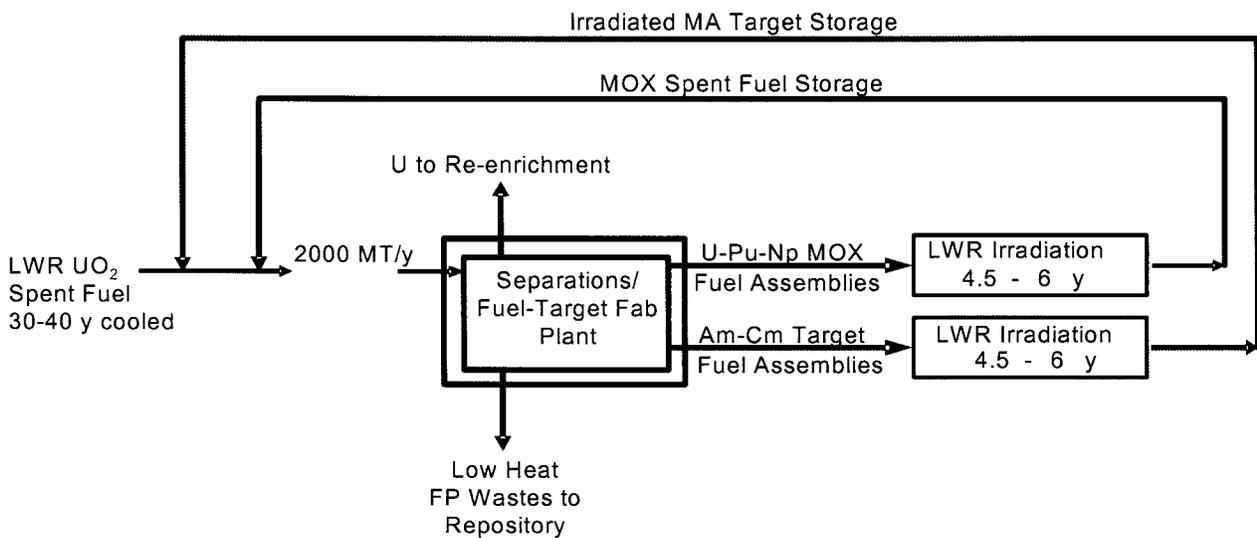


Fig. 2. P-T scenario using thermal transmutation in LWRs.

FIRST CYCLE RESULTS

TRU actinide compositions before and after irradiation for each of the fast and thermal spectrum reactors are shown in Table 1. The feed composition was the same in each case and is shown in the column on the left side of the table. The ABRs with conversion ratios of 0.50 and 0.25 are designated “FR/0.50CR” and “FR/0.25CR,” respectively.

The data in Table 1 were used to calculate and compare burn-up percentages (net destruction and net production), as shown in Table 2. These comparisons showed that the burn-up rates of ^{239}Pu , ^{240}Pu , ^{237}Np , and ^{241}Am achieved in an LWR are greater than in an ABR. However, the net production (negative burn-up in Table 2) of ^{241}Pu , ^{242}Pu , ^{243}Am , and ^{244}Cm is significantly greater in the LWR. A net production of the isotope, ^{238}Pu , occurs in both reactors by transmutation of ^{237}Np and ^{241}Am , but the accumulation of ^{238}Pu is greater in the LWR.

Table 1. TRU actinide compositions before and after irradiation

	1 st cycle transmutation feed			
	LEU spent fuel 30-year decay	FR/0.50 CR spent fuel @ discharge	FR/0.25 CR spent fuel @ discharge	LWR recycle combined spent fuel @ discharge
^{237}Np , MT/y	1.097	0.712	0.645	0.44
^{238}Pu , MT/y	0.302 (1.5%)	0.728 (4/5%)	0.747 (5.2%)	1.57 (12.1%)
^{239}Pu , MT/y	13.05 (66.6%)	9.56 (58.7%)	7.68 (53.6%)	3.53 (27.2%)
^{240}Pu , MT/y	4.66 (23.8%)	4.51 (27.7%)	4.41 (30.8%)	3.97 (30.6%)
^{241}Pu , MT/y	0.703 (2.6%)	0.620 (3.8%)	0.622 (4.3%)	2.06 (15.9%)
^{242}Pu , MT/y	<u>0.888 (4.5%)</u>	<u>0.880 (5.4%)</u>	<u>0.870 (6.1%)</u>	<u>1.83 (14.1%)</u>
Total Pu, MT/y	19.60	16.30	14.33	12.96
^{241}Am , MT/y	2.31	1.41	1.29	0.47
^{243}Am , MT/y	<u>0.177</u>	<u>0.190</u>	<u>0.191</u>	<u>0.40</u>
Total Am, MT/y	2.49	1.67	1.547	0.88
^{244}Cm , MT/y	<u>0.0166</u>	<u>0.057</u>	<u>0.063</u>	<u>0.340</u>
Total Cm, MT/y	0.0198	0.181	0.205	0.559

Table 2. Burn-up^a comparisons
(Values in parentheses are net production)

	FR/0.50 CR	FR/0.25CR	LWR
^{239}Pu	27%	41%	73%
^{240}Pu	3%	5%	15%
^{241}Pu	12%	12%	(193%)
^{242}Pu	1%	2%	(106%)
^{243}Am	(7%)	(8%)	(126%)
^{244}Cm	(243%)	(280%)	(1950%)
^{237}Np	35%	41%	60%
^{241}Am	39%	44%	80%
^{238}Pu	(141%)	(147%)	(420%)

^a Burn-up = (amount in feed – discharge amount)/amount in feed

Interpretation of Results

Expected differences in distribution of TRU actinides in fast and thermal spectrum irradiations are implied in many reports to be based only on the increased ratio of fission cross sections to capture cross sections, resulting in greater amounts of fissioning for the TRU actinides in fast spectrum irradiations. However, the creation of TRU actinides does not depend only on fission cross sections, but does depend on the capture cross sections of the parent isotopes (or on decay of the parent isotopes in some cases). Also, the accumulation of a particular isotope depends on the differences in formation versus destruction rates; and the destruction rates are dependent on the sum of capture, fission, and decay of the isotope.

It is well known from comparative cross-section data for TRU actinides that both capture and fission cross sections for most isotopes are significantly decreased in fast spectrum irradiations versus those in thermal spectrum irradiations, but that the relative decreases are different for the different isotopes. However, the neutron flux in fast reactors is typically about 10 times higher than in LWRs, thus mitigating the lower cross sections in the calculation of reaction rates.

Still, most of the formation and destruction reaction rates per unit of mass of each isotope are lower in the ABR (FR/0.25CR) than in the LWR, as shown in Table 3. Only the formation rate of ^{239}Pu and the destruction rates of ^{238}U and ^{238}Pu are greater in the ABR (FR/0.25CR). For ^{239}Pu , the formation rate is increased by a factor of 2.91 and the destruction rate is decreased by a factor of 2.84, resulting in a gain in the accumulation of ^{239}Pu in the ABR (FR/0.25CR) relative to that in the LWR. For ^{240}Pu , the destruction rate is decreased by a larger factor (12.2) than the formation rate decrease factor (6.6), again resulting in a gain in the accumulation in the ABR (FR/0.25CR) relative to the LWR.

Table 3. Relative TRU isotope formation and destruction rates in the ABR(FR/0.25CR) in comparison to those in the LWR

Product isotope	Parent isotope	(+ = increase; - = decrease) in relative formation rate ^{a,c}	(+ = increase; - = decrease) in relative destruction rate ^{b,c}	Effect ^d
^{238}U		—	+ 3.39 x	
^{239}Pu	(^{238}U)	+2.91x	- 2.84 x	gain
^{240}Pu	(^{239}Pu)	- 6.60 x	- 12.2 x	gain
^{241}Pu	(^{240}Pu)	- 29.6 x	- 1.2 x	reduction
^{242}Pu	(^{241}Pu)	- 2.5 x	- 1.5 x	reduction
^{243}Am	(^{242}Pu)	- 3.27 x	- 1.92 x	reduction
^{244}Cm	(^{243}Cm)	- 4.01 x	- 1.11 x	reduction
^{241}Am	^{241}Pu	(decay)	- 2.10 x	
^{238}Pu	(^{241}Am)	- 2.70 x	+ 1.34 x	reduction
^{242}Pu	(^{241}Am)	- 2.70 x	- 1.54 x	reduction
^{237}Np	(^{236}U)	- 1.00 x	- 1.24 x	gain
^{238}Pu	(^{237}Np)	- 1.73 x	+ 1.34 x	reduction

^a “Increased formation rate” = capture rate of parent isotope in FR/capture rate of parent isotope in LWR, and “decreased formation rate” = (-1) capture rate of parent isotope in LWR/capture rate of parent isotope in FR.

^b “Increased destruction rate” = destruction rate (capture + fission) of product isotope in FR/destruction rate of product isotope in LWR, and “decreased destruction rate” = destruction rate of product isotope in LWR/destruction rate of product isotope in FR.

^c Rates are per unit mass of each isotope.

^d Effect on product isotope in FR relative to LWR.

More significantly, the formation rate of ^{241}Pu is greatly reduced (by a factor of ~ 30) in the ABR (FR/0.25CR) relative to that in the LWR, while the destruction rate is decreased by only a small factor (1.2). This results in a massive reduction in the accumulation of ^{241}Pu and the sequentially produced heavier isotopes, ^{242}Pu , ^{243}Am , and ^{244}Cm . Thus, it appears that irradiation of plutonium in a fast spectrum is optimum, because the production of ^{241}Pu (and its decay daughter, ^{241}Am) as well as of ^{242}Pu , ^{243}Am , and ^{244}Cm are minimized.

However, when minor actinide isotopes, predominantly ^{237}Np and ^{241}Am , are initially present in the transmutation feed, as they are in the spent fuel produced by irradiation of low-enriched uranium, the destruction rates are slower in the ABR (FR/0.25CR) than in the LWR, as shown in Table 3. This indicates that the transmutation of minor actinides in an LWR is optimum.

The isotope ^{238}Pu is produced by transmutation of ^{247}Np and ^{241}Am in both fast and thermal spectrum irradiations. Table 3 shows that the destruction rate of ^{238}Pu is increased and the formation rates are decreased in the ABR (FR/0.25CR) relative to those in the LWR; thus, a smaller accumulation of ^{238}Pu is achieved in the ABR (FR/0.25CR).

MULTIPLE P-T CYCLE COMPARISONS

Two cases were evaluated for the fast reactor scenario, both using the 0.25 conversion ratio core. In one of these cases, the fast reactor spent fuel was recycled after a 5-year decay period and, in the second case, the spent fuel was recycled after a 30-year decay period. In the case evaluated for LWR irradiation, the spent fuel was recycled after a 30-year decay period.

Plutonium Isotope Inventories

Plutonium isotope inventories in the feed to each cycle are compared for the three cases in Fig. 3. The multi-cycle profiles for the fast reactor cases are nearly identical; this is because the ^{241}Pu content is low in both cases.

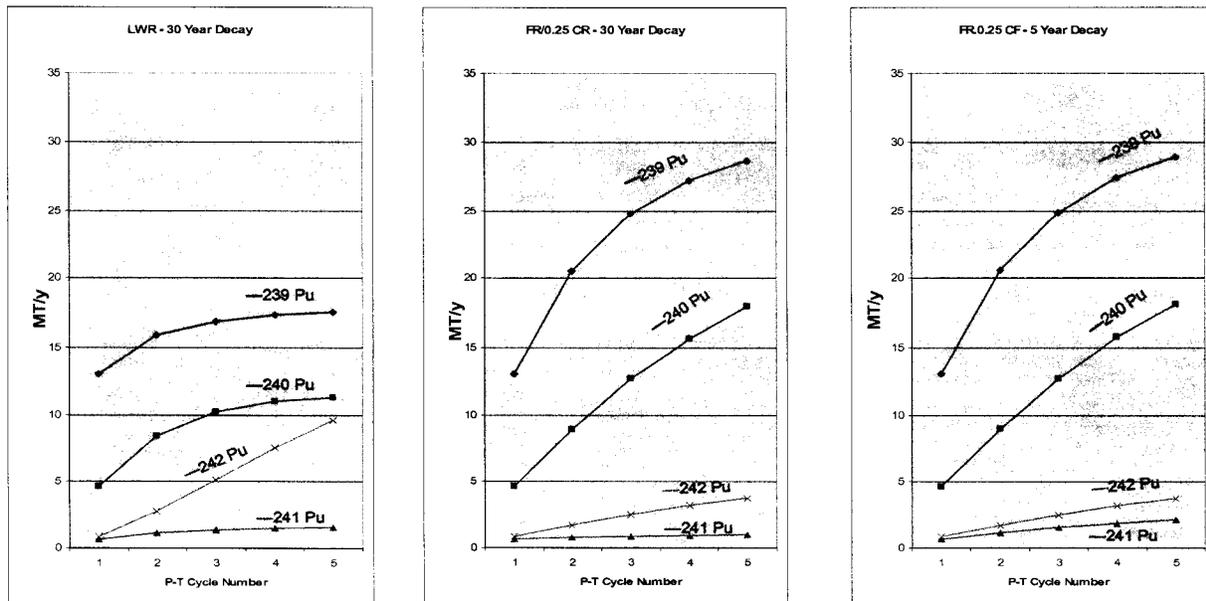


Fig. 3. Comparison of plutonium isotope inventories in the feed to each cycle during multiple P-T cycles.

In comparison, the ^{239}Pu and ^{240}Pu content in the multi-cycle profiles for the LWR case approach equilibrium more quickly and at lower equilibrium levels because of the larger burnup in each cycle. Also, the ^{242}Pu content increases at a significantly greater rate than in the fast reactor cases because of the increased generation rate in each cycle. The amount of ^{241}Pu generated in each cycle also increases, but $\sim 75\%$ of the ^{241}Pu decays to ^{241}Am during the 30-year decay period. Thus, the ^{241}Pu content in the feed to each cycle remains relatively low.

^{238}Pu and Precursors

The ^{238}Pu inventory, as well as the pre-cursor isotopes (^{241}Am , ^{237}Np) inventories, are compared in Fig. 4 during multiple P-T cycles. For the fast reactor cases, the ^{238}Pu and ^{241}Pu contents are higher and the ^{241}Am content is smaller when the shorter (5-year) decay period is used in each P-T cycle.

In comparison, the ^{238}Pu content is greater in the LWR multi cycles. Also, the ^{241}Am is equally high or slightly greater than in the fast reactor multi cycles. This occurs even though the ^{241}Am burnup in the LWR is greater in each cycle, because an almost equal amount of ^{241}Am is generated by decay from ^{241}Pu . It is this phenomenon that indicates the optimum results would result from irradiating ^{241}Am in LWRs where greater burn-up occurs and from irradiating plutonium in fast reactors to prevent formation of ^{241}Pu and its decay daughter, ^{241}Am .

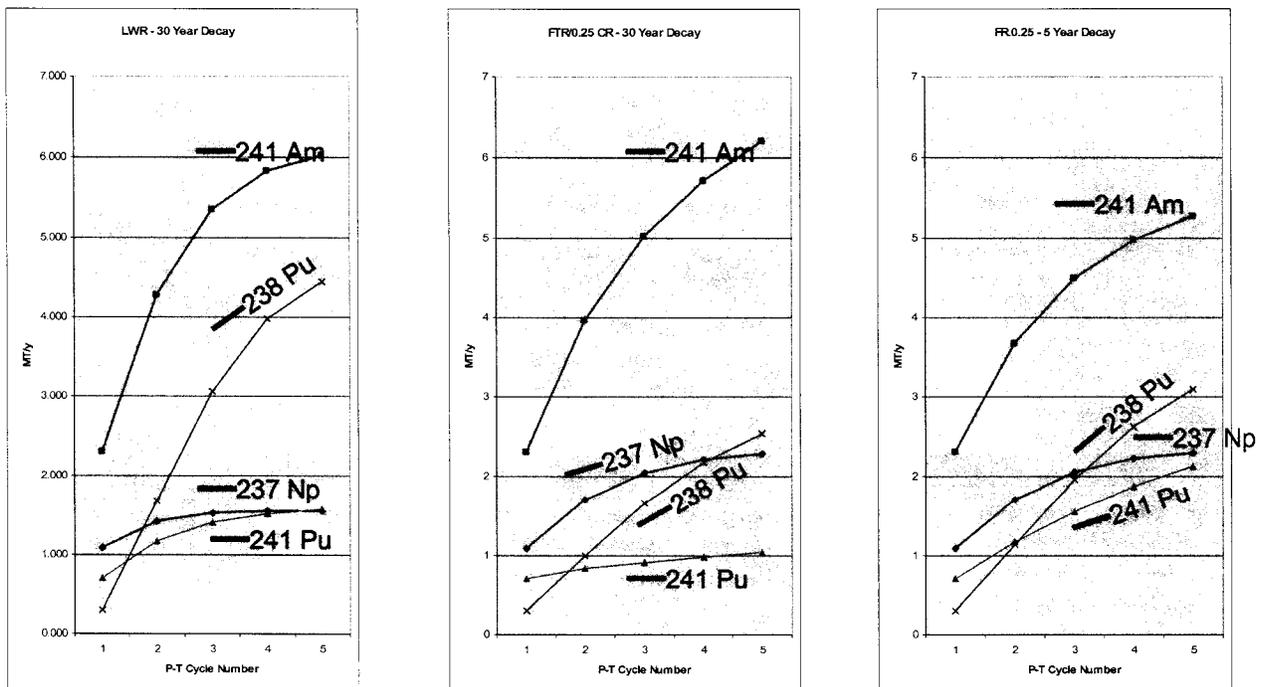


Fig. 4. Comparison of ^{238}Pu and its precursor isotope inventories in the feed to each cycle during multiple P-T cycles.

TRU Elements

The elemental isotopes were summed to compare the TRU element inventories during multiple P-T cycles in Fig. 5. The inventory profiles for the two fast reactor cases are similar. In the LWR case, the minor TRU actinide inventories are similar to the fast reactor cases, but the plutonium inventory

approaches equilibrium more quickly and at a lower amount. Overall, the performance using compact ABRs (FR/0.25 CR) is not greatly different from that obtained in LWRs with 30-year decay periods.

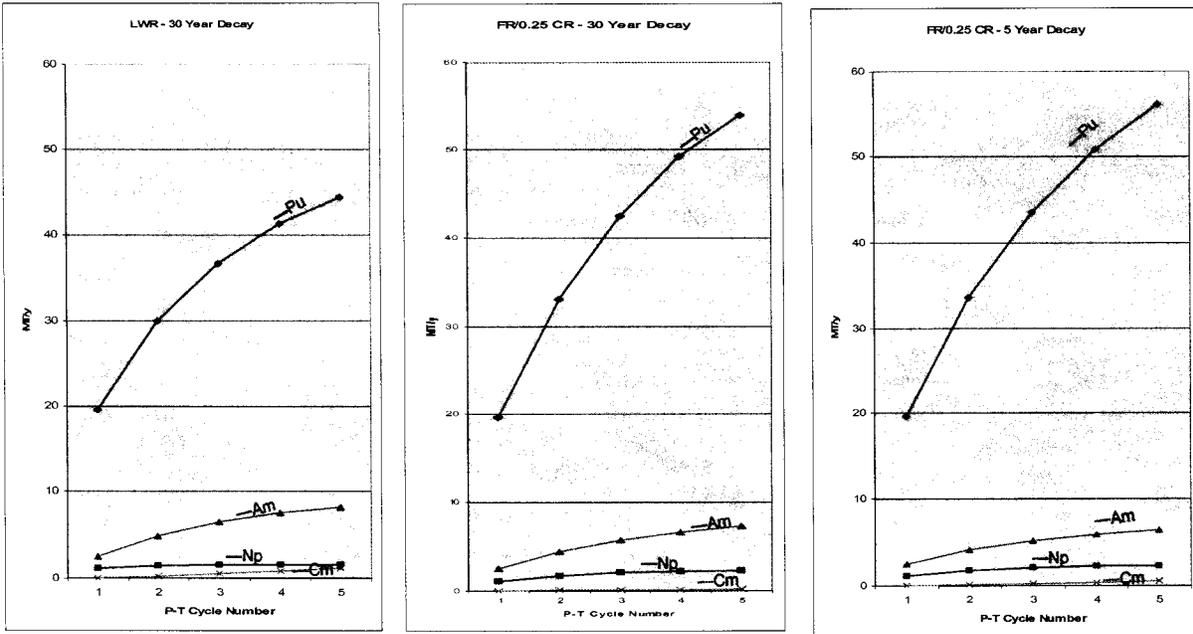


Fig. 5. Comparison of TRU element inventories in the feed to each cycle during multiple P-T cycles.

Total TRUs

All TRU elements were summed to compare the multi P-T cycle performance for the compact ABR (FR/0.25 CR) with that obtained from LWRs with 30-year decay periods. Figure 6 shows the Total TRU mass profiles and Fig. 7 shows the TRU integral heat profile. In both figures, the mass and heat accumulations are compared with once-through operations.

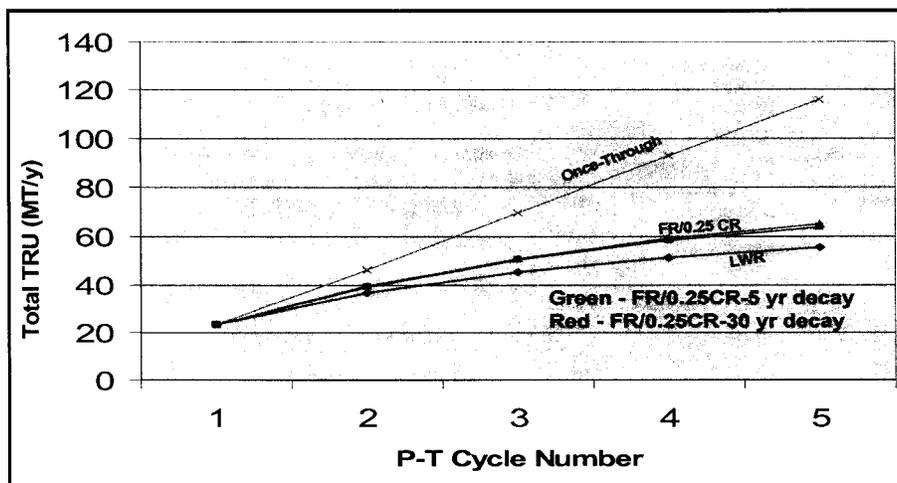


Fig. 6. Comparison of TRU inventory in the feed to each cycle during multiple P-T cycles.

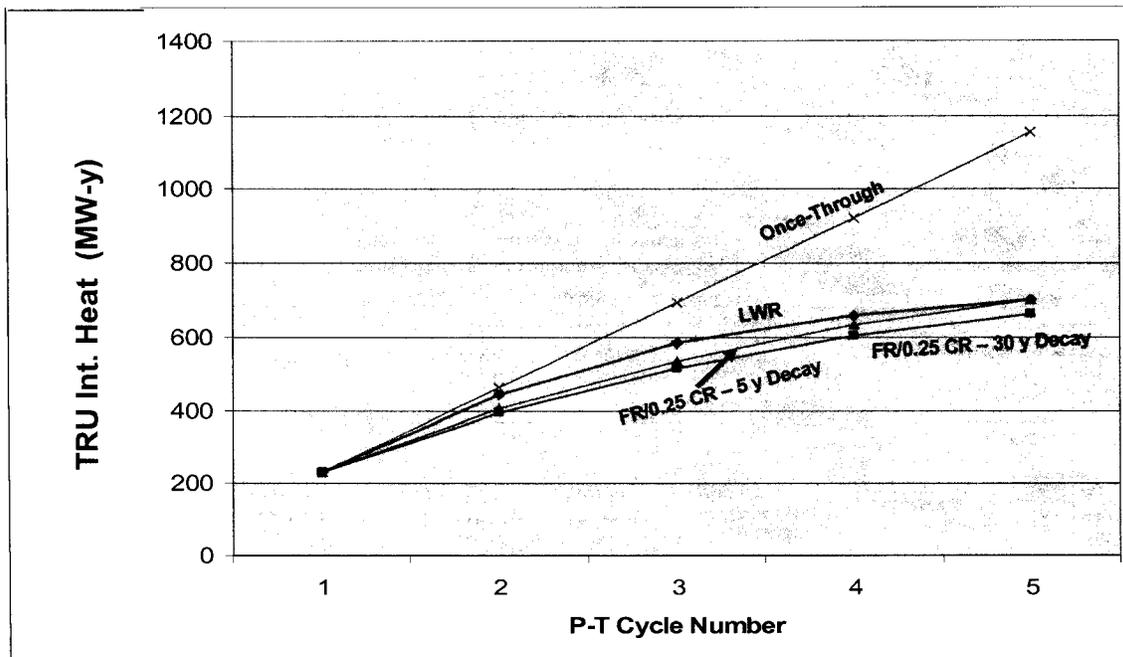


Fig. 7. Comparison of integral heat from TRUs in the feed to each cycle during multiple P-T cycles.

Figure 6 shows that the mass of TRU actinides is actually greater in the spent fuel from the fast spectrum irradiations (FR/0.25 CR) than from the thermal spectrum irradiation (LWR); while Fig. 7 shows that the TRU integral heat content [4] is greater in the spent fuel from the thermal spectrum irradiation (LWR).

Conclusions Based on Comparison of Data with Generic Accumulation Chart

Figure 8 shows the generic accumulation of spent fuel (and TRU actinides) without dimensions because of the uncertainty regarding when recycling will begin. The accumulation in the once-through case is linear (currently ~2,000 MT/year spent fuel and ~23 MT/year TRU actinides). At the point that recycling is begun at the rate of generation, the accumulation begins to decrease. At equilibrium (several P-T cycles), a state of approximate “no net production” is reached.

Starting at the point that recycling is begun at the rate of generation, Fig. 6 shows a similar decrease in TRU accumulation in the spent fuels from both fast spectrum irradiation (FR/0.25 CR) and thermal spectrum irradiation (LWR). However, in all cases, the accumulated inventory on hand at the time recycling begins is not reduced. Even if the “oldest fuel first” concept is used, then the amount of inventory on hand at the beginning of recycling is maintained but not reduced. No additional storage capacity is required for spent fuel assemblies, but the TRU in the existing inventory continues to increase until the “no net production” condition is reached.

Based on this comparison, the conclusions reached are

- (1) Decreasing the inventory will be difficult, even after deployment of ABRs.
- (2) Starting recycling as soon as possible is the most effective way to stop the inventory growth.
- (3) Burning minor actinides (or at least Am-Cm) in LWRs and Pu (or Pu-Np) in FRs appears to be optimum.

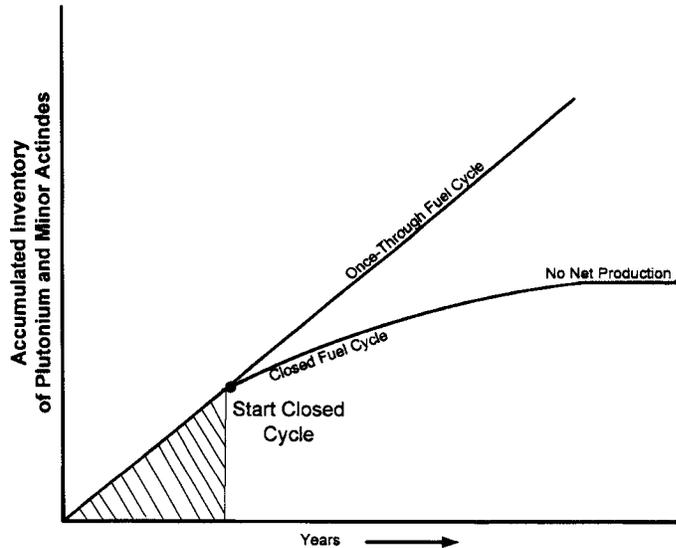


Fig. 8. Generic accumulation rates.

Optimum Burnout of ^{241}Pu - ^{241}Am

Based on the last conclusion, further calculations were done using the HELIOS model to confirm the previous indication. Table 4 shows the same data as Table 2 for one core burnup (first P-T cycle) plus the results in the right-most column for the case where Pu-Np is irradiated in the FR/0.25 CR and Am-Cm is irradiated in the LWR. The comparative results of the ^{241}Pu - ^{241}Am burn-up are shown for each case at the bottom of the chart, and confirm the third conclusion above.

Table 4. Comparative burn-up rates for ^{241}Pu - ^{241}Am

	1 st cycle transmutation feed				Pu-Np in FR/0.25 CR Am-Cm in LWR combined spent fuel @ discharge
	LEU spent fuel 30-year decay	LWR recycle spent fuel @ discharge	FR/0.50 CR spent fuel @ discharge	FR/0.25 CR spent fuel @ discharge	
^{237}Np , MT/y	1.097	0.44	0.712	0.645	0.73
^{238}Pu , MT/y	0.302 (1.5%)	1.57 (12.1%)	0.728 (4.5%)	0.747 (5.2%)	1.32 (8.4%)
^{239}Pu , MT/y	13.05 (66.6%)	3.53 (27.2%)	9.56 (58.7%)	7.68 (53.6%)	8.15 (51.8%)
^{240}Pu , MT/y	4.66 (23.8%)	3.97 (30.6%)	4.51 (27.7%)	4.41 (30.8%)	4.55 (28.9%)
^{241}Pu , MT/y	0.703 (2.6%)	2.06 (15.9%)	0.620 (3.8%)	0.622 (4.3%)	0.69 (4.4%)
^{242}Pu , MT/y	<u>0.888 (4.5%)</u>	<u>1.83 (14.1%)</u>	<u>0.880 (5.4%)</u>	<u>0.870 (6.1%)</u>	<u>1.01 (6.4%)</u>
Total Pu, MT/y	19.60	12.96	16.30	14.33	15.72
^{241}Am , MT/y	2.31	0.47	1.41	1.29	0.40
^{243}Am , MT/y	<u>0.177</u>	<u>0.40</u>	<u>0.190</u>	<u>0.191</u>	<u>0.18</u>
Total Am, MT/y	2.49	0.88	1.67	1.547	0.59
^{244}Cm , MT/y	<u>0.0166</u>	<u>0.340</u>	<u>0.057</u>	<u>0.063</u>	<u>0.142</u>
Total Cm, MT/y	0.0198	0.559	0.181	0.205	0.298
$\Sigma^{241}\text{Pu} + ^{241}\text{Am}$, MT/y	3.01	2.53	2.03	1.91	1.09
$\Sigma^{241}\text{Pu} + ^{241}\text{Am}$, % burn-up	-----	16.0%	32.6%	36.5%	63.8%

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