Evaluation of Actinide Partitioning and Transmutation in Light-Water Reactors

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Abstract *−Advanced Fuel Cycle Initiative (AFCI) studies were made to evaluate the feasibility of multicycle transmutation of plutonium and the minor actinides (MAs) in light-water reactors (LWRs). Results showed that significant repository benefits, cost reductions, proliferation resistance, and effective use of facilities can be obtained. Key advantages are shown to be made possible by processing 30-year-decayed spent fuel rather than the more traditional 5-year-decayed fuel.*

INTRODUCTION

AFCI studies were made to assess effects of existing LWR spent fuel accumulation in the United States, coupled with the absence of fast spectrum reactors and accelerator-driven systems, on the capability to partition/transmute actinides during the next ~300 years. Existing and advanced light water reactors (LWRs) would be used.

Because of the absence of spent fuel processing and mixed oxide (MOX) fuel utilization in the United States, the LWR spent fuel accumulation was ~45,000 MT in 2001. The annual generation rate is currently about 2000 MT per year from the 103 reactors, which produce approximately 100 GW of electricity. The spent fuel is currently being stored at the reactor sites in water pools and dry casks, awaiting future transfer to and disposal in the Yucca Mountain Repository.

The legal capacity limit for commercial fuel storage in the Repository is currently 63,000 MT. The amount of spent fuel in storage will likely exceed the legal capacity of the Repository by the time it is expected to begin receiving spent fuel in 2010. Thus, alternatives to expansion of Yucca Mountain or opening of a second repository are being studied. One of the alternatives is to deploy spent fuel processing and transmutation of plutonium and minor actinides in existing and future LWRs.

Previous studies on deployment of partitioning and transmutation in the United States have indicated several advantages of processing the oldest fuel first.[1] If a plant is built and operated to process spent fuel at the same rate (2000 MT/year) that the fuel is generated and the "oldest-fuel-first" approach is adopted, then the average age of the spent fuel processed will never be less than 35 to 45 years, depending on a starting time in the period 2015-2025. The obvious benefits are the reduced amount of radioactive material in the spent fuel to be processed and the reduced amount of decay heat that must be handled. Recent studies, reported herein, have shown that significant benefits in transmutation also are possible with 35-year fuel cycles (30-year decay periods).

SCENARIO EVALUATED

The processing scenario evaluated (Fig. 1) assumed that (1) 2000 MT/year of spent fuel, irradiated to 45 GWd/MT and decayed for 30 years, is processed; (2) recovered plutonium and 90% of the neptunium are transmuted in LWR MOX fuel; and (3) MAs, consisting of americium, curium, and 10% of the neptunium, are transmuted in "burnable-poison" -type targets.

BENEFITS OF SCENARIO EVALUATED

The scenario evaluated offers significant benefits, which include (1) extended lifetime for the repository, (2) lower costs for partitioning and transmutation of plutonium and the MAs, and for storage of spent fuel, (3) maintenance of proliferation resistance for the fissile plutonium in spent fuels, and, (4) efficient use of fuel/target fabrication facilities. The lifetime of the repository would be extended significantly because all of the plutonium and MAs would be "in process" or "in storage" and only fission products would be put into the repository. The lower costs would be achieved primarily because no capital

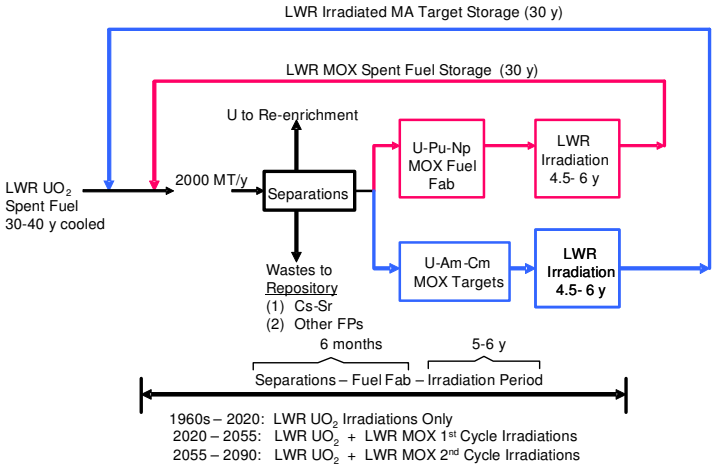


Fig. 1. Scenario evaluated.

investment for a special transmuter reactor (fast reactor, accelerator-driven system, etc.) would be required. Instead, only existing and new LWRs would be utilized.

Moreover, no new storage capacity would be needed for spent fuels and irradiated targets because the number of spent fuel assemblies would remain the same after the scenario is begun. Even though the total inventory of plutonium would rise during the early cycles, ~98% of the plutonium would be contained in stored spent fuel and would be protected by high radiation (the "Spent Fuel Standard"). This is because the spent fuel would be reprocessed and re-irradiated at intervals within which the fission products, ^{137}Cs and ^{90}Sr, both with half-lives of ~30 years, exist in significantly high concentrations.

Further, the scenario evaluated would allow efficient use of the fabrication facilities because the larger fraction (uranium-plutonium-neptunium) can be fabricated into MOX fuel in conventional glove-box-contained equipment, whereas the smaller fraction (americium-curium-diluent) will require more expensive shielded containment. Also, the scenario would allow different irradiation and/or decay times for the MOX fuel and MA targets if necessary to optimize the transmutation process.

IRRADIATION CONFIGURATION

In most of the calculations made in this study, the MA "target" rods consisted of a loading of 10.0 wt % MAs in a matrix of UO_2 containing 5.0 wt % ^{235}U. Each fuel assembly consisted of 48 MA target rods inserted into a standard 17 H 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.

The MOX rods consisted of a loading of 9.28 wt % plutonium plus neptunium in a matrix of depleted UO_2. Each MOX fuel assembly consisted of 104 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-MW(t) core, which contained 193 fuel assemblies. Detailed multi-dimensional neutronics calculations were performed with the HELIOS code using 45 neutron groups.

Calculations were also performed to determine the void reactivity coefficients for the MA target and the MOX fuel assemblies. These calculations were performed at full power conditions for a relative void fraction of 90% and 50%. The void reactivity coefficients were negative for all cases. The peak power density for the MA target assemblies typically increased by 6% at the beginning-of-life.

INITIAL SCOUTING CALCULATIONS

Initial calculations were made with MA targets containing a loading of 10 wt % in either a fertile (UO_2) or an inert matrix (Zr) diluent to compare the effectiveness of transmutation. This was measured by the comparative burnup of ^{241}Am, which is the predominant (~88%) MA component in LWR-UO_2 spent fuel, after 30-year decay. With the inert matrix diluent, the ^{241}Am burnup was 96%, and with the fertile matrix diluent, the burnup was 87%. This was not considered to be significantly different. Future fuel development tests may show that an inert matrix diluent is required to obtain satisfactory target performance. However, the use of an inert matrix diluent will add significant amounts of material that eventually becomes additional waste. All further calculations were made using UO_2 as the MA diluent.

ACTINIDE COMPOSITIONS

Table 1 shows the distribution of neptunium, plutonium, americium, and curium radionuclides in the first transmutation cycle. The left-side column indicates the distribution of 30-year decayed LWR-UO_2 spent fuel components into LWR MOX fuel containing uranium, plutonium, and 90% of the neptunium and into MA targets containing uranium, americium, curium, and 10% of the neptunium. The middle column shows the composition of the MA targets after three 18-month reactor cycles, followed by a 30-year decay period. Similarly, the right-side column shows the composition of the LWR MOX spent fuel.

Table 1. Actinide Mass/Isotopic Compositions in 1st Transmutation Cycle

	Spent LWR-UO_2		Irradiated MA Targets	Spent LWR-MOX
	to MA Targets	to MOX Fuel		
Np, MT/year	0.11	0.98	0.06	0.38
Pu, MT/year	—	19.6	1.6	9.7
^{238}Pu, %	—	1.5	51.9	5.7
^{239}Pu, %	—	66.6	25.1	33.5
^{240}Pu, %	—	23.8	6.8	39.6
^{241}Pu, %	—	3.6	1.0	4.8
^{242}Pu, %	—	4.5	15.3	16.4
Am, MT/year	2.49	—	0.46	1.9
^{241}Am, %	92.8	—	74.4	84.6
^{243}Am, %	7.1	—	24.1	15.3
Cm, MT/year	0.020	—	0.068	0.095
^{243}Cm, %	1.3	—	5.8	0.7
^{244}Cm, %	83.7	—	61.1	70.5
^{245}Cm, %	13.9	—	27.6	25.4
^{246}Cm, %	1.0	—	5.5	3.3
Total HM/MT/year	26	221	24	204

SIGNIFICANT FEATURES OF THE SCENARIO EVALUATED

Two features of the scenario enable multiple cycles to be attainable. The first is the blending of components from the LWR-MOX spent fuel, irradiated MA targets, and LWR-UO_2 spent fuel at the beginning of the separations process, as illustrated in Table 2. Because of the relatively small content (<1%) of plutonium and MAs in spent LWR-UO_2 fuel, only 11.4% of the feed materials for the first cycle MOX and MA irradiations contain recycled actinides and the remaining 88.6% is fresh UO_2 fuel. As the number of cycles increase, the ratio of recycled actinides to fresh UO_2 fuel increases, but only to ~20%. Thus, the bulk of the feed is always fresh UO_2 fuel and the heavier actinides from the LWR MOX spent fuel and irradiated MAs are diluted significantly.

Table 2. Actinide Mass/Isotopic Compositions for 2nd-Cycle Feed

	Spent LWR-MOX	+	Irradiated Am-Cm	+	Spent LWR-UO_2	=	2nd Cycle Feed
Np, MT/year	0.38		0.06		0.96		1.40
Pu, MT/year	9.7		1.6		17.4		28.7
^{238}Pu, %	5.7		51.9		1.5		5.7
^{239}Pu, %	33.5		25.1		66.6		53.1
^{240}Pu, %	39.6		6.8		23.8		28.2
^{241}Pu, %	4.8		1.0		3.6		3.9
^{242}Pu, %	16.4		15.3		4.5		9.1
Am, MT/year	1.9		0.46		2.2		4.6
^{241}Am, %	84.6		74.4		92.8		87.5
^{243}Am, %	15.3		24.1		7.1		12.2
Cm, MT/year	0.095		0.068		0.018		0.18
^{243}Cm, %	0.7		5.8		1.3		2.7
^{244}Cm, %	70.5		61.1		83.7		68.3
^{245}Cm, %	25.4		27.6		13.9		25.1
^{246}Cm, %	3.3		5.5		1.0		3.9
Total HM/MT/year	204 10.2%		24 1.2%		1772 88.6%		2000/ 32 1/48 100%

The second feature is an alteration of the transmutation path which is made possible by the long (30-year) decay period. This is primarily because of different transmutation paths that can be seen in Fig. 2. In this chart, the primary path toward production of heavier nuclides (curium) is through

^{241}Pu 6^{242}Pu 6^{243}Am 6^{244m}Am 6^{244}Cm

by means of neutron capture and beta decay reactions.

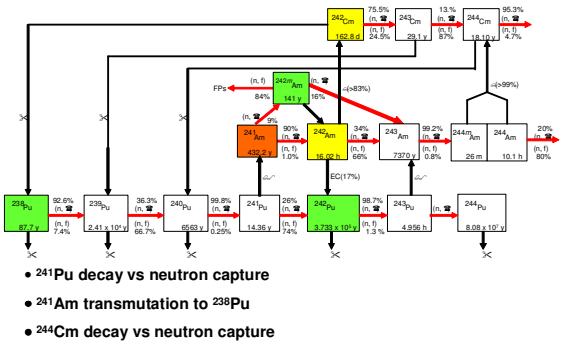
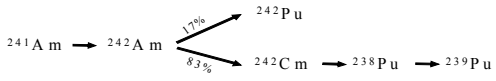


Fig. 2. Nuclear reactions of interest.

With a 30-year decay period, more than 75% of the ^{241}Pu decays to ^{241}Am. Then, during subsequent irradiation, most of the ^{241}Am is transmuted through the pathway



to produce predominantly ^{238}Pu and ^{239}Pu. Still, as indicated, some (~17%) of the ^{241}Am is transmuted to ^{242}Pu and thence to the heavier curium isotopes.

However, during the 30-year decay period, ~2/3 of the previously produced ^{244}Cm will decay to ^{240}Pu. Thus, much of the transmutation pathway is altered to produce lighter plutonium nuclides rather than the heavy curium nuclides.

The net effect of the two significant features of the scenario enables the fissile content of the blended plutonium product from each separation to remain sufficiently high (≥ 40%) for multiple partitioning-transmutation (P-T) cycles to be achieved. Moreover, the rate of production of curium isotopes is slowed significantly (Fig. 3). After 10 P-T cycles, the production rates of the radionuclides in the pathway to heavier elements (^{242}Pu, ^{243}Am, and curium isotopes) are still increasing, but at a relatively low rate. Production rates of the elements neptunium, plutonium, and americium (not shown) are near equilibrium.

COMPARATIVE RESULTS WITH 5-YEAR DECAY (10-YEAR P-T CYCLES)

A similar series of calculations was made to compare the results obtained when using 5-year decay periods (10-year P-T cycles) with the previously obtained results using 30-year decay periods (35-year P-T cycles). Figure 4 illustrates the production rates of the key radionuclides, ^{242}Pu, ^{243}Am, and ^{244}Cm. Although the production rates of ^{243}Am are similar and those of ^{242}Pu are not greatly different, the rate of production of ^{244}Cm is significantly greater with the shorter decay periods and cycle lengths.

When plotted against actual time beginning with the start of the recycling scenario (Fig. 5), the differences are more prolific and indicate the difficulty that would be encountered in the near term (~50 years after start of recycling). During this time, multiple tons per year of ^{244}Cm would need to be handled if the 5-year decay fuel is processed and expensive separations steps (to separate curium from americium) and storage requirements (for curium) may be needed.

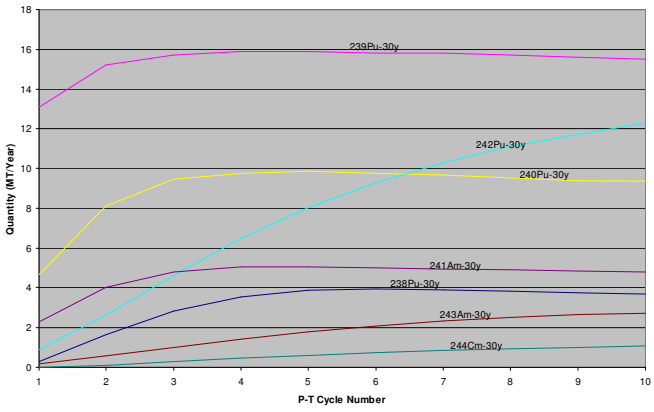


Fig. 3. Production rates of key radionuclides with 30-year decay cycles.

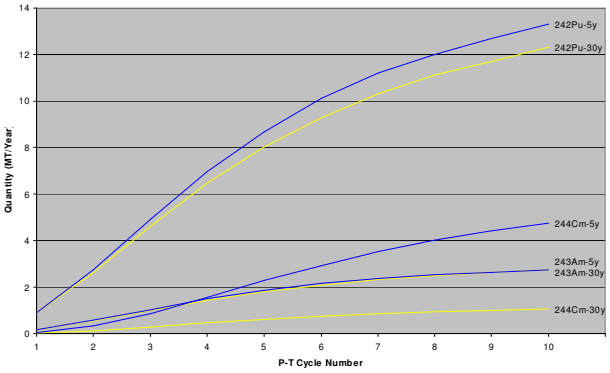


Fig. 4. Comparison of 5-year decay and 30-year decay production rates for each cycle.

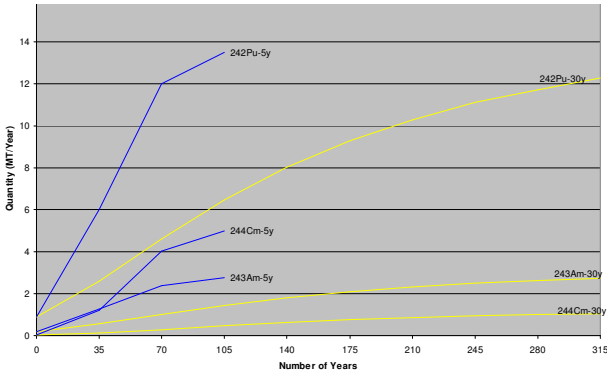


Fig. 5. Comparison of 5-year decay and 30-year decay production rates with time.

SUMMARY

Current studies have shown that significant benefits can be obtained by multicycle transmutation of plutonium and the MAs in LWRs and that key advantages are obtained by processing ~30-year-decayed spent fuel rather than the more traditional 5- or 10-year-decayed fuel.[2,3,4] A key feature in the scenarios evaluated is the dilution effect of bringing spent MOX fuel and irradiated MA targets together with fresh LWR-UO_2 spent fuel at the beginning of the separations processing. Because of the relatively

small amounts of plutonium and MAs produced, the LWR-UO_2 spent fuel will always provide ≥ 80% of the feed material for the next irradiation cycle. The second feature of the scenario evaluated is that key reactions (decay of ^{241}Pu and ^{244}Cm) occur during the 30-year decay period and cause an alteration in the transmutation path, which results in greater production of the lighter plutonium nuclides via transmutation of ^{241}Am and a minimization of the production of ^{244}Cm.

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