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Preliminary Multicycle Transuranic Actinide Partitioning-Transmutation Studies

February 2007

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Nuclear Science and Technology Division

PRELIMINARY MULTICYCLE TRANSURANIC ACTINIDE PARTITIONING-TRANSMUTATION STUDIES

Emory D. Collins, John-Paul Renier, Guillermo D. DelCul, Barry B. Spencer

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Systems Analysis Working Group

This document is a comprehensive system analysis report on preliminary multicycle transuranic actinide partitioning-transmutation studies done at Oak Ridge National Laboratory over the past three years and was prepared at the request of the AFCI/GNEP Systems Analysis Working Group.

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ACRONYMS AND ABBREVIATIONS

ABR	advanced burner reactor
BOC	beginning-of-cycle
BWR	boiling water reactor
EOC	end-of-cycle
IDH	integrated decay heat
IMF	inert-matrix fuel
LEU	low enriched uranium
LWR	light water reactor
MA	minor actinide
MOX	mixed oxide
MT	metric ton
MWt	megawatts thermal power
ORNL	Oak Ridge National Laboratory
P-T	partitioning-transmutation
PWR	pressurized water reactor
R&D	research and development
TRU	transuranic

PREFACE

This document is a comprehensive system analyses report on preliminary multicycle transuranic actinide partitioning-transmutation studies done at Oak Ridge National Laboratory over the past three years and was prepared at the request of the AFCI/GNEP Systems Analysis Working Group.

EXECUTIVE SUMMARY

Preliminary multicycle transuranic (TRU) actinide partitioning-transmutation (P-T) studies have been conducted at Oak Ridge National Laboratory (ORNL) during the past three years to determine the capabilities for consumption of the TRU actinides and to reveal the limiting constraints that must be eliminated or diminished by future research, development, and demonstration. The costs of designing, building, licensing, and operating the required facilities (reactors and separations fuel fabrication plants) were recognized as constraints that must be minimized if multicycle P-T in closed fuel cycles can be deployed successfully. Another significant constraint is the need to ensure that sufficient proliferation resistance methods are provided to prevent the diversion of fissile plutonium to covert use in a nuclear weapon.

Nuclear power reactors [predominantly thermal-spectrum light water reactors (LWRs), either pressurized water reactors (PWRs) or boiling water reactors (BWRs)] are used worldwide for production of electricity. More than 100 power reactors in the United States are currently operated, and significant growth is expected in the future. Spent fuel separation and uranium-plutonium mixed oxide (MOX) fuel fabrication plants are currently operated in Europe, Russia, and Japan, but not in the United States. No large country has, as yet, deployed a geologic waste repository, although several locations are being considered. In the United States, the Yucca Mountain Site in Nevada has been selected, but the process for licensing and deployment has been slowed by sociopolitical factors. The need to begin spent fuel recycle (P-T operations) to conserve the available repository space at Yucca Mountain and to enable sustained use of nuclear power has become apparent.

Early deployment systems studies[1] at ORNL recognized the significant difference in spent fuel accumulation in the United States from that in other countries, especially France, where a closed fuel cycle has already been deployed. Data on spent fuel discharges in the United States have been compiled by the U.S. Energy Information Administration and were summarized in the December 2004 issue of Nuclear News[2]. The data indicate that the rate of accumulation is now approximately 2200 MT per year of heavy metal and that the total inventory is now greater than 55,000 MT. This is a major concern for the future use of the Yucca Mountain Repository because its current legislated capacity is only 70,000 MT; thus, the U.S. Congress is currently considering a bill to allow the legislated capacity to be increased to 120,000 MT. Even then, without spent fuel recycle, a second repository will be needed within the next 30 to 40 years.

Both mass and decay heat of the radioactive waste are significant factors in determining the total capacity of the Yucca Mountain Repository[3]. Removal of the heat generators from the waste that is placed into the repository can result in more compact storage and thus a more efficient utilization of the repository space. Decay heat from the TRU actinides is approximately constant and will be the dominant heat source after ~100 years. During the first ~50 years, the dominant heat sources are the fission products, principally ^{137}Cs and ^{90}Sr, each with a half-life of approximately 30 years. Thus, attainment of the maximum repository benefit requires P-T of all transuranic actinides and managed storage of the fission product waste containing cesium and strontium for ≥ 100 years.

The multicycle P-T studies at ORNL began with an evaluation of the potential benefits that could be obtained by using existing and future thermal spectrum LWRs for the actinide transmutation[4,5]. A specific goal of these studies was to evaluate the approach to equilibrium for each of the TRU actinide isotopes during the early P-T cycles and to interpret the specific transmutation pathway for each of the isotopes.

The effects of several options and constraints were evaluated during the evaluation phase of the study, and the most significant effects were found to be the age (decay time, "cooling time") of the spent fuel and the blending method used (if any). Also, the effect of using inert or fertile diluents for americium-curium targets and the effect of separating and storing the curium instead of recycling it were determined[6].

More recently, the use of compact advanced burner reactors (ABRs), employing fast spectrum irradiation at low conversion ratios (< 0.5) was selected for future transmutation of TRU actinides produced in U.S. LWRs. Previous studies have indicated that ABRs are required to consume the TRU actinides[7]. However, no direct comparison of the approach to equilibrium isotopic compositions during multiple recycling operations with that previously determined for thermal spectrum transmutation had been made. Thus, the ORNL studies were extended to provide a direct comparison of actinide transmutation in LWRs and ABRs.

Efforts were made to evaluate realistic scenarios that would apply to closing the fuel cycle in the United States, including full actinide recycle, during the next ~100 years. Existing conditions in the accumulation of spent fuel in the United States were recognized constraints. Variable conditions were chosen to minimize costs and to provide essential proliferation resistance.

For the scenarios evaluated, the following observations and conclusions were made

- The United States has accumulated more than 55,000 MT of heavy metal in spent fuel and is generating ~2200 MT/year. The legacy fuel must be processed, and the actinides must be recycled to minimize the number of high-level waste repositories required in the future.

- Processing the oldest fuel first will enable numerous technical, environmental, and economic advantages. The spent fuel will have decayed for more than 30 years, and many radionuclides will have decayed significantly prior to processing.

- Utilization of a blending strategy in which recycled actinides are blended with low-enriched uranium spent fuel at the head-end of the separations plant will provide a sufficiently high fissile content for subsequent recycle of the actinide mix and will enable continuous, multicycle operation using either LWRs or ABRs, or combinations of the two types of reactors.

- Utilization of large spent fuel processing (separations and fuel/target fabrication) facilities with overall capacities of 2000 to 3000 MT/year is practical and provides the lowest unit cost for processing.

- Utilization of large co-located and integrated separations and fuel/target fabrications operations located within a physically protected facility will provide significant cost reduction and maximized proliferation resistance.

- Utilization of heterogeneous actinide recycling will provide (1) cost reduction in separations processes, fuel development, and fuel/target fabrication facilities and operation; (2) flexibility of P-T deployment; and (3) improved technical performance. Proliferation risk will not be increased.

The results of the study provided the following conclusions

- Use of multiple P-T cycles (continuous recycle) using only existing and new LWRs is feasible. Use of long decay periods (≥ 30 years) in the P-T cycles using LWRs will enable significant

suppression of the production of curium and heavier actinides during the continued multiple P-T cycles.

- Alternatively, use of short decay periods (~5 years) for irradiation of plutonium in LWRs, as currently practiced in France and other countries, would significantly increase the production of heavier actinides (e.g., ^{241}Pu, ^{242}Pu, ^{243}Am, ^{244}Cm).

- Use of short decay periods (~5 years) can be done effectively by irradiating plutonium in fast reactors and Am-Cm in LWRs because production of ^{241}Pu and heavier actinides is suppressed significantly in those cases.

- Minor actinides (Np, Am, Cm) are more effectively burned in LWRs than in ABRs.

- Optimum performance can be obtained by irradiating Pu (or Pu-Np) in fast spectrum reactors and by irradiating Am-Cm targets in thermal spectrum reactors.

- The approach to equilibrium of the actinides during multiple P-T cycles was not significantly different in cases evaluated for (1) all-thermal-spectrum (LWR) irradiations, (2) all-fast-spectrum (ABR) irradiations, or (3) hybrid irradiations (Pu-Np in fast reactors and Am-Cm in thermal reactors).

- Because the ABR design size has been optimized at ~840 MWt, a large number (33–90) of ABRs would be required to transmute the ~23 MT/year TRU actinides currently produced in ~2000 MT/year of low-enriched uranium spent fuel; in comparison, 10–24 existing (or new) 3400 MWt LWRs would be sufficient.

Based on these conclusions, full near-term implementation of P-T in the United States using only ABRs will be difficult; whereas, near-term deployment using LWRs could be utilized. Similar results would be achieved if the oldest (legacy) spent fuel is processed first.

Because the actinides are only partially destroyed in each P-T cycle, using either thermal or fast reactor transmutation, multiple cycles can only be used to stabilize the growth to a state of approximately "no net production." The inventory of TRU actinides in spent fuel that exists when recycling is begun cannot be reduced unless significantly more spent fuel is processed than is being produced. Production is currently ~2000 MT/year (~23 MT/year of TRU actinides) and is expected to grow in the future. Thus, processing facilities with capacities significantly greater than 2000 MT/year will be required to reduce the inventory and to provide the fissile plutonium needed to fuel future fast and thermal reactors.

Starting recycle as soon as possible is the most effective way to stop actinide inventory growth and the continuing need to provide for storage of the spent fuel assemblies that are generated each year. Cost of providing this storage has been estimated to be more than $500 million per year.

1. INTRODUCTION

1.1. Objectives

Preliminary multicycle transuranic (TRU) actinide partitioning-transmutation (P-T) studies have been conducted at Oak Ridge National Laboratory (ORNL) to determine the capabilities for consumption of the TRU actinides and to reveal the limiting constraints that must be eliminated or diminished by future research, development, and demonstration. The costs of designing, building, licensing, and operating the required facilities (reactors, separations fuel fabrication plants) were recognized as constraints that must be minimized if multicycle P-T in closed fuel cycles can be deployed successfully. Another significant constraint is the need to ensure that sufficient proliferation resistance methods are provided to prevent the diversion of fissile plutonium to covert use in a nuclear weapon.

1.2. History

Nuclear power reactors [predominantly thermal-spectrum light water reactors (LWRs), either pressurized water reactors (PWRs) or boiling water reactors (BWRs)] are used worldwide for production of electricity. More than 100 power reactors in the United States are currently operated, and significant growth is expected in the future. Spent fuel separation and uranium-plutonium mixed oxide (MOX) fuel fabrication plants are currently operated in Europe, Russia, and Japan, but not in the United States. No large country has, as yet, deployed a geologic waste repository, although several locations are being considered. In the United States, the Yucca Mountain Site in Nevada has been selected, but the process for licensing and deployment has been slowed by sociopolitical factors. The need to begin spent fuel recycle (P-T operations) to conserve the available repository space at Yucca Mountain and to enable sustained use of nuclear power has become apparent.

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Both mass and decay heat of the radioactive waste are significant factors in determining the total capacity of the Yucca Mountain Repository.[3] Removal of the heat generators from the waste that is placed into the repository can result in more compact storage and thus a more efficient utilization of the repository space. Figure 1 shows that decay heat from the TRU actinides is approximately constant and that it is the dominant heat source after ~100 years. During the first ~50 years, the dominant heat sources are the fission products, principally ^{137}Cs and ^{90}Sr, each with a half-life of approximately 30 years. Thus, attainment of the maximum repository benefit requires P-T of all transuranic actinides and managed storage of the fission product waste containing cesium and strontium for ≥ 100 years.

1.3. Scope

The multicycle P-T studies at ORNL began with an evaluation of the potential benefits that could be obtained by using existing and future thermal spectrum LWRs for the actinide transmutation.[4 5] A specific

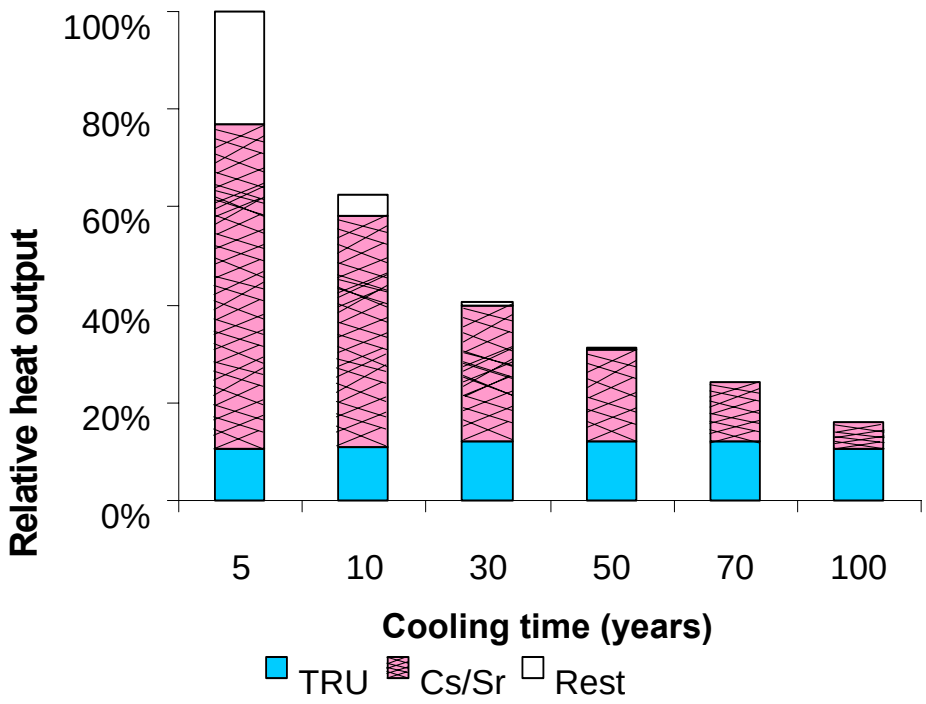

Fig 1. Heat generators in spent fuel.

goal of the studies was to evaluate the approach to equilibrium for each of the TRU actinide isotopes during the early P-T cycles and to interpret the specific transmutation pathway for each of the isotopes.

The effects of several options and constraints were evaluated during the evaluation phase of the study, and the most significant effects were found to be the age (decay time, "cooling time") of the spent fuel and the blending method used (if any). Also, the effect of using inert or fertile diluents for americium-curium targets and the effect of separating and storing the curium instead of recycling it were determined.[6]

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2. SCENARIO SELECTION FOR THE BASE-CASE EVALUATION USING THERMAL TRANSMUTATION IN LWRs

Efforts were made to select a realistic scenario that would apply to closing the fuel cycle, including full actinide recycle, in the United States during the next ~100 years. Prime consideration was given to cost minimization and to proliferation resistance. The scenario selected for evaluation using existing and new LWRs for the actinide transmutation is shown in Fig. 2**.**

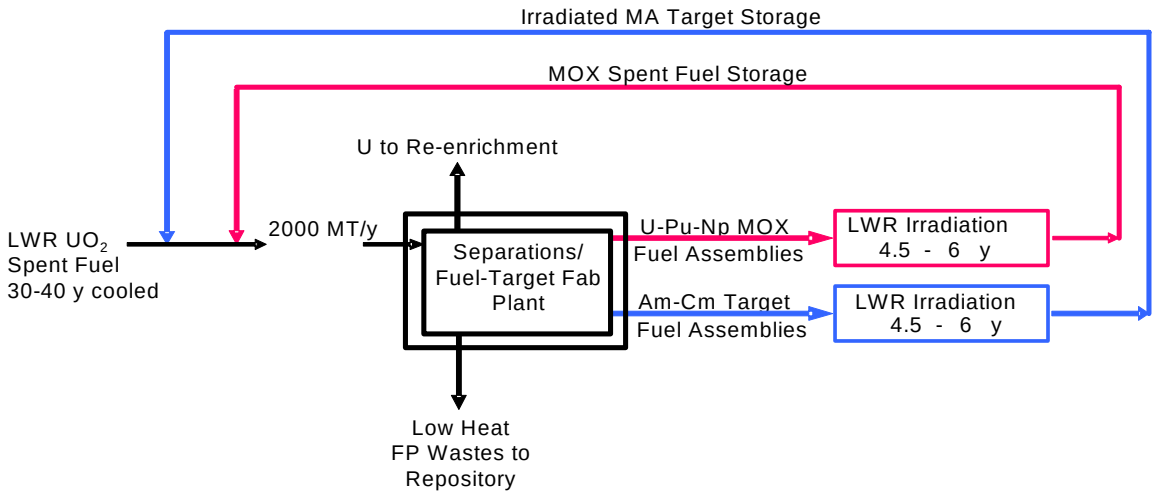


Fig. 2. P-T scenario using thermal transmutation in LWRs.

The optional conditions chosen for the scenario evaluated and the rationale for selection are described below. The options included (1) spent fuel age; (2) blending strategy for the recycled actinides with the actinides contained in LWR UO_2 spent fuel; (3) co-location and integration of facilities for spent fuel separation, recycle fuel/targets fabrication, and waste solidification; (4) processing capacity (plant size); (5) use of heterogeneous cores (Pu-Np and Am-Cm) for actinide transmutation; and (6) irradiation configuration.

2.1. Spent Fuel Age

Commercial plants in France and the United Kingdom typically process spent fuels that have aged for 5 to 10 years after removal from the reactor. Therefore, many P-T systems studies have been done previously with the assumption of 5 or 10 years aging. However, the very large inventory of heavy metal in spent fuel in the United States (~55,000 MT) contains significant amounts of older legacy fuel. A previous ORNL study[1] showed that if a separations/fuel fab plant was designed and built in the United States by 2020, with a capacity to process spent fuel at the current rate of generation (~2000 MT/year), and using the "oldest-fuel-first" methodology, the plant would, during its 30 to 50 year lifetime, never process fuel less than 39 years old. Figure 3, from that study, shows (1) the average age of the fuel processed, (2) the average burnup, and (3) the accumulated amount of fuel processed while using the oldest-fuel-first method.

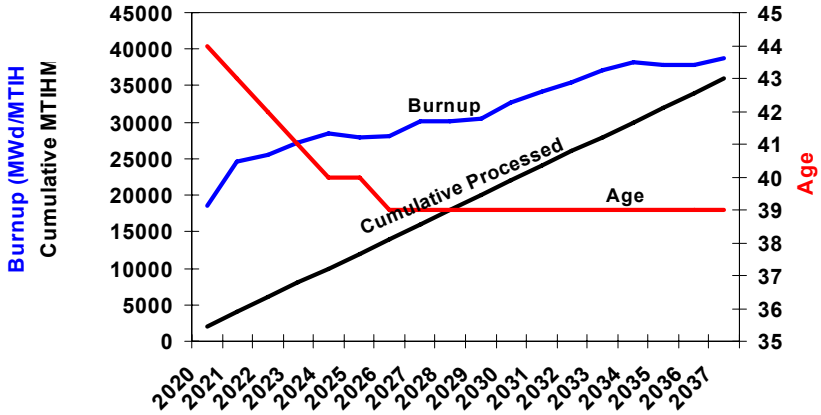


Fig. 3. Processing of 2000 MT/year of older-fuel-first.

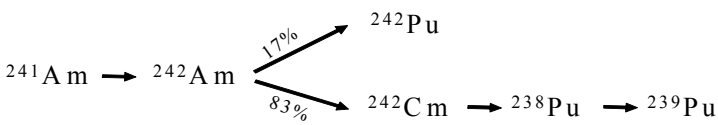
The benefits of processing the oldest fuel first are that there would be (1) lower radioactive emissions, (2) less radiation damage to equipment and instrumentation components and process fluids, and (3) less heat emission from stored waste. Thus, the plant investment and operating costs would be reduced. Moreover, the P-T performance would be improved significantly because the transmutation pathway would be altered to produce lighter plutonium isotopes (predominantly ^{238}Pu) rather than the heavier radionuclides, ^{242}Pu, ^{243}Am, and curium isotopes. The manner in which this occurs is described below.

In the chart of the nuclides (Fig. 4), the primary path toward production of heavier nuclides (curium) is through

$$^{241}\text{Pu} \rightarrow {}^{242}\text{Pu} \rightarrow {}^{243}\text{Am} \rightarrow {}^{244m}\text{Am} \rightarrow {}^{244}\text{Cm}$$

by means of neutron capture and beta decay reactions.

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, ~67% of the previously produced ^{244}Cm will decay to ^{240}Pu.

The conclusion is that much of the transmutation pathway is altered to produce lighter plutonium nuclides rather than the heavy curium nuclides.

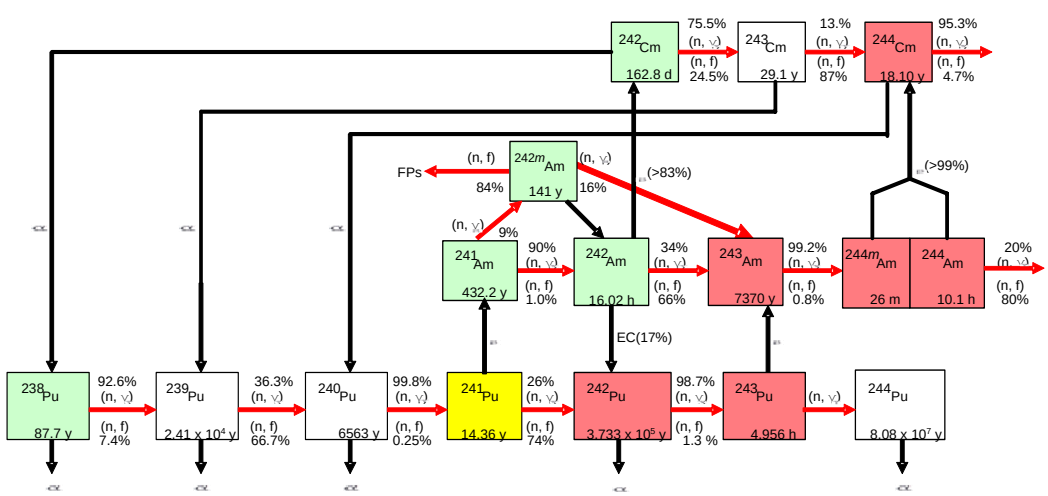


Fig. 4. Thermal spectrum nuclear reactions of interest.

2.2. Blending Strategy

In the evaluated scenario (Fig. 2), after disassembly and dissolution, the spent fuel components from recycled fuels are blended with those from LWR UO_2 spent fuels. The blended liquor is then processed to recover the actinide products and to fabricate new recycle fuel.

There is a basis of experience for blending in existing spent fuel processing plants (THORP, LaHague) where several types of spent fuel are processed routinely. For example, THORP processes stainless-steel-clad advanced gas reactor fuel, zircaloy-clad PWR spent fuel, and zircaloy-clad BWR spent fuel in the same equipment. This experience indicates that blending of the actinide components from different spent fuels [such as LWR UO_2 (LEU) spent fuel, recycled MOX spent fuels, and irradiated targets] can be done within the same plant, and that the extra costs of providing special separations plants for each type of spent fuel can be avoided.

The blending strategy, illustrated in Fig. 5, enables (1) dilution of the heavier plutonium isotopes contained in the MOX spent fuel with the lighter plutonium isotopes in the irradiated Am-Cm and LWR UO_2 (LEU) spent fuel and (2) maintenance of a sufficiently high fissile plutonium ($^{239}Pu + {}^{241}Pu$) concentration in the feed to the next cycle. The scenario assumed a constant 100 GWe power generation equivalent to a total of 2000 MT/year of LWR spent fuel. Thus, the recycled spent fuels were blended with a sufficient amount of LWR-UO_2 (LEU) spent fuel to prepare a total of 2000 MT/year of feed to the next P-T cycle, as illustrated in Table 1.

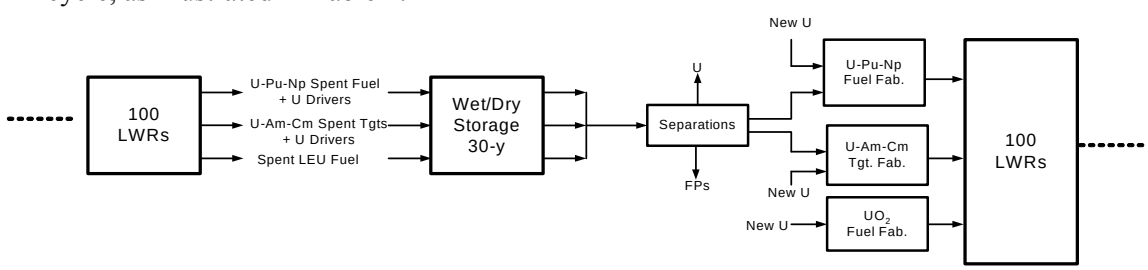


Fig. 5. Blending strategy.

Table 1. Actinide mass/isotopic compositions for second cycle feed

	Recycled MOX Spent Fuel	+	Recycled Irradiated Am-Cm Targets	+	"Fresh" LWR-UO_2 (LEU) Spent Fuel LWR-UO_2	=	2nd P-T Recycle 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		13.7		25.1
^{246}Cm, %	3.3		5.5		1.0		3.9
Total HM/MT/yr	204		24		1772		2000
	10.2%		1.2%		88.6%		100%

Previous studies at Argonne National Laboratory (ANL) and other institutions[8] have shown clearly that only one or two cycles of TRU actinide P-T is possible if blending with LWR UO_2 spent fuel is not done. This is because of the depletion of fissile isotopes and the accumulation of heavier, more highly radioactive actinides in the recycled materials. The early conclusion was that recycle using thermal spectrum irradiation is limited to one or two cycles ("limited recycle"). However, blending is feasible as described above and enables extended recycling through additional P-T cycles ("continuous recycling").

2.3. Plant Size (Capacity)

Existing plants have processing capabilities of 400 to 1200 MT/year of spent fuel, whereas the U.S. plant, when built, will need to process > 2000 MT/year to maintain equilibrium with the current generation rate. If nuclear power grows, as expected, even larger plant sizes will be required. In a previous ORNL study,[9] capital costs were shown to be essentially the same for small-scale plants as for larger-capacity installations because of the fixed infrastructure costs for a plant that processes highly radioactive materials. Thus, the data showed that the unit cost decreases exponentially with increasing capacity and reaches a broad near-minimum unit cost between 2000 and 8000 MT/year (Fig. 6). Further, a more recent study done for ORNL by NEXIA Solutions (U.K.) [10] and based on experience with the THORP plant, indicated that, if a 2000-MT/year plant is built in two 1000-MT/year modules or three 670-MT/year modules, the capital costs would be increased by 40% (for two modules) or 80% (for three modules). However, the increased cost may be worthwhile because there will likely be advantages in operation and maintenance flexibility to build the plant in multiple modules.

Operating cost data on existing plants are less reliable because the availability of spent fuel feed to those plants has been limited. This will not be the case for a U.S. plant because of the large inventory of spent fuel available for processing.

The conclusion is that a plant with an overall capacity of 2000 to 3000 MT/year is practical from a cost and operations feasibility standpoint.

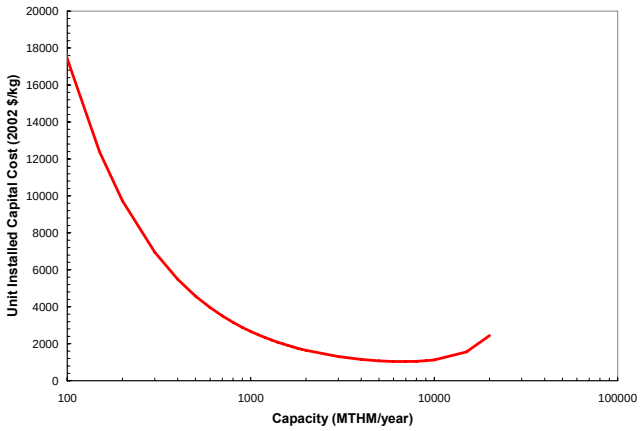


Fig. 6. Unit-installed cost vs plant capacity.

2.4. Co-Location and Integration of Separations and Fuel/Target Fabrication

The driving factors to co-locate and integrate the spent fuel separations and fuel/target fabrication operations within a physically protected facility (illustrated in Fig. 7) are cost reduction and maximization of proliferation resistance. Within the co-located configuration, the operations are integrated and excessive product storage and transportation are avoided. Thus, cost reduction is achieved. Moreover, all spent fuel separations are conducted within the physically protected facility, and the fissile material input and output are contained within large, easily accountable, fuel assembly modules. Effective monitoring of

waste materials and exiting personnel are the remaining links to ensure that no significant amount of fissile material leaves the plant covertly**.** Development, refinement, and implementation of effective monitoring technologies for waste materials and exiting personnel should be a high priority to provide the primary closure to safeguards requirements. Measurements of process streams, products, and wastes within the plant could become more of a process control function and a secondary safeguards requirement.

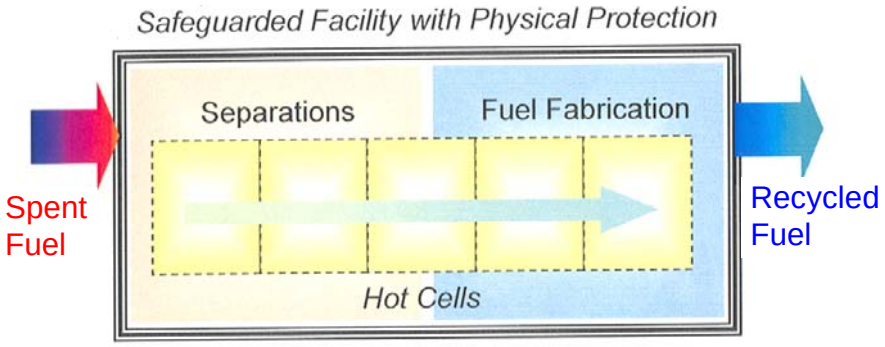


Fig. 7. Co-located and integrated separations – fuel fabrication plant.

2.5. Heterogeneous Actinide Recycling

Initial calculations indicated no appreciable difference in TRU actinide transmutation when using either homogenous or heterogeneous core irradiations. However, out-of-reactor factors are significantly different, and heterogeneous actinide recycling was selected for this study primarily to minimize the effort, time, and costs for fuel/target development and fabrication. This is achieved because U-Pu MOX fuel has already been developed, licensed, and industrially proven and can be conducted in glove-box-contained equipment. In contrast, fabrication of any fuel or target containing americium and/or curium requires shielded, remotely operated equipment, which is more expensive to operate and maintain. Moreover, the plutonium is present in much greater amounts (by a factor of ~10) than the americium and curium.

The conclusion is that if the americium and curium are combined with the plutonium, then the shielded, remotely operated fabrication facility becomes larger and more expensive to build, operate, and maintain.

In addition, efficient, industrially proven separations chemistry is more amenable to the heterogeneous separation of the predominantly tetravalent and hexavalent actinides (plutonium and neptunium) from the predominantly trivalent americium and curium and the predominantly trivalent lanthanide fission products. By taking advantage of these chemical properties, the plutonium and neptunium can be recovered, along with part of the uranium, in a single-step solvent extraction process similar to that already used successfully in existing industrial-scale operations. The priority of current research and development (R&D) can be focused on the more difficult trivalent actinide-lanthanide separation process to develop a robust process that can be used on an industrial scale.

In early studies, homogenous actinide recycling was selected, apparently, to provide added proliferation resistance. However, subsequent studies[11] have shown that the added resistance is minimal. Safeguards studies now indicate that other components (large amounts of uranium) are more effective for reducing the “attractiveness level.”

2.6. Irradiation Configuration

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 were kept constant during the multiple P-T cycles. In most of the calculations made in this study, the minor actinide (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 × 17 PWR fuel rod configuration, together with 216 standard "driver rods" containing UO_2 fuel enriched to 5.0 wt % ^{235}U (Fig. 8).

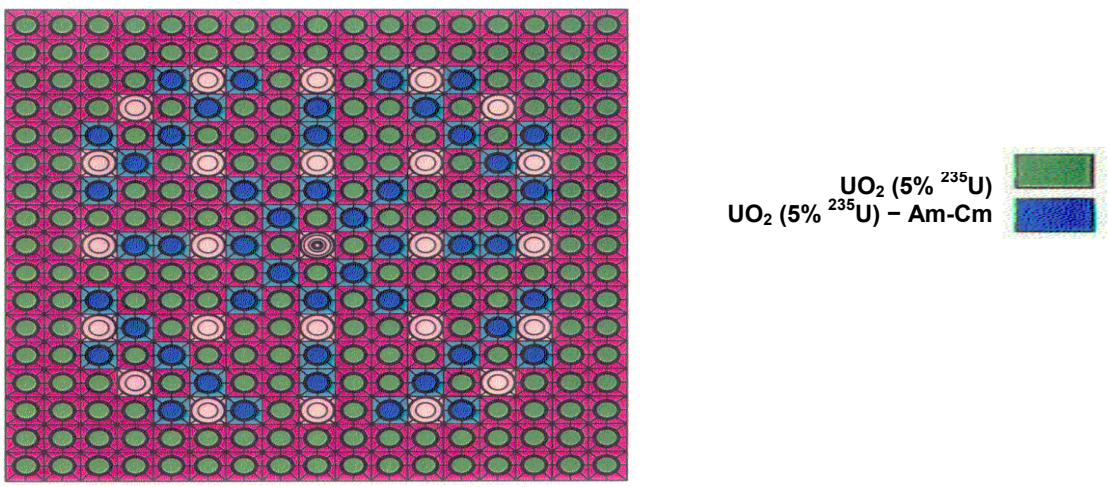


Fig. 8. Irradiation configuration.

Similarly, 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-MWt core, which contained 193 fuel assemblies. Detailed two dimensional neutronics depletion calculations were performed with the HELIOS code[12] using 47 neutron groups.

Special calculations of reactor core safety factors were made for full-power conditions to determine the effects of (1) beginning irradiation with TRU actinides recovered from either 5-year or 30-year decayed spent fuel; (2) fuel assembly loadings of TRU actinides of either 10% or 15%; (3) actinides including and not including curium; (4) matrices of depleted uranium, low-enriched uranium, or inert metal (zirconium); and (5) heterogeneous (Pu-Np and Am-Cm) or homogenous core loadings. The reactivity change (void coefficients) at beginning-of-cycle (BOC) and end-of-cycle (EOC) were determined at coolant void levels of 10% to 100%. Detailed results are given in Appendix A.

In summary, the results showed that negative void coefficients were found for all heterogeneous core configurations and for homogeneous cores with 10% TRU actinide loadings. The homogeneous cores with 15% TRU actinide loadings encountered positive void coefficients at BOC 90% and 100% coolant voiding conditions and at EOC 100% coolant voiding.

3. RESULTS FROM THE BASE-CASE EVALUATION USING THERMAL TRANSMUTATION IN LWRs

3.1. Base Case

The results of the base case (30-year decay, 35-year P-T cycles) evaluation are shown in Figs. 9 and 10. The results in Fig. 9 show that both the Pu-Np and the Am-Cm can be brought to near equilibrium, and that the production of heavy elements (Am and Cm) can be suppressed. Also, the scenario enables the fissile content of the blended plutonium product from each separation to remain sufficiently high (≥ 40%) for multiple P-T cycles to be achieved.

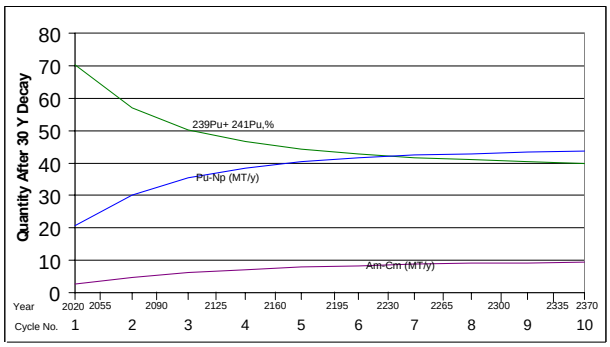


Fig. 9. TRU element inventories and fissile plutonium concentration in the feed to each P-T cycle.

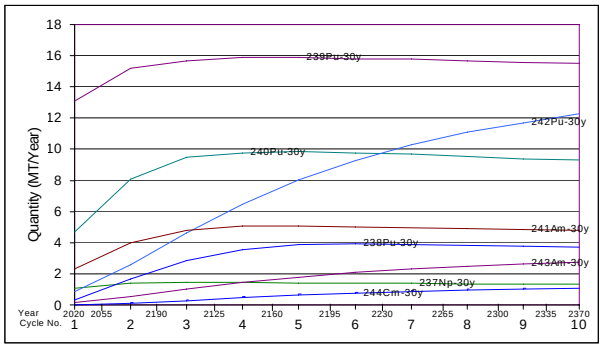


Fig. 10. TRU actinide inventories in the feed to each P-T cycle.

Figure 10 shows that, after ten 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. This is because of the 17% branching decay of ^{242}Am (16-h half-life) to ^{242}Pu and subsequent neutron capture, as described earlier. Overall, the production rates of all of the actinide elements are near equilibrium.

The conclusion is that the overall time span of ~350 years for ten P-T cycles is indicative of a sustainable strategy of continuous actinide recycle with only process losses going into the repository.

3.2. 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 11 illustrates the production rates of the heavier radionuclides, ^{242}Pu, ^{243}Am, and ^{244}Cm during each P-T cycle. 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 5-year decay periods and cycle lengths. This is because the 5-year decay period allows only 22% decay of ^{241}Pu to ^{241}Am, with subsequent transmutation primarily to ^{238}Pu, whereas the 30-year decay period allows 77% decay of ^{241}Pu to ^{241}Am. Also, of the ^{244}Cm that is produced, the 5-year decay period allows only 17% decay of the ^{244}Cm to ^{240}Pu, whereas the 30-year decay period allows 68% decay of the ^{244}Cm.

When plotted against actual time beginning with the start of the recycling scenario (Fig. 12), the differences are more prolific and indicate the difficulty that would be encountered in the near term (~50 to 100 years after start of recycling). During that time, multiple tons per year of ^{244}Cm would need to be

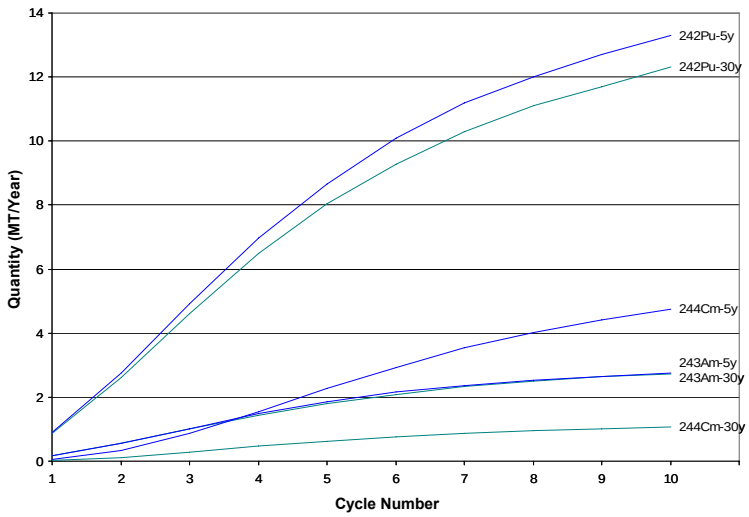


Fig. 11. Comparison of 5-year decay and 30-year decay production rates for each cycle.

handled if the 5-year decay fuel were processed and expensive separations steps (to separate curium from americium) and storage provisions (for curium) would be needed. Apparently, this effect has caused a dilemma in countries such as France, where recycling is already in progress using a ~5-year decay period. The apparent dilemma has led to a decision to limit plutonium recycle in LWR-MOX fuel to one cycle.

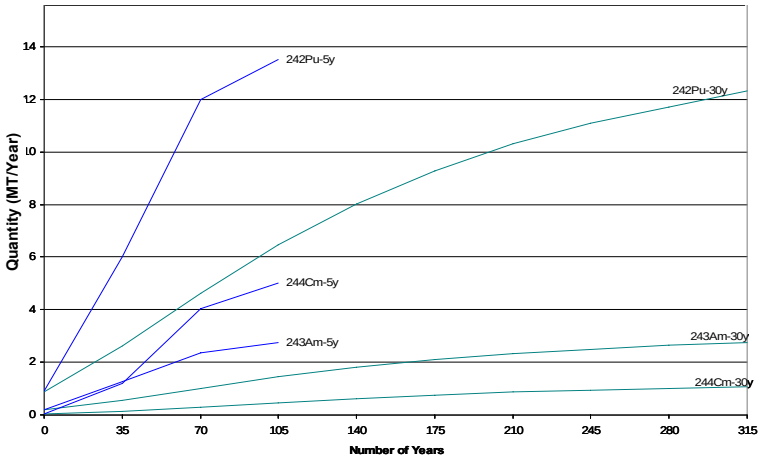


Fig. 12. Comparison of 5-year decay and 30-year decay production rates with time

3.3. Studies on the Effect of Removing Curium from the Americium Target

It is well recognized that curium isotopes emit significantly greater amounts of neutrons than are emitted by plutonium, neptunium, and americium. Therefore, fabrication of targets or fuel containing curium must be done in shielded facilities. Also, most studies recognize that the americium in spent fuel contains significant amounts of gamma-emitting isotopes (^{241}Am and ^{239}Np, the 2.35-d–half-life daughter of ^{243}Am). This factor will make the fabrication of recycle targets or fuel containing americium, with or without curium, a process that must be performed in shielded facilities. Nevertheless, because multiple recycling of actinides produces increasing amounts of curium isotopes, many studies have recommended separation and storage of curium, rather than recycling.[10,13,14]

Thus, a systems study[6] was performed to examine the effects of multiple recycling with americium and curium, or with only americium, in the recycle targets. In both cases the plutonium and neptunium were mixed with depleted uranium and were recycled as MOX fuel while the americium and curium were, or only americium was, irradiated in the oxide form with either LEU oxide (5% ^{235}U) or zirconium as the diluent.

The results from five P-T cycles in which either a 5-year or a 30-year decay period was used for the irradiated fuel, and targets in each cycle, are shown in Table 2. These results show that recycling with or without curium does not significantly affect the composition of the other actinide elements and key isotopes. The amounts of curium produced in each cycle are reduced when curium is removed after each cycle. This is an effect of the purging of one of the products instead of recycling. However, when the cumulative amount of curium removed (without regard to decay) is considered, the overall production of curium is much greater. The overall amount of curium produced is less when curium is recycled because a significant amount of the curium is transmuted to fission products and heavier elements, such as berkelium and californium. Fortunately, the isotopes of berkelium and californium are produced in relatively small amounts and most have relatively short half-lives; thus, the accumulation of the transcurium element isotopes and their radioactive emissions are relatively insignificant in the recycled actinide mix, especially when longer decay times are used between each P-T cycle and the one that follows.

Table 2. Curium removal effect using low-enriched uranium (LEU) diluent

P-T Cycle	5-y Decay					30-y Decay				
	1	2	3	4	5	1	2	3	4	5
Year	2020-2030	2030-2040	2040-2050	2050-2060	2060-2070	2020-2055	2055-2090	2090-2125	2125-2160	2160-2195
Total Pu, MT/y										
Am-Cm in LEU	21.3	34.5	43.2	49.0	53.0	19.6	31.8	40.0	45.6	49.5
Am in LEU	21.3	34.5	43.2	48.9	52.8	19.6	31.8	40.0	45.6	49.4
^{242}Pu, MT/y										
Am-Cm in LEU	0.89	2.36	4.19	6.10	7.92	0.89	2.30	4.02	5.81	7.49
Am in LEU	0.89	2.36	4.19	6.09	7.89	0.89	2.30	4.02	5.80	7.48
^{241}Am, MT/y										
Am-Cm in LEU	0.73	1.56	2.13	2.43	2.57	2.31	4.37	5.78	6.61	7.07
Am in LEU	0.73	1.57	2.14	2.47	2.62	2.31	4.37	5.78	6.63	7.09
^{243}Am, MT/y										
Am-Cm in LEU	0.18	0.51	0.93	1.37	1.77	0.18	0.52	0.95	1.38	1.77
Am in LEU	0.18	0.51	0.94	1.38	1.81	0.18	0.52	0.95	1.38	1.78
^{244}Cm, kg/y										
Am-Cm in LEU	44	288	721	1291	1933	17	110	252	412	571
Am in LEU **(*)**	(44)	(262)	(575)	(915)	(1245)	(17)	(107)	(231)	(365)	(493)
Total Cm, kg/y										
Am-Cm in LEU	47	340	865	1560	2348	20	165	395	668	950
Am in LEU **(*)**	(47)	(307)	(677)	(1079)	(1458)	(20)	(158)	(346)	(546)	(733)

(*) Parenthesis indicate the amount of ^{244}Cm or Total Cm removed to storage at the end of each P-T cycle.

The results of this study indicate that curium recycle produces less curium overall. Shielding for target fabrication is required with or without curium, even though curium will require special neutron shielding.

The conclusion is that when an economic comparison is made, the significant costs of separating, storing, and disposing of curium will likely make that option more difficult and expensive than curium recycling.

3.4. Studies on the Effect of Inert Matrix for Americium-Curium Irradiation

During the spent fuel storage part of the recycle process, significant changes occur in the actinide composition as radioactive decay occurs. Specifically, the most significant change is the decay of ~14-year half-life ^{241}Pu to produce ^{241}Am. When the subsequent irradiation of the actinides occurs, the ^{241}Am transmutes predominantly to short-lived ^{242}Cm (162-d half-life). This produces a relatively large

amount of helium from the ^{242}Cm alpha decay and introduces the possibility of over-pressurization of the transmutation target rods. Resolution of this potential problem must come from the selection of diluent and from the form of the target matrix as well as from the pellet and/or rod configuration and design.

There are both advantages and disadvantages in selecting the diluent such that the target matrix is either inert or fertile. A uranium (fertile) diluent would provide compatibility with the existing components of spent fuel but may not accommodate heat transfer and helium-containment requirements. In contrast, an inert diluent could provide better heat transfer and helium containment properties but would introduce a new component into the spent fuel mix and would require additional separation and disposal requirements. The analysis and selection of the optimal diluent must be made during the transmutation target fabrication R&D.

A comparative transmutation analysis using LWR irradiation was made to see if the choice of diluent for an Am-Cm oxide target would make a difference in other actinide compositions during multiple recycling. The diluents compared were low-enriched uranium oxide (5% ^{235}U) and zirconium metal. The results of five P-T cycles are shown in Table 3 for cases where the decay storage period was either 5 years or 30 years. These results showed that with the inert matrix, about 5 to 10% greater production of curium occurred; however, in general, the results showed no greatly significant difference in the production of plutonium, americium, or curium isotopes.

Table 3. Effect of low-enriched uranium (LEU) vs inert material (IM) as the matrix for Am-Cm targets

P-T Cycle	5-y Decay					30-y Decay				
	1	2	3	4	5	1	2	3	4	5
Year	2020-2030	2030-2040	2040-2050	2050-2060	2060-2070	2020-2055	2055-2090	2090-2125	2125-2160	2160-2195
Total Pu, MT/y										
Am-Cm in LEU	21.3	34.5	43.2	49.0	53.0	19.6	31.8	40.0	45.6	49.5
Am-Cm in IMF	21.3	34.4	42.8	48.2	51.8	19.6	31.6	39.2	44.2	47.5
^{242}Pu, MT/y										
Am-Cm in LEU	0.89	2.36	4.19	6.10	7.92	0.89	2.30	4.02	5.81	7.49
Am-Cm in IMF	0.89	2.37	4.22	6.17	8.05	0.89	2.33	4.07	5.86	7.54
^{241}Am, MT/y										
Am-Cm in LEU	0.73	1.56	2.13	2.43	2.57	2.31	4.37	5.78	6.61	7.07
Am-Cm in IMF	0.73	1.49	1.98	2.24	2.35	2.31	4.14	5.33	6.00	6.34
^{243}Am, MT/y										
Am-Cm in LEU	0.18	0.51	0.93	1.37	1.77	0.18	0.52	0.95	1.38	1.77
Am-Cm in IMF	0.18	0.51	0.91	1.29	1.62	0.18	0.55	0.96	1.37	1.72
^{244}Cm, kg/y										
Am-Cm in LEU	44	288	721	1291	1933	17	110	252	412	571
Am-Cm in IMF	44	304	760	1340	1960	17	126	286	459	622
Total Cm, kg/y										
Am-Cm in LEU	47	340	865	1560	2348	20	165	395	668	950
Am-Cm in IMF	47	355	897	1595	2355	20	182	430	715	1001

The conclusion is that the choice of diluent will not affect transmutation yields significantly and can be made entirely from the standpoint of target design and performance.

4. COMPARISON OF ACTINIDE P-T IN LWRs AND ABRs

In the previous studies, multicycle P-T in existing and future thermal spectrum LWRs was evaluated to determine the approach to equilibrium mass levels of TRU actinides.[5] The use of compact ABRs, using fast spectrum irradiation at low conversion ratios (< 0.5) is now planned. Previous studies indicated that ABRs are required to consume the TRU actinides.[7] 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.[15]

4.1. P-T Scenarios Evaluated

Figure 13 shows the P-T scenario evaluated for fast burner reactor transmutation. It is similar to the scenario previously evaluated for thermal reactor transmutation (Fig. 2). The TRU actinide feed material for both cases was obtained from LWR UO_2 (LEU) spent fuel that had been irradiated for ~45 GWd/MT and that 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. 13), a homogenous core with uranium-TRU-zirconium metal fuel was used. Conversion ratios of 0.50 and 0.25 were evaluated.

4.2. Fast Reactor Model Irradiation Configuration

The lattice representing the seven-ring, compact, sodium-cooled, low-conversion-ratio fast reactor design described by Argonne National Laboratory [16] was modeled using the HELIOS code[12], 112 neutron groups, and the fast reactor cross-section library. In the reactor lattice model, each fuel assembly 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 MWt, using a capacity factor of 85%. Total irradiation time was ~3 years. Three fuel rod diameters (0.67, 0.62, and 0.59 cm) were used to provide conversion ratios of 0.50, 0.35, and 0.25, respectively.

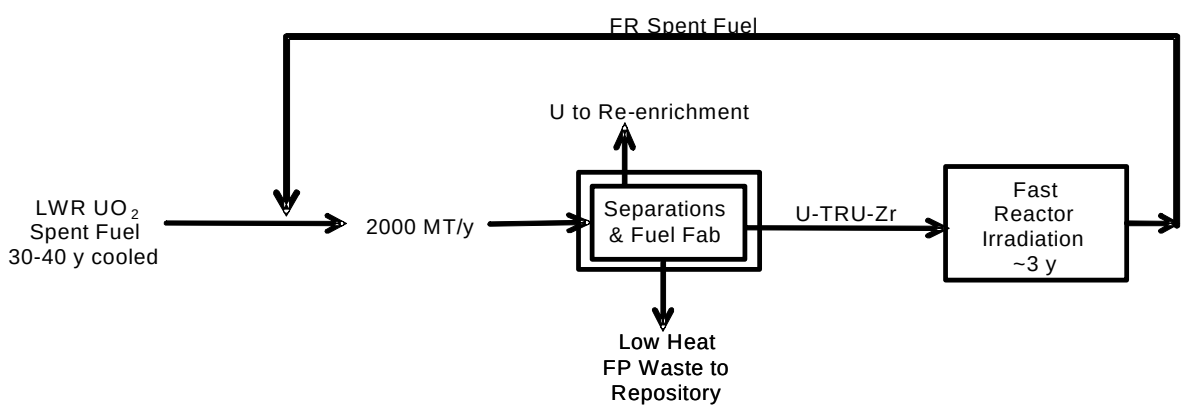


Fig. 13. Fast burner reactor transmutation scenario.

4.3. First Cycle Results

TRU actinide compositions before and after irradiation for each of the fast and thermal spectrum reactors are shown in Table 4. 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.

Table 4. TRU actinide compositions before and after irradiation

	1st 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	0.888 (4.5%)	0.880 (5.4%)	0.870 (6.1%)	1.83 (14.1%)
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	0.177	0.190	0.191	0.40
Total Am, MT/y	2.49	1.67	1.547	0.88
^{244}Cm, MT/y	0.0166	0.057	0.063	0.340
Total Cm, MT/y	0.0198	0.181	0.205	0.559
Total TRUs, MT/y	23.2	18.9	16.7	14.8
Sum ^{241}Pu + ^{241}Am, MT/y	3.01	1.03	1.91	2.53

The data in Table 4 were used to calculate and compare burnup percentages (net destruction and net production), as shown in Table 5**.** The comparisons showed that the burnup rates of ^{239}Pu, ^{240}Pu, ^{237}Np, and ^{241}Am achieved in LWRs are greater than in ABRs. However, the net production (negative burnup in Table 5) of ^{241}Pu, ^{242}Pu, ^{243}Am, and ^{244}Cm is significantly greater in the LWRs. A net production of the ^{238}Pu isotope occurs in both reactors by transmutation of ^{237}Np and ^{241}Am, but the accumulation of ^{238}Pu is greater in the LWRs.

Table 5. Burnup[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%)
Total TRUs	18.5%	28.0%	36.2%
Sum ^{241}Pu + ^{241}Am	32.6%	36.5%	15.9%

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

4.4. Interpretation of First-Cycle 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 accumulation (net production) of TRU actinides does not depend only on fission cross sections. Table 6 shows that (1) accumulation of each actinide isotope depends on the differences in formation vs destruction rates; (2) the formation rates do not depend at all on fission reactions; and, (3) fission is only one component of the destruction rates. Except in a few cases where the radionuclides are produced predominantly by decay (for example, ^{241}Am is produced only by decay of ^{241}Pu), most of the radionuclide formation rates are dependent on the neutron capture rate of the precursor isotopes. The destruction rate of each actinide is dependent on the sum of neutron capture, fission, and decay of that particular radionuclide.

Table 6. Factors affecting accumulation (net production) of each actinide isotope

Formation:	From decay of parent isotope or neutron capture in precursor isotope
Destruction:	Sum of decay, neutron capture and fission of the product isotope
Formation minus Destruction = Accumulation (net production) of the product isotope	

It is well known from comparative cross-section data for TRU actinides that both neutron capture and fission cross sections for most isotopes are significantly decreased in fast spectrum irradiations vs 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 an order of magnitude higher than in LWRs, thus mitigating the lower cross sections in determining neutron reaction rates.

The factors described in Table 6 which affect formation and destruction were calculated for each actinide isotope by the HELIOS code and the relative rates in ABRs in comparison to those in LWRs are shown in Table 7**.** These relative rates show that most of the formation and destruction reaction rates per unit of mass of each isotope are lower in ABRs (FR/0.25CR) than in LWRs. Only the formation rate of ^{239}Pu and the destruction rates of ^{238}U and ^{238}Pu are greater in the ABRs (FR/0.25CR). The formation rate of ^{239}Pu 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 ABRs (FR/0.25CR). The destruction rate of ^{240}Pu is decreased by a larger factor than the formation rate decrease factor (12.2 vs 6.6), again resulting in a gain in the accumulation in ABRs (FR/0.25CR) relative to that in LWRs.

Table 7. Relative TRU isotope formation and destruction rates in ABRs(FR/0.5CR) in comparison to those in LWRs

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] Rates are per unit mass of each isotope.

[c] "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.

[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 ABRs (FR/0.25CR) relative to that in LWRs, 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.

The conclusion is that transmutation of plutonium in a fast spectrum is optimum because the production of ^{241}Pu (and its decay daughter, ^{241}Am) as well as the sequentially produced ^{242}Pu, ^{243}Am, and ^{244}Cm are minimized.

The ^{238}Pu isotope is produced by transmutation of the minor actinides, ^{237}Np and ^{241}Am, in both fast and thermal spectrum irradiations. Table 7 shows that the destruction rate of ^{238}Pu is increased and that the formation rates are decreased in ABRs(FR/0.25CR) relative to those in LWRs; thus, a smaller accumulation of ^{238}Pu is achieved in ABRs(FR/0.25CR). However, the production of ^{238}Pu has two positive attributes. First, upon recycle, the ^{238}Pu either fissions directly or is converted to more fissile ^{239}Pu, thus enhancing the fissile content of the recycled actinides. Second, when blended with the plutonium from LWR UO_2 spent fuel, the increased ^{238}Pu content decreases the "attractiveness" of the blended plutonium for use in weapons production because of the increased radioactivity and decay heat emission.

When the 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 ABRs(FR/0.25CR) than in LWRs, as shown in Table 7**.** Thus hybrid cases evaluated in which the plutonium or the plutonium-neptunium combination are transmuted in ABRs (FR/0.25CR) and the remaining minor actinides (Np-Am-Cm or Am-Cm) are transmuted in LWRs. Very little difference occurred in the results from the two cases, indicating that the neptunium could be included with either the plutonium or with the other minor actinides. In either case, the production of both ^{241}Pu and its decay daughter, ^{241}Am, are suppressed effectively. (The ^{241}Pu-^{241}Am combination is a major contributor to integrated decay heat (IDH) if the TRU actinides are sent to a repository.) Tables 8 and 9 compare the results shown previously in Tables 4 and 5 with the "optimum" hybrid case and indicate that the burnup of the ^{241}Pu + ^{241}Am combination is increased by a factor of ~2 in comparison to the burnup when using homogeneous cores in ABRs. The hybrid case provided 63.8% burnup, compared to 36.5% in ABRs(FR/0.25CR) and only 15.9% in LWRs.

Table 8. Comparison of hybrid case TRU actinide compositions before and after irradiation

	1st 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	Hybrid Pu-Np in FR/0.25 CR Am-Cm in LWR @ discharge
^{237}Np, MT/y	1.097	0.712	0.645	0.44	0.73
^{238}Pu, MT/y	0.302 (1.5%)	0.728 (4/5%)	0.747 (5.2%)	1.57 (12.1%)	1.32 (8.4%)
^{239}Pu, MT/y	13.05 (66.6%)	9.56 (58.7%)	7.68 (53.6%)	3.53 (27.2%)	8.15 (51.8%)
^{240}Pu, MT/y	4.66 (23.8%)	4.51 (27.7%)	4.41 (30.8%)	3.97 (30.6%)	4.55 (28.9%)
^{241}Pu, MT/y	0.703 (2.6%)	0.620 (3.8%)	0.622 (4.3%)	2.06 (15.9%)	0.69 (4.4%)
^{242}Pu, MT/y	0.888 (4.5%)	0.880 (5.4%)	0.870 (6.1%)	1.83 (14.1%)	1.01 (6.4%)
Total Pu, MT/y	19.60	16.30	14.33	12.96	15.72
^{241}Am, MT/y	2.31	1.41	1.29	0.47	.40
^{243}Am, MT/y	0.177	0.190	0.191	0.40	0.18
Total Am, MT/y	2.49	1.67	1.547	0.88	0.59
^{244}Cm, MT/y	0.0166	0.057	0.063	0.340	0.142
Total Cm, MT/y	0.0198	0.181	0.205	0.559	0.298
Total TRUs, MT/y	23.2	18.9	16.7	14.8	17.3
Sum ^{241}Pu + ^{241}Am, MT/y	3.01	1.03	1.91	2.53	1.09

Table 9. Comparison of hybrid case burnup percentages
(Values in parenthesis are net production)

	FR/0.50 CR	FR/0.25CR	LWR	Hybrid Pu-Np in FR/0.25 CR Am-Cm in LWR
^{239}Pu	27%	41%	73%	38%
^{240}Pu	3%	5%	15%	2%
^{241}Pu	12%	12%	(193%)	2%
^{242}Pu	1%	2%	(106%)	(14%)
^{243}Am	(7%)	(8%)	(126%)	(2%)
^{244}Cm	(243%)	(280%)	(1950%)	(755%)
^{237}Np	35%	41%	60%	33%
^{241}Am	39%	44%	80%	83%
^{238}Pu	(141%)	(147%)	(420%)	(337%)
Total TRUs	18.5%	28.0%	36.2%	25.4%
Sum ^{241}Pu + ^{241}Am	32.6%	36.5%	15.9%	63.8%

The conclusion is that the transmutation of minor actinides, especially americium and curium, are optimum in a thermal spectrum reactor (an LWR), while the transmutation of plutonium or plutonium plus neptunium is optimum in a fast-spectrum reactor.

4.5. Multiple P-T Cycle Comparisons

The baseline case was the P-T scenario shown in Fig. 2, in which a heterogeneous transmutation (U-Pu-Np MOX and U-Am-Cm targets) was made in LWRs and the spent MOX fuel and irradiated Am-Cm targets were allowed to decay for 30 years before recycle. Evaluation cases were made for comparison using the P-T scenario shown in Fig. 13 with a homogeneous transmutation (U-TRU-Zr) alloy in fast burner reactors that had a 0.25 conversion ratio (FR/0.25CR).

In the first case evaluated, the recycled fuel was allowed to decay for 30 years before the next cycle; in the next case, the irradiated recycle fuel was allowed to decay for only 5 years. Results from these two cases were similar because the amount of ^{241}Pu produced in the fast reactor transmutation was sufficiently low, such that the mass difference in decay of ^{241}Pu was not significantly greater during the longer decay periods. Thus, the shorter decay period can be used, and the amount of recycle fuel inventory in storage can be reduced significantly. Similarly, the "optimum" hybrid case, in which a heterogeneous transmutation (U-Pu-Np MOX in FR/0.25CR reactors and Am-Cm targets in LWRs) is used, can utilize the shorter (5-year) decay periods because neither the irradiated recycle MOX fuel nor the irradiated Am-Cm targets contain a relatively large mass of ^{241}Pu.

Multicycle calculations were made to evaluate the approach to equilibrium for (1) the baseline case using heterogeneous cores in LWRs; (2) the comparative case using homogenous cores in ABRs (FR/0.25CR); and (3) the "optimum" hybrid case using heterogeneous cores (U-Pu-Np MOX in FR/0.25CR ABRs and Am-Cm targets in LWRs). Detailed actinide compositions in the feed and spent fuel/irradiated targets for each P-T cycle are given in Appendices B, C, and D. The approaches to equilibrium for each case are compared in Figures 14, 15, 16, and 17**,** which respectively show for the total recycle from each P-T cycle, the mass (metric tons per year from ~2000 MT/year of spent fuel processed) of plutonium isotopes (Fig. 14); the mass of heat generator isotopes (Fig. 15); the megawatts per year of integral decay heat (Fig. 16); and the mass (metric tons per year) of total TRU actinides, as well as each actinide element (Fig. 17). In each of the sets of charts, the baseline all-thermal-spectrum transmutation case is shown in left-side-chart, the all-fast-spectrum transmutation case is shown in the center chart and the "optimum" hybrid case is shown in the right-side chart.

Figure 14 shows several significant differences in thermal spectrum and fast spectrum transmutations of plutonium isotopes. In the thermal spectrum, the ^{239}Pu and ^{240}Pu approach equilibrium more quickly and at a lower mass level. The mass level of ^{241}Pu is also suppressed in the thermal spectrum irradiation case, but this is largely due to decay of the ^{241}Pu (14.3-year half-life) to ^{241}Am during the 30-year decay periods. The heavier plutonium isotope, ^{242}Pu, continues to grow to higher mass levels. Both the homogenous fast spectrum irradiation case and the heterogeneous hybrid case in which the Pu-Np is irradiated in the fast spectrum ABRs show similar enhancement of ^{239}Pu and ^{240}Pu growth to increasing mass levels throughout the five P-T cycles. The hybrid case shows a slightly higher mass level due to the increased amount of ^{238}Pu produced by ^{241}Am in the thermal target irradiation, followed by conversion of the ^{238} Pu to ^{239}Pu by neutron capture.

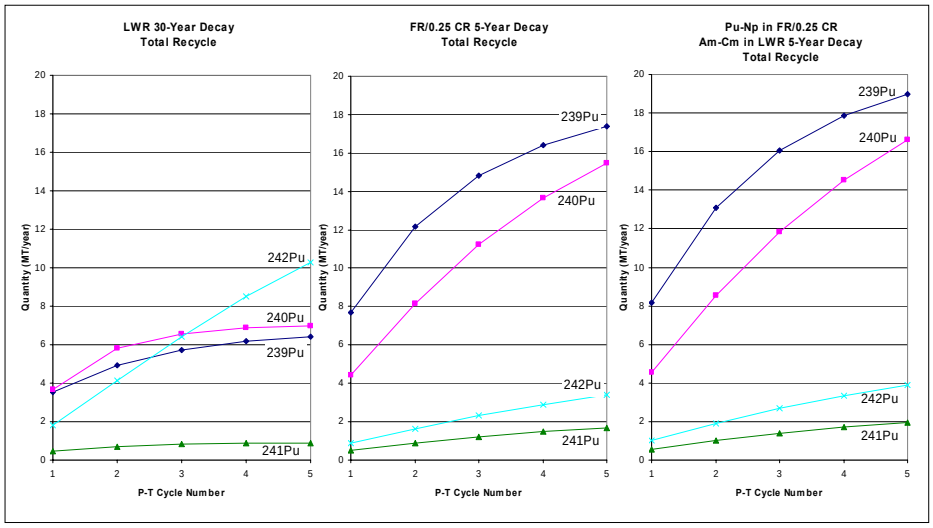


Fig. 14. Comparison of plutonium isotope inventories in the total recycle from each cycle during multiple P-T cycles.

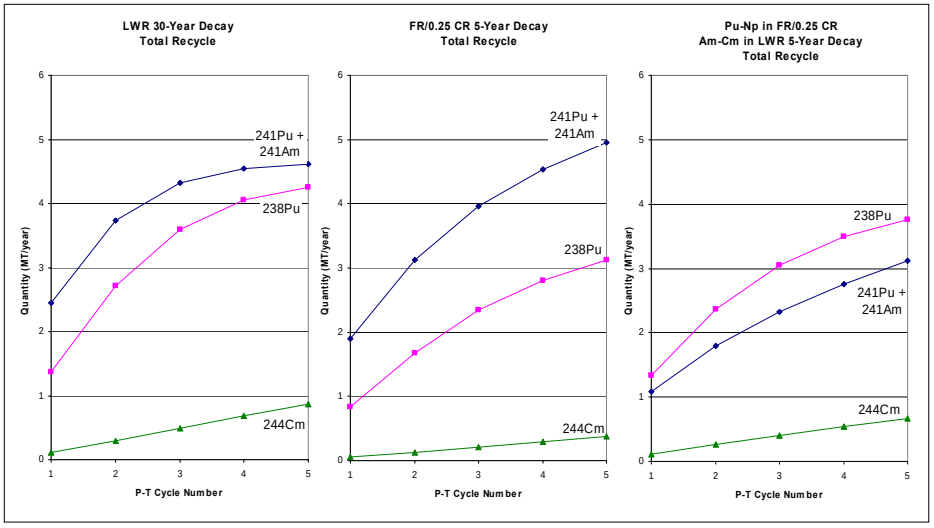


Fig. 15. Comparison of heat generator isotope inventories in the total recycle from each cycle during multiple P-T cycles.

Figure 15 compares the behavior of several "heat generator" isotopes in the thermal, fast, and hybrid irradiations. In the thermal spectrum case, the ^{241}Pu-^{241}Am pair reaches an equilibrium level of ~4.5 MT/year due to a combination of higher burnup of ^{241}Am and continued production of ^{241}Pu and decay to ^{241}Am. The ^{241}Pu-^{241}Am pair continues to grow to 5+MT/year in the all-fast-spectrum irradiation because of the smaller burnup, primarily of ^{241}Am. In the optimum "hybrid" case, the growth of the ^{241}Pu-^{241}Am pair is significantly suppressed because of the higher burnup of ^{241}Am in the thermal irradiation of Am-Cm targets and the minimization of ^{241}Pu production in the fast spectrum irradiation of the U-Pu-Np MOX fuel.

Production of ^{238}Pu from both ^{237}Np and ^{241}Am occurs in all of these cases. However, the ^{238}Pu produced is destroyed faster in the fast spectrum irradiation. Production of ^{244}Cm is suppressed in all three cases.

Figure 16 continues comparison of the heat generators in terms of integral decay heat (IDH) during hypothetical long-term storage in a repository for 1250 years, using the method of calculations described in ANL-AFCI-164.[17] The IDH is a characteristic of recycled spent fuel that has been used to indicate the "Yucca Mountain Repository Benefit Factor" if the assumption is made that the recycled spent fuel is sent directly to the Yucca Mountain Repository instead of being recycled. The approach to equilibrium occurs more quickly in the all-thermal-spectrum irradiation case, but the heat levels after five cycles are not greatly different in the three cases, even though the ^{241}Pu-^{241}Am pair content is suppressed in the hybrid case. Examination of the major heat-generating actinide elements shows a marked contrast in plutonium and americium generators. (Curium contribution is insignificant in comparison in all three cases.) In the all-thermal-spectrum case, heat generation comes almost equally from plutonium and americium, but in

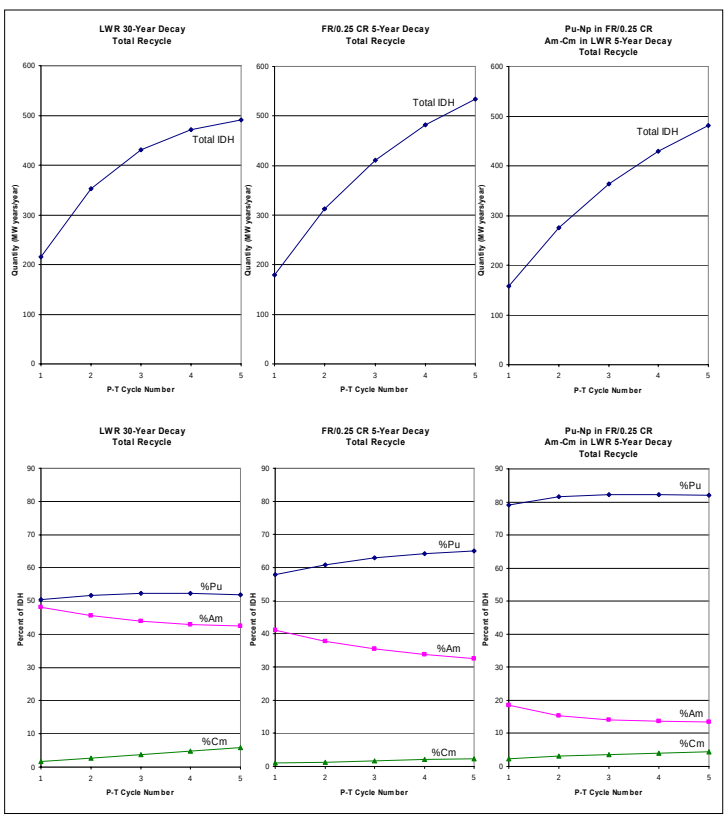


Fig. 16. Comparison of integral decay heat from total recycle inventories from each cycle during multiple P-T cycles.

the all-fast-spectrum and the hybrid cases, the plutonium isotopes are the major generators, largely due to increased contributions from ^{239}Pu and ^{240}Pu. In all cases, ^{238}Pu is a major contributor. In the hybrid case, heat generation from the ^{241}Pu-^{241}Am pair is significantly suppressed, as described above.

Figure 17 compares the mass level approach to equilibrium of the individual actinide elements with the total. Since the all-fast spectrum and the hybrid cases have shorter cycle time periods (~10 years vs ~35 years for the all-thermal-spectrum case), the full equilibrium levels will be reached in ~50 years while the all-thermal case will have completed only between one and two cycles during the 50-year period.

The major conclusion is that similar results are obtained for all three cases during the five-cycle comparison.

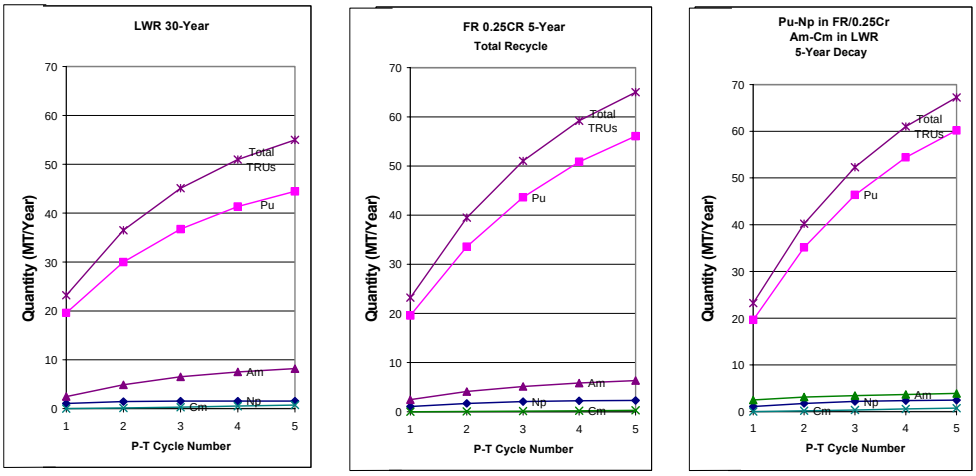


Fig. 17. Comparison of total TRU and element inventories in the total recycle from each cycle during multiple P-T cycles.

Figure 18 shows, for each of three cases, the number of reactors required to transmute the TRU actinides (~23 MT/year) produced in ~2000 MT/year of LWR UO_2 spent fuel. Considering the large number of 840 MWt ABRs required and the uncertain time required for full deployment, near-term P-T in the United States will likely need to utilize LWRs. Much earlier closing of the fuel cycle can be done effectively, using existing LWRs and the P-T scenario shown in Fig. 2.

The data obtained in this study showed that, because only part of the TRU actinides are destroyed in each P-T cycle in all of the cases evaluated (all-thermal-spectrum irradiations, all-fast-spectrum irradiations, or combinations of the two methods), the actinide inventory continues to grow even after recycling begins. Multiple P-T cycles are required to achieve a state of approximately "no net production," as illustrated in the generic diagram shown in Fig. 19**.** Moreover, the inventory that exists when recycling begins is not reduced unless the spent fuel is processed at a rate significantly greater than the rate at which it is being generated. Since the generation rate in the United States has risen to ~2200 MT/year and is expected to grow in the future, spent fuel processing plants that are, much larger than those that currently exist in France, the United Kingdom, Japan, and Russia, will be required.

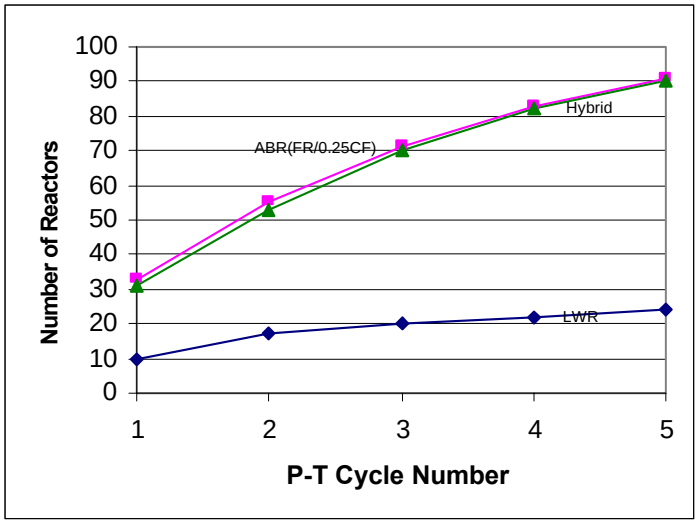


Fig. 18. Number of reactors required to transmute TRU actinides produced in 2000 MT/year of LWR UO_2 spent fuel (~100 LWRs).

The conclusion is that the most effective way to stop the inventory growth is to start recycling as soon as possible.

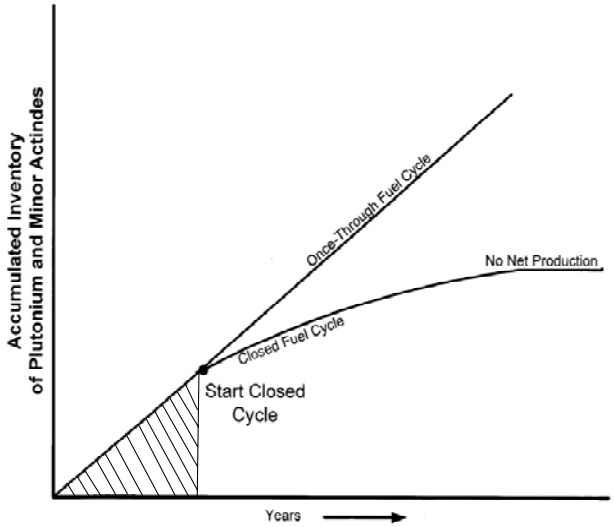


Fig. 19. Generic diagram of inventory accumulation rate when recycling begins at a rate equal to the rate of inventory generation.

5. SUMMARY AND CONCLUSIONS

Efforts were made to evaluate realistic scenarios that would apply to closing the fuel cycle in the United States, including full actinide recycle, during the next ~100 years. Existing conditions in the accumulation of spent fuel in the United States were recognized constraints. Variable conditions were chosen to

minimize costs and to provide essential proliferation resistance. For the scenarios evaluated, the following observations and conclusions were made.

- The United States has accumulated more than 55,000 MT of heavy metal in spent fuel and is generating ~2200 MT/year. The legacy fuel must be processed, and the actinides must be recycled to minimize the number of high-level waste repositories required in the future.

- Processing the oldest fuel first will enable numerous technical, environmental, and economic advantages. The spent fuel will have decayed more than 30 years, and many radionuclides will have decayed significantly prior to processing.

- Utilization of a blending strategy in which recycled actinides are blended with low-enriched uranium spent fuel at the head-end of the separations plant will provide a sufficiently high fissile content for subsequent recycle of the actinide mix and will enable continuous, multicycle operation of LWRs, ABRs, or combinations of the two types of reactors.

- Utilization of large spent fuel processing (separations and fuel/target fabrication) facilities with overall capacities of 2000 to 3000 MT/year is practical and provides the lowest unit cost for processing.

- Utilization of large co-located and integrated separations and fuel/target fabrications operations located within a physically protected facility will provide significant cost reduction and maximized proliferation resistance.

- Utilization of heterogeneous actinide recycling will provide (1) cost reduction in separations processes, fuel development, and fuel/target fabrication facilities and operation; (2) flexibility of P-T deployment; and (3) improved technical performance. Proliferation risk will not be increased.

The results of the study provided the following conclusions:

- Use of multiple P-T cycles (continuous recycle) using only existing and new LWRs is feasible. Use of long decay periods ($\geq$ 30 years) in the P-T cycles using LWRs would enable significant suppression of the production of curium and heavier actinides during the continued multiple P-T cycles.

- Alternatively, use of short decay periods (~5 years) for irradiation of plutonium in LWRs, as currently practiced in France and other countries, would significantly increase the production of heavier actinides (e.g., ^{241}Pu, ^{242}Pu, ^{243}Am, ^{244}Cm).

- Use of short decay periods (~5 years) can be done effectively by irradiating plutonium in fast reactors and Am-Cm in LWRs because production of ^{241}Pu and heavier actinides is suppressed significantly in those cases.

- Minor actinides (Np, Am, Cm) are more effectively burned in LWRs than in ABRs.

- Optimum performance can be obtained by irradiating Pu (or Pu-Np) in fast spectrum reactors and by irradiating Am-Cm targets in thermal spectrum reactors.

- The approach to equilibrium of the actinides during multiple P-T cycles was not significantly different in cases evaluated for all-thermal-spectrum (LWR) irradiations, (2) all-fast-spectrum

(ABR) irradiations, or (3) hybrid irradiations (Pu-Np in fast reactors and Am-Cm in thermal reactors).

- Because the ABR design sign has been optimized at ~840 MWt, a large number (33–90) of ABRs would be required to transmute the ~23 MT/year TRU actinides currently produced in ~2000 MT/year of low-enriched uranium spent fuel; in comparison, 10–24 existing (or new) 3400 MWt LWRs would be sufficient.

Based on these conclusions, full near-term implementation of P-T in the United States using only ABRs will be difficult; whereas, near-term deployment using LWRs could be utilized. Similar results would be achieved if the oldest (legacy) spent fuel is processed first.

Because the actinides are only partially destroyed in each P-T cycle, using either thermal or fast reactor transmutation, multiple cycles can only be used to stabilize the growth to a state of approximately "no net production." The inventory of TRU actinides in spent fuel that exists when recycling is begun cannot be reduced unless significantly more spent fuel is processed than is being produced. Production is currently > 2000 MT/year (> 23 MT/year of TRU actinides) and is expected to grow in the future. Thus, large processing facilities with capacities significantly greater than 2000 MT/year will be required to reduce the inventory and to provide the fissile plutonium needed to fuel future fast and thermal reactors.

Starting recycle as soon as possible is the most effective way to stop both actinide inventory growth and the continuing need to provide for storage of the spent fuel assemblies that are generated each year. Cost of providing this storage has been estimated to be over $500 million per year.

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Appendix A

Safety Evaluation of Transmutation Fuel Performance in LWRs

Appendix A. Safety Evaluation of Transmutation Fuel Performance in LWRs

Special calculations of reactor core safety factors were made for full power conditions to determine the effects of (a) beginning irradiation with TRU actinides recovered from either 5-year or 30-year-decayed spent fuel; (b) fuel assembly loadings of TRU actinides of either 10% of 15%; (c) actinides including and not including curium; (d) matrices of either depleted uranium, low-enriched uranium or inert metal (zirconium); and (e) heterogeneous (Pu-Np and Am-Cm) or homogenous core loadings. Detailed results are given below. The reactivity change (void coefficients) at beginning-of-cycle (BOC) and end-of-cycle (EOC) were determined at coolant void levels of 10% to 100%.

In summary, negative void coefficients were found for all heterogeneous core configurations and for homogenous cores with 10% TRU actinide loadings. The homogenous cores with 15% TRU actinide loadings encountered positive void coefficients at BOC 90% and 100% coolant voiding conditions and at EOC 100% coolant voiding.

A.1. Reactor Core Containing Fuel Assemblies Composed of 264 Fuel Rods with 10 wt % Np-Pu-Am-Cm Fuel Loaded in a Depleted Uranium Matrix

Reactivity change (pcm) for coolant voiding

	10%	30%	50%	70%	90%	100%
BOC	-1,470	-4,555	-7,653	-10,022	-9,690	-7,393
EOC	-1,535	-4,902	-8,382	-11,225	-11,343	-9,295

Total actinide masses (gram) per fuel assembly

	Pu-tot	Am-tot	Cm-tot	Np-tot
BOL	3.974+04	5.085+03	4.091+01	2.206+03
EOL	3.340+04	2.192+03	9.718+02	1.094+03
30 yr decay	2.931+04	5.636+03	2.900+02	1.061+03

A.2. Reactor Core Containing Fuel Assemblies Composed of 264 Fuel Rods with 15 wt % Np-Pu-Am-Cm Fuel Loaded in a Depleted Uranium Matrix

Reactivity change (pcm) for coolant voiding

	10%	30%	50%	70%	90%	100%
BOC	-965	-2,657	-3,719	-3,392	+91	+3,687
EOC	-1,010	-2,872	-4,136	-4,058	-797	+2,793

Total actinide masses (gram) per fuel assembly

	Pu-tot	Am-tot	Cm-tot	Np-tot
BOL	5.962+04	7.628+03	6.137+01	3.309+03
EOL	5.203+04	4.047+03	1.160+03	1.840+03
30 yr decay	4.686+04	8.283+03	3.243+02	1.811+03

A.3. Reactor Core Containing Fuel Assemblies Composed of 264 Fuel Rods with 10 wt % Np-Pu-Am Fuel Loaded in a Depleted Uranium Matrix

Reactivity change (pcm) for coolant voiding

	10%	30%	50%	70%	90%	100%
BOC	-1,469	-4,554	-7,650	-10,017	-9,680	-7,385
EOC	-1,534	-4,901	-8,380	-11,222	-11,333	-9,287

Total actinide masses (gram) per fuel assembly

	Pu-tot	Am-tot	Cm-tot	Np-tot
BOL	3.978+04	5.090+03	0.000+00	2.208+03
EOL	3.343+04	2.194+03	9.479+02	1.095+03
30 yr decay	2.934+04	5.640+03	2.767+02	1.062+03

A.4. Reactor Core Containing Fuel Assemblies Composed of 264 Fuel Rods with 15 wt % Np-Pu-Am Fuel Loaded in a Depleted Uranium Matrix

Reactivity change (pcm) for coolant voiding

	10%	30%	50%	70%	90%	100%
BOC	-965	-2,657	-3,719	-3,392	+91	+3,687
EOC	-1,010	-2,872	-4,058	-4,058	-797	+2,793

Total actinide masses (gram) per fuel assembly

	Pu-tot	Am-tot	Cm-tot	Np-tot
BOL	5.967+04	7.635+03	0.000+00	3.311+03
EOL	5.207+04	4.051+03	1.120+03	1.842+03
30 yr decay	4.690+04	8.289+03	3.020+02	1.813+03

A.5. Reactor Core Containing Fuel Assemblies Composed of 264 Fuel Rods with 10 wt % Np-Pu-Am-Cm Fuel Loaded in an Inert-Matrix Fuel (IMF) Matrix

Reactivity change (pcm) for coolant voiding

	10%	30%	50%	70%	90%	100%
BOC	-1,069	-3,502	-6,176	-8,450	-8,093	-5,221
EOC	-977	-4,309	-8,504	-13,365	-16,830	-15,610

Total actinide masses (gram) per fuel assembly

	Pu-tot	Am-tot	Cm-tot	Np-tot
BOL	3.392+04	4.340+03	3.492+01	1.882+03
EOL	1.317+04	1.323+03	1.252+03	5.692+03
30 yr decay	1.096+04	3.126+03	3.921+02	5.673+02

A.6. Reactor Core Containing Fuel Assemblies Composed of 264 Fuel Rods with 15 wt % Np-Pu-Am-Cm Fuel Loaded in an IMF Matrix

Reactivity Change (pcm) for Coolant Voiding

	10%	30%	50%	70%	90%	100%
BOC	-740	-2,139	-3,052	-2,495	+1,495	+5,504
EOC	-1,010	-2,872	-4,136	-4,058	-797	+2,793

Total actinide masses (gram) per fuel assembly

	Pu-tot	Am-tot	Cm-tot	Np-tot
BOL	5.130+04	6.564+03	5.281+01	2.847+03
EOL	3.036+04	2.790+03	1.400+03	1.219+03
30 yr decay	2.584+04	6.533+03	4.177+02	1.217+03

A.7. Reactor Core Containing Fuel Assemblies Composed of 264 Fuel Rods with 10 wt % Np-Pu-Am Fuel Loaded in an IMF Matrix

Reactivity change (pcm) for coolant voiding

	10%	30%	50%	70%	90%	100%
BOC	-1,069	-3,500	-6,171	-8,440	-8,080	-5,206
EOC	-975	-4,307	-8,500	-13,355	-16,812	-15,589

Total actinide masses (gram) per fuel assembly

	Pu-tot	Am-tot	Cm-tot	Np-tot
BOL	3.395+04	4.344+03	0.000+00	1.884+03
EOL	1.319+04	1.324+03	1.236+03	5.698+02
30 yr decay	1.097+04	3.130+03	3.841+02	5.679+02

A.8. Reactor Core Containing Fuel Assemblies Composed of 264 Fuel Rods with 15 wt % Np-Pu-Am Fuel Loaded in an IMF Matrix

Reactivity change (pcm) for coolant voiding

	10%	30%	50%	70%	90%	100%
BOC	-739	-2,137	-3,047	-2,487	+1,505	+5,514
EOC	-888	-2,855	-4,533	-4,918	-1,790	+2,178

Total actinide masses (gram) per fuel assembly

	Pu-tot	Am-tot	Cm-tot	Np-tot
BOL	5.135+04	6.569+03	0.000+00	2.849+03
EOL	3.040+04	2.793+03	1.370+03	1.221+03
30 yr decay	2.587+04	6.538+03	4.011+02	1.218+03

For reactor cores composed of fuel assemblies, each of which containing 216 fuel rods with Np-Pu-Am or Np-Pu-Am-Cm in an IMF or depleted uranium matrix and 48 low-enriched uranium fuel rods, and one-third of the core being replaced at each fuel cycle, the void reactivity coefficients were negative for voiding fractions ranging from 10% to 100% and having an Np-Pu-Am-(Cm) density in the matrix of 10 wt % and 15 wt %.

Appendix B

Actinide Composition Data for Scenario In Which All Actinides are Transmuted in LWRs With Heterogeneous Cores (Pu-Np plus Am-Cm)

LWR 30-year decay
Actinides in Feed to Each P-T cycle

P-T Cycle No.	1		2		3		4		5		6	
	(MT/y)	(%)	(MT/y)	(%)	(MT/y)	(%)	(MT/y)	(%)	(MT/y)	(%)	(MT/y)	(%)
Np-237	1.087	100	1.434	100	1.536	100	1.557	100	1.555	100	1.549	100
Pu-238	0.302	1.5	1.69	5.6	3.061	8.3	3.967	9.6	4.442	10.0	4.640	9.9
Pu-239	13.05	66.6	15.90	53.0	16.87	45.9	17.34	41.9	17.54	39.4	17.59	37.7
Pu-240	4.66	23.8	8.435	28.1	10.24	27.9	10.99	26.6	11.27	25.3	11.35	24.3
Pu-241	0.703	3.6	1.182	3.9	1.422	3.9	1.531	3.7	1.577	3.5	1.595	3.4
Pu-242	0.887	4.5	2.794	9.3	5.16	14.0	7.515	18.2	9.65	21.7	11.50	24.6
Total Pu	19.60	100.0	30.00	100.0	36.75	100.0	41.34	100.0	44.48	100.0	46.68	100.0
Am-241	2.313	92.9	4.279	87.5	5.36	82.3	5.842	77.6	6.03	73.5	6.078	70.1
Am-242m	0.0015	0.1	0.0119	0.2	0.019	0.3	0.022	0.3	0.023	0.3	0.023	0.3
Am-243	0.177	7.1	0.601	12.3	1.134	17.4	1.669	22.2	2.15	26.2	2.566	29.6
Total Am	2.49	100.1	4.892	100.0	6.508	100.0	7.533	100.0	8.203	100.0	8.667	100.0
Cm-243	0.0003	1.5	0.005	2.6	0.009	1.8	0.011	1.3	0.012	1.0	0.012	0.8
Cm-244	0.0166	83.0	0.132	68.6	0.3196	65.8	0.532	64.3	0.737	62.9	0.919	61.6
Cm-245	0.0028	14.0	0.048	24.9	0.1245	25.6	0.206	24.9	0.279	23.8	0.338	22.7
Cm-246	0.0002	1.0	0.0074	3.8	0.033	6.8	0.079	9.5	0.144	12.3	0.223	14.9
Total Cm	0.020	99.5	0.1924	100.0	0.486	100.0	0.828	100.0	1.172	100.0	1.492	100.0
Total TRUs	23.2		36.5		45.3		51.3		55.4		58.4	
Sum Pu-241 + Am-241	3.02		5.46		6.78		7.37		7.61		7.67	
Total IDH (MW-y/y)	231		445		585		663		704		723	
Np (%)	0.014		0.010		0.008		0.007		0.007		0.006	
Pu (%)	47.9		48.7		49.7		50.2		50.3		50.0	
Am(%)	51.7		50.1		48.3		47.0		46.1		45.6	
Cm(%)	0.23		0.97		1.79		2.62		3.42		4.15	

LWR 30-year decay
Irradiated Pu-Np from Each P-T Cycle

P-T Cycle No.	1		2		3		4		5		6	
	(MT/y)	(%)	(MT/y)	(%)	(MT/y)	(%)	(MT/y)	(%)	(MT/y)	(%)	(MT/y)	(%)
Np-237	0.381	100	0.4875	100	0.5137	100	0.5162	100	0.5121	100	0.5068	100
Pu-238	0.557	5.8	1.234	8.0	1.758	9.0	2.056	9.2	2.183	8.9	2.213	8.5
Pu-239	3.167	32.9	4.22	27.4	4.78	24.5	5.094	22.7	5.250	21.4	5.31	20.4
Pu-240	3.85	40.0	5.606	36.4	6.307	32.4	6.56	29.3	6.604	26.9	6.566	25.2
Pu-241	0.466	4.8	0.687	4.5	0.783	4.0	0.821	3.7	0.833	3.4	0.833	3.2
Pu-242	1.593	16.5	3.671	23.8	5.846	30.0	7.874	35.2	9.656	39.4	11.17	42.8
Total Pu	9.63	100.0	15.41	100.0	19.47	100.0	22.40	100.0	24.53	100.0	26.10	100.0
Am-241	1.601	84.6	2.365	79.1	2.69	74.1	2.821	69.9	2.859	66.4	2.855	63.6
Am-242m	0.0027	0.1	0.0040	0.1	0.0044	0.1	0.0045	0.1	0.0045	0.1	0.0044	0.1
Am-243	0.289	15.3	0.6187	20.7	0.935	25.8	1.211	30.0	1.442	33.5	1.632	36.3
Total Am	1.89	100.0	2.988	100.0	3.631	100.0	4.037	100.0	4.306	100.0	4.491	100.0
Cm-243	0.0007	0.7	0.0011	0.5	0.001	0.4	0.0014	0.3	0.0015	0.3	0.0015	0.2
Cm-244	0.0661	70.5	0.152	70.3	0.2387	70.4	0.3157	70.6	0.3807	70.9	0.4343	71.1
Cm-245	0.0238	25.4	0.055	25.4	0.0854	25.2	0.1112	24.9	0.132	24.6	0.1485	24.3
Cm-246	0.0031	3.3	0.0081	3.7	0.0135	4.0	0.0186	4.2	0.023	4.3	0.0268	4.4
Total Cm	0.0937	100.0	0.2164	100.0	0.3389	100.0	0.4469	100.0	0.5372	100.0	0.6111	100.0
Total TRUs	12.0		19.1		24.0		27.4		29.9		31.7	
Sum Pu-241 + Am-241	2.07		3.05		3.48		3.64		3.69		3.69	
Total IDH (MW-y/y)	166		260		311		337		348		352	
Np (%)	0.007		0.006		0.005		0.005		0.004		0.004	
Pu (%)	47.8		49.6		50.7		51.2		51.1		50.8	
Am(%)	50.7		48.3		46.5		45.6		45.1		45.0	
Cm(%)	1.30		1.91		2.51		3.06		3.58		4.03	

LWR 30-year decay
Irradiated Am-Cm from Each P-T Cycle

P-T Cycle No.	1		2		3		4		5		6	
	(MT/y)	(%)	(MT/y)	(%)	(MT/y)	(%)	(MT/y)	(%)	(MT/y)	(%)	(MT/y)	(%)
Np-237	0.0058	100	0.0879	100	0.1035	100	0.1125	100	0.1184	100	0.1227	100
Pu-238	0.810	52.2	1.478	51.2	1.842	50.1	2.005	49.0	2.067	48.0	2.082	47.1
Pu-239	0.382	24.6	0.731	25.3	0.96	26.0	1.095	26.8	1.183	27.5	1.24	28.1
Pu-240	0.105	6.8	0.205	7.1	0.276	7.5	0.33	7.9	0.359	8.3	0.385	8.7
Pu-241	0.015	1.0	0.030	1.0	0.040	1.1	0.047	1.1	0.052	1.2	0.055	1.2
Pu-242	0.240	15.5	0.445	15.4	0.561	15.3	0.617	15.1	0.642	14.9	0.65	14.8
Total Pu	1.552	100.0	2.889	100.0	3.676	100.0	4.089	100.0	4.303	100.0	4.416	100.0
Am-241	0.362	75.6	0.659	68.5	0.808	61.4	0.863	55.1	0.873	49.9	0.865	45.8
Am-242m	0.0074	1.5	0.0130	1.4	0.016	1.2	0.017	1.1	0.0166	0.9	0.0161	0.9
Am-243	0.110	23.0	0.290	30.1	0.491	37.3	0.685	43.8	0.858	49.1	1.006	53.3
Total Am	0.479	100.1	0.962	100.0	1.315	100.0	1.565	100.0	1.748	100.0	1.887	100.0
Cm-243	0.0039	5.8	0.0073	3.2	0.0092	2.1	0.010	1.5	0.0103	1.2	0.0103	0.9
Cm-244	0.0415	61.2	0.135	59.7	0.2540	58.0	0.3767	56.6	0.4898	55.0	0.5881	53.5
Cm-245	0.0186	27.4	0.061	27.0	0.1108	25.3	0.1558	23.4	0.1928	21.6	0.2222	20.2
Cm-246	0.0038	5.6	0.023	10.2	0.064	14.6	0.1232	18.5	0.1984	22.3	0.2783	25.3
Total Cm	0.0678	100.0	0.2263	100.0	0.4380	100.0	0.6657	100.0	0.8913	100.0	1.0989	100.0
Total TRUs	2.1		4.2		5.5		6.4		7.1		7.5	
Sum Pu-241 + Am-241	0.38		0.69		0.85		0.91		0.93		0.92	
Total IDH (MW-y/y)	50		94		121		135		144		148	
Np (%)	0.000		0.003		0.003		0.002		0.002		0.002	
Pu (%)	58.5		57.1		55.7		54.4		53.2		52.1	
Am(%)	38.8		38.2		37.4		36.5		35.6		34.9	
Cm(%)	2.72		4.70		6.89		9.10		11.15		12.94	

LWR 30-year decay
Total Recycle from Each P-T Cycle

P-T Cycle No.	1		2		3		4		5		6	
	(MT/y)	(%)	(MT/y)	(%)	(MT/y)	(%)	(MT/y)	(%)	(MT/y)	(%)	(MT/y)	(%)
Np-237	0.387	100	0.575	100	0.617	100	0.629	100	0.6305	100	0.630	100
Pu-238	1.367	12.2	2.712	14.8	3.600	15.6	4.061	15.3	4.250	14.7	4.295	14.1
Pu-239	3.549	31.7	4.946	27.0	5.736	24.8	6.189	23.4	6.43	22.3	6.556	21.5
Pu-240	3.955	35.3	5.811	31.8	6.583	28.4	6.880	26.0	6.963	24.2	6.951	22.8
Pu-241	0.481	4.3	0.717	3.9	0.823	3.6	0.868	3.3	0.885	3.1	0.888	2.9
Pu-242	1.833	16.4	4.116	22.5	6.407	27.7	8.491	32.1	10.30	35.7	11.82	38.7
Total Pu	11.19	100.0	18.30	100.0	23.15	100.0	26.49	100.0	28.83	100.0	30.51	100.0
Am-241	1.968	82.8	3.024	76.5	3.500	70.7	3.684	65.8	3.732	61.6	3.72	58.3
Am-242m	0.010	0.4	0.017	0.4	0.020	0.4	0.022	0.4	0.021	0.3	0.021	0.3
Am-243	0.397	16.7	0.9087	23.0	1.426	28.8	1.896	33.8	2.300	38.0	2.64	41.4
Total Am	2.38	99.9	3.95	100.0	4.95	100.0	5.60	100.0	6.05	100.0	6.38	100.0
Cm-243	0.0046	2.8	0.0084	1.9	0.0105	1.4	0.011	1.0	0.012	0.8	0.012	0.7
Cm-244	0.1076	66.5	0.287	64.9	0.4933	63.4	0.692	62.2	0.871	61.0	1.022	59.8
Cm-245	0.0424	26.2	0.116	26.2	0.1962	25.2	0.267	24.0	0.325	22.8	0.371	21.7
Cm-246	0.0069	4.3	0.031	7.0	0.0775	10.0	0.142	12.8	0.219	15.3	0.305	17.8
Total Cm	0.162	99.9	0.442	100.1	0.778	100.0	1.112	100.0	1.427	100.0	1.71	100.0
Total TRUs	14.1		23.3		29.5		33.8		36.9		39.2	
Sum Pu-241 + Am-241	2.45		3.74		4.32		4.55		4.62		4.61	
Total IDH (MW-y/y)	216		353		431		471		491		500	
Np(%)	0.01		0.00		0.00		0.00		0.00		0.00	
Pu(%)	50.5		51.8		52.4		52.3		51.9		51.4	
Am(%)	48.1		45.7		44.0		43.0		42.4		42.1	
Cm(%)	1.63		2.7		3.7		4.8		5.8		6.7	

Appendix C

Actinide Composition Data for Scenario In Which All Actinides are Transmuted in the FR/0.25CR

FR/0.25CR 5-year decay
Actinides in Feed to Each P-T cycle

P-T Cycle No.	1		2		3		4		5	
	(MT/y)	(%)	(MT/y)	(%)	(MT/y)	(%)	(MT/y)	(%)	(MT/y)	(%)
Np-237	1.097	100	1.722	100	2.061	100	2.232	100	2.311	100
Pu-238	0.302	1.5	1.137	3.4	1.973	4.5	2.634	5.2	3.101	5.5
Pu-239	13.05	66.6	20.55	61.2	24.90	57.1	27.46	54.0	29.00	51.7
Pu-240	4.66	23.8	8.98	26.7	12.68	29.1	15.70	30.9	18.08	32.2
Pu-241	0.703	3.6	1.179	3.5	1.561	3.6	1.879	3.7	2.139	3.8
Pu-242	0.887	4.5	1.74	5.2	2.509	5.8	3.18	6.3	3.75	6.7
Total Pu	19.60	100.0	33.59	100.0	43.62	100.0	50.85	100.0	56.07	100.0
Am-241	2.313	92.9	3.682	89.6	4.49	87.0	4.972	84.7	5.27	82.9
Am-242m	0.0015	0.1	0.065	1.6	0.12	2.3	0.156	2.7	0.179	2.8
Am-243	0.177	7.1	0.365	8.9	0.555	10.8	0.74	12.6	0.909	14.3
Total Am	2.49	100.1	4.11	100.0	5.16	100.1	5.87	100.0	6.36	100.0
Cm-243	0.0003	1.5	0.0051	6.0	0.010	5.6	0.014	4.8	0.0167	4.1
Cm-244	0.0166	83.0	0.068	80.6	0.141	79.2	0.225	77.6	0.312	76.5
Cm-245	0.0028	14.0	0.0106	12.6	0.025	14.0	0.046	15.9	0.071	17.4
Cm-246	0.0002	1.0	0.0007	0.8	0.002	1.1	0.0045	1.6	0.0083	2.0
Total Cm	0.020	99.5	0.0844	100.0	0.178	100.0	0.290	99.8	0.408	100.0
Total TRUs	23.2		39.5		51.0		59.2		65.1	
Sum Pu-241 + Am-241	3.02		4.86		6.05		6.85		7.41	
Total IDH (MW-y/y)	231		406		536		632		702	
Np (%)	0.01		0.01		0.01		0.01		0.01	
Pu (%)	48.0		52.4		55.6		57.7		59.2	
Am (%)	51.8		47.1		43.6		41.1		39.3	
Cm (%)	0.23		0.55		0.86		1.16		1.45	

FR/0.25CR 5-year decay
Total actinide recycle from each P-T cycle

P-T Cycle No.	1		2		3		4		5	
	(MT/y)	(%)	(MT/y)	(%)	(MT/y)	(%)	(MT/y)	(%)	(MT/y)	(%)
Np-237	0.645	100	0.998	100	1.18	100	1.266	100	1.30	100
Pu-238	0.841	5.9	1.681	6.9	2.345	7.3	2.814	7.6	3.125	7.6
Pu-239	7.68	53.7	12.16	49.6	14.81	46.4	16.40	44.1	17.38	42.3
Pu-240	4.41	30.9	8.17	33.3	11.23	35.2	13.64	36.6	15.48	37.7
Pu-241	0.489	3.4	0.881	3.6	1.205	3.8	1.47	3.9	1.68	4.1
Pu-242	0.870	6.1	1.65	6.7	2.328	7.3	2.90	7.8	3.38	8.2
Total Pu	14.29	100.0	24.54	100.0	31.92	100.0	37.22	100.0	41.05	100.0
Am-241	1.414	84.8	2.252	81.8	2.76	79.2	3.073	77.0	3.276	75.1
Am-242m	0.063	3.8	0.118	4.3	0.155	4.5	0.177	4.4	0.191	4.4
Am-243	0.191	11.5	0.384	13.9	0.570	16.4	0.741	18.6	0.893	20.5
Total Am	1.668	100.0	2.754	100.0	3.483	100.0	3.99	100.0	4.36	100.0
Cm-243	0.0049	7.5	0.0098	6.1	0.014	5.2	0.016	4.1	0.018	3.6
Cm-244	0.052	79.6	0.125	78.3	0.209	77.1	0.296	76.3	0.381	75.1
Cm-245	0.0079	12.1	0.023	14.4	0.044	16.2	0.068	17.5	0.095	18.7
Cm-246	0.0005	0.8	0.0018	1.1	0.0043	1.6	0.0081	2.1	0.013	2.6
Total Cm	0.0653	100	0.1596	100.0	0.271	100.1	0.388	100.0	0.507	100.0
Total TRUs	16.7		28.5		36.9		42.9		47.2	
Sum Pu-241 + Am-241	1.90		3.13		3.96		4.54		4.96	
Total IDH (MW-y/y)	179		313		411		482		534	
Np (%)	0.011		0.010		0.009		0.008		0.007	
Pu (%)	57.9		60.9		63.0		64.2		65.0	
Am (%)	41.1		37.8		35.4		33.7		32.6	
Cm (%)	0.95		1.3		1.7		2.0		2.3	

Appendix D

Actinide Composition Data for Scenario In Which Pu-Np is Transmuted in the FR/0.25CR and Am-Cm in the LWR

Pu-Np in FR/0.25CR Am-Cm in LWR 5-year decay

Actinides in Feed to Each P-T cycle

P-T Cycle No.	1		2		3		4		5	
	(MT/y)	(%)	(MT/y)	(%)	(MT/y)	(%)	(MT/y)	(%)	(MT/y)	(%)
Np-237	1.097	100	1.773	100	2.157	100	2.363	100	2.465	100
Pu-238	0.302	1.5	1.658	4.7	2.689	5.8	3.38	6.2	3.81	6.3
Pu-239	13.05	66.6	21.06	59.9	25.88	55.8	28.77	52.9	30.50	50.7
Pu-240	4.66	23.8	9.14	26.0	13.08	28.2	16.36	30.1	19.01	31.6
Pu-241	0.703	3.6	1.372	3.9	1.88	4.1	2.28	4.2	2.61	4.3
Pu-242	0.887	4.5	1.91	5.4	2.82	6.1	3.59	6.6	4.24	7.0
Total Pu	19.60	100.0	35.14	100.0	46.35	100.0	54.38	100.0	60.17	100.0
Am-241	2.313	92.9	2.726	87.9	2.90	84.5	3.017	82.0	3.11	80.2
Am-242m	0.0015	0.1	0.0118	0.4	0.0146	0.4	0.016	0.4	0.017	0.4
Am-243	0.177	7.1	0.361	11.6	0.513	15.0	0.643	17.5	0.757	19.5
Total Am	2.49	100.1	3.10	100.0	3.43	99.9	3.68	99.9	3.88	100.1
Cm-243	0.0003	1.5	0.0076	4.3	0.0093	2.6	0.010	1.8	0.010	1.4
Cm-244	0.0166	83.0	0.140	80.0	0.286	79.9	0.430	78.6	0.567	78.8
Cm-245	0.0028	14.0	0.023	13.1	0.047	13.1	0.069	12.6	0.0875	12.2
Cm-246	0.0002	1.0	0.004	2.3	0.015	4.2	0.038	6.9	0.055	7.6
Total Cm	0.020	99.5	0.175	99.8	0.358	99.8	0.547	100.0	0.720	99.9
Total TRUs	23.2		40.2		52.3		61.0		67.2	
Sum Pu-241 + Am-241	3.02		4.10		4.78		5.30		5.72	
Total IDH (MW-y/y)	231		389		505		592		658	
Np(%)	0.01		0.01		0.01		0.01		0.01	
Pu(%)	48.2		62.5		68.3		71.0		72.4	
Am(%)	51.8		36.5		30.1		26.9		25.0	
Cm(%)	0.23		1.2		1.9		2.4		2.8	

Pu-Np in FR/0.25CR Am-Cm in LWR 5-year decay

Irradiated Pu-Np from Each P-T Cycle

P-T Cycle No.	1		2		3		4		5	
	(MT/y)	(%)	(MT/y)	(%)	(MT/y)	(%)	(MT/y)	(%)	(MT/y)	(%)
Np-237	0.665	100	1.056	100	1.267	100	1.372	100	1.419	100
Pu-238	0.397	2.9	1.257	5.1	1.869	5.7	2.257	5.9	2.49	5.8
Pu-239	7.78	56.1	12.63	50.8	15.56	47.4	17.31	44.9	18.37	43.0
Pu-240	4.44	32.0	8.40	33.8	11.70	35.6	14.36	37.2	16.44	38.5
Pu-241	0.489	3.5	0.948	3.8	1.32	4.0	1.624	4.2	1.87	4.4
Pu-242	0.767	5.5	1.631	6.6	2.39	7.3	3.026	7.8	3.56	8.3
Total Pu	13.87	100.0	24.87	100.0	32.84	100.0	38.58	100.0	42.73	100.0
Am-241	0.199	73.7	0.385	71.2	0.53	69.5	0.654	68.3	0.751	67.7
Am-242m	0.0017	0.6	0.0035	0.6	0.0049	0.6	0.0060	0.6	0.0070	0.6
Am-243	0.068	25.2	0.152	28.1	0.229	29.8	0.297	31.0	0.356	32.1
Total Am	0.27	99.5	0.541	99.9	0.768	100.0	0.957	100.0	1.11	100.4
Cm-243	0.0001	1.0	0.0002	0.9	0.0003	0.8	0.0004	0.8	0.0005	0.8
Cm-244	0.009	90.0	0.021	91.7	0.033	91.4	0.044	91.5	0.054	91.4
Cm-245	0.0007	7.0	0.00165	7.2	0.00265	7.3	0.0036	7.5	0.0045	7.6
Cm-246	0.00001	0.1	0.00003	0.1	0.0001	0.3	0.0001	0.2	0.0001	0.2
Total Cm	0.010	98.1	0.0229	99.9	0.0361	99.9	0.0481	100.0	0.0591	100.0
Total TRUs	14.8		26.5		34.9		41.0		45.3	
Sum Pu-241 + Am-241	0.69		1.33		1.85		2.28		2.62	
Total IDH (MW-y/y)	101		204		284		344		389	
Np (%)	0.02		0.02		0.01		0.01		0.01	
Pu (%)	89.1		89.5		89.4		89.2		89.0	
Am (%)	10.6		10.2		10.2		10.4		10.6	
Cm (%)	0.29		0.34		0.38		0.42		0.45	

Pu-Np in FR/0.25CR Am-Cm in LWR 5-year decay

Irradiated Am-Cm from Each P-T Cycle

P-T Cycle No.	1		2		3		4		5	
	(MT/y)	(%)	(MT/y)	(%)	(MT/y)	(%)	(MT/y)	(%)	(MT/y)	(%)
Np-237	0.016	100	0.021	100	0.024	100	0.026	100	0.029	100
Pu-238	0.941	42.3	1.11	54.3	1.182	53.4	1.231	52.6	1.27	51.6
Pu-239	0.368	16.5	0.458	22.4	0.509	23.0	0.551	23.5	0.59	24.0
Pu-240	0.101	4.5	0.13	6.3	0.15	6.6	0.162	6.9	0.18	7.3
Pu-241	0.049	2.2	0.063	3.1	0.072	3.3	0.079	3.4	0.09	3.7
Pu-242	0.767	34.5	0.284	13.9	0.304	13.7	0.319	13.6	0.33	13.4
Total Pu	2.226	100.0	2.044	100.0	2.214	100.0	2.342	100.0	2.46	100.0
Am-241	0.343	74.2	0.390	67.5	0.400	62.3	0.403	58.4	0.404	55.3
Am-242m	0.0085	1.8	0.0096	1.7	0.0098	1.5	0.0098	1.4	0.0100	1.4
Am-243	0.11	23.8	0.178	30.8	0.232	36.1	0.277	40.1	0.316	43.3
Total Am	0.462	99.9	0.578	99.9	0.642	100.0	0.690	100.0	0.730	100.0
Cm-243	0.0071	5.2	0.0087	2.8	0.0093	2.0	0.0097	1.5	0.0099	1.2
Cm-244	0.108	78.8	0.240	78.5	0.372	78.1	0.496	77.5	0.612	76.8
Cm-245	0.0185	13.5	0.042	13.7	0.063	13.2	0.080	12.5	0.095	11.9
Cm-246	0.0038	2.8	0.015	4.9	0.032	6.7	0.054	8.4	0.080	10.0
Total Cm	0.137	100.3	0.3057	100.0	0.4763	100.0	0.6397	100.0	0.7969	100.0
Total TRUs	2.8		2.9		3.4		3.7		4.0	
Sum Pu-241 + Am-241	0.39		0.45		0.47		0.48		0.49	
Total IDH (MW-y/y)	57.2		71.1		79.5		86.2		92.4	
Np (%)	0.0008		0.0009		0.0009		0.0009		0.0009	
Pu (%)	61.7		59.0		56.8		54.9		53.4	
Am (%)	32.1		29.9		27.9		26.3		24.9	
Cm (%)	6.2		11.0		15.3		18.8		21.6	

Pu-Np in FR/0.25CR Am-Cm in LWR 5-year decay
Total Recycle Irradiated Actinides from Each P-T Cycle

P-T Cycle No.	1		2		3		4		5	
	(MT/y)	(%)	(MT/y)	(%)	(MT/y)	(%)	(MT/y)	(%)	(MT/y)	(%)
Np-237	0.681	100	1.077	100	1.291	100	1.398	100	1.45	100
Pu-238	1.338	8.6	2.367	8.8	3.05	8.7	3.49	8.5	3.76	8.3
Pu-239	8.15	52.3	13.09	48.6	16.07	45.8	17.86	43.6	18.96	42.0
Pu-240	4.54	29.2	8.53	31.7	11.85	33.8	14.52	35.5	16.62	36.8
Pu-241	0.538	3.5	1.01	3.8	1.39	4.0	1.7	4.2	1.96	4.3
Pu-242	1.006	6.5	1.92	7.1	2.69	7.7	3.35	8.2	3.89	8.6
Total Pu	15.57	100.0	26.92	100.0	35.05	100.0	40.92	100.0	45.19	100.0
Am-241	0.542	74.2	0.775	69.3	0.934	66.2	1.057	64.2	1.16	62.7
Am-242m	0.010	1.4	0.013	1.2	0.015	1.1	0.016	1.0	0.0170	0.9
Am-243	0.178	24.4	0.33	29.5	0.461	32.7	0.574	34.9	0.672	36.3
Total Am	0.730	100.0	1.118	100.0	1.410	100.0	1.647	100.0	1.849	100.0
Cm-243	0.007	4.8	0.009	2.7	0.0096	1.9	0.010	1.5	0.010	1.2
Cm-244	0.117	79.6	0.261	79.3	0.405	79.0	0.540	78.4	0.666	77.8
Cm-245	0.019	12.9	0.044	13.4	0.0655	12.8	0.084	12.2	0.100	11.7
Cm-246	0.004	2.7	0.015	4.6	0.0325	6.3	0.055	8.0	0.080	9.3
Total Cm	0.147	100.0	0.329	100.0	0.5126	100.0	0.689	100.0	0.856	100.0
Total TRUs	17.1		29.4		38.3		44.7		49.3	
Sum Pu-241 + Am-241	1.08		1.79		2.32		2.76		3.12	
Total IDH (MW-y/y)	158		275		364		430		481	
Np (%)	0.01		0.01		0.01		0.01		0.01	
Pu (%)	79.1		81.6		82.3		82.3		82.1	
Am (%)	18.4		15.3		14.1		13.6		13.4	
Cm (%)	2.4		3.1		3.6		4.1		4.5	

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