ICONE
Conference Paper

Proceedings of ICONE 10:
10th International Conference on Nuclear Engineering
April 14-18, 2002, Arlington, VA, USA

22575

MULTIPLE TIER FUEL CYCLE STUDIES FOR WASTE TRANSMUTATION

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ABSTRACT

As part of the U.S. Department of Energy Advanced Accelerator Applications Program, a systems study was conducted to evaluate the transmutation performance of advanced fuel cycle strategies. Three primary fuel cycle strategies were evaluated: dual-tier systems with plutonium separation, dual-tier systems without plutonium separation, and single-tier systems without plutonium separation. For each case, the system mass flow and TRU consumption were evaluated in detail. Furthermore, the loss of materials in fuel processing was tracked including the generation of new waste streams. Based on these results, the system performance was evaluated with respect to several key transmutation parameters including TRU inventory reduction, radiotoxicity, and support ratio. The importance of clean fuel processing (~0.1% losses) and inclusion of a final tier fast spectrum system are demonstrated. With these two features, all scenarios capably reduce the TRU and plutonium waste content, significantly reducing the radiotoxicity; however, a significant infrastructure (at least 1/10 the total nuclear capacity) is required for the dedicated transmutation system.

I. INTRODUCTION

The primary goal of the transmutation mission remains – to transmute the hazardous components of spent nuclear fuel. Thus, the majority of the transuranic (TRU) material needs to be fissioned, producing ~1 MW-day of energy for every gram. In previous work, the focus was on direct utilization of the TRUs contained in LWR spent fuel in a dedicated accelerator transmutation of waste (ATW) transmuter. However, other systems may be more efficient in consuming this material, particularly early in the process when the fissile content is still high. Thus, a variety of alternative fuel cycle strategies have been proposed where the transmutation campaign is divided into several phases, denoted tiers. For example, the TRU material could first be irradiated in a thermal reactor (to burn the plutonium) with subsequent irradiation in a fast spectrum system (to burn the minor actinides). The purpose of this study was to identify promising fuel cycle options and provide a self-consistent evaluation of their performance.

A sustained nuclear power industry analogous to the current U.S. situation was presumed. A distinct commercial nuclear power production sector was assumed which provides a continuing feed of spent nuclear fuel; for this study, an evolutionary advanced light water reactor (ALWR) with burnup of 50 GWd/MT was used. This spent fuel is treated in a subsequent government owned waste transmutation enterprise to improve the disposed waste characteristics (e.g., reduce the long-lived toxicity), and reduce the quantity of the fissile material discharged to waste. The specific power production and transmutation systems are described in Section II.

Three primary fuel cycle strategies were evaluated in this study: dual-tier systems with plutonium separation, dual-tier systems without plutonium separation, and single-tier fast systems without plutonium separation. For each case, the system mass flow and TRU consumption were evaluated in detail based on existing designs for each technology option. Furthermore, the loss of materials in fuel processing was tracked (0.1% loss each pass) including the generation of new waste streams and the material characteristics at each point in

the extended fuel cycle. The analytical techniques are briefly described in Section III. The key modeling assumptions and performance objectives are discussed in more detail in a companion paper [1].

Based on these results, the system performance was evaluated with respect to a wide variety of transmutation criteria. The basic mass flow and TRU consumption results are given in Section IV. Several of the key derived performance parameters are also summarized including comparisons of the waste mass loading (Section V.1), radiotoxicity (Section V.2), and support ratio (Section V.3). The impact of transmutation on repository design, operation, and performance is discussed in a companion paper [2].

II. DESCRIPTION OF TRANSMUTATION SYSTEMS

The transmutation systems are designed to transmute the spent fuel inventory resulting from a sustained capacity of ALWRs that operate on a once-through fuel cycle. Single and double tier transmutation scenarios were considered. For the first tier, thermal reactor systems (using plutonium or TRU-based fuel) were employed. Both advanced light water reactor (ALWR) and advanced gas cooled reactor (GT-MHR) technologies were considered, using both mixed oxide (MOX) fuel and nonfertile fuel (NFF) forms. In all cases, a final tier fast spectrum system was utilized; both subcritical accelerator-driven (with non-uranium fuel) and fast reactor options (with conventional fertile fuel forms) were considered. Existing system designs for the five distinct systems types noted above: commercial ALWR, transmutation ALWR, GT-MHR, fast spectrum accelerator-driven (i.e., ATW), and fast reactor, are briefly referenced in this section. The systems were adapted to the transmutation mission (e.g., modified fuel type), but were not optimized for this application.

For the commercial system, it is presumed that an evolutionary LWR design will be employed. The precise design parameters of these power production systems will be determined by commercial concerns. A key assumption is that enriched uranium fuel will be utilized, but that a discharge burnup level higher than current LWRs will be achieved. The discharge fuel composition was based on a previous high burnup (50,000 MWd/MT) PWR spent fuel evaluation. In particular, the extended burnup PWR model developed in the Yucca Mountain spent fuel evaluations [3] was utilized. This model is based on detailed depletion computations for a "generic" PWR assembly design using an enrichment of 4.2% U-235/U to obtain the higher burnup level.

For the ALWR transmutation system, it is presumed that a LWR is adapted for the transmutation mission to utilize 100% core loading of alternate (transmutation) fuel forms. In particular, mixed oxide (MOX) fuel and nonfertile fuel (NFF) forms were considered using the 100% MOX and NFF full-core scenarios that were proposed by *Paul Scherrer Institute* [4]. The MOX fuel design is similar to that employed in operating French LWRs. The NFF fuel design employs TRU oxides embedded in an inert ZrO_2 matrix. The presence of the diluent matrix severely reduces the heavy metal loading for the proposed NFF, this should allow *much* higher heavy metal burnup to be achieved without fuel failure. The NFF is also loaded with burnable poison to compensate reactivity losses associated with the elimination of the fertile uranium.

For the thermal (spectrum) gas reactor transmutation system, it is presumed that a General Atomics GT-MHR design is adapted for the transmutation mission to utilize 100% core loading of particles based on the "conventional" TRISO design but with a transuranic or plutonium oxide fuel kernel. The reactor design is identical to the standard 600MWt GT-MHR [5]. Each fuel element block has provision for 216 fuel/burnable poison channels, and 102 coolant passages. Ten blocks are stacked to form a fuel column, and 108 fuel columns are formed in three rings between inner and outer graphite reflectors to form the active core.

For the fast spectrum accelerator-driven system, the ATW system point design employing a sodium cooled transmutation blanket [6] was utilized. The subcritical transmutation blanket is assumed to be fueled with a non-uranium metallic alloy (TRU with ~40% Zr) fuel; pyrochemical techniques are used for recycle of residual transuranics in this fuel after irradiation. A transmuter fission power level of 840 MWt was assumed. The point design layout consists of a central LBE target/buffer surrounded by 132 fuel assemblies. Flattening of the blanket power distribution is accomplished by dividing the blanket into two "enrichment" zones, with the outer zone utilizing a higher TRU fraction in the alloy.

For the transmutation fast reactor, the 840 MWt Advanced Liquid Metal Reactor (ALMR) design was utilized for this study. This design was developed in the former U.S. fast reactor program by General Electric and Argonne National Laboratory; conventional and burner configurations for a weapons plutonium disposition mission are described in Ref. 7. The ALMR burner design utilizes standard fertile fuel forms – ternary metal U/TRU-10Zr alloy with maximum TRU content of ~30%. The core geometry in this configuration was "spoiled" by reducing the core height to 18 inches. This increases the leakage and reduces the conversion ratio (CR) to ~0.5; this allows net consumption of the TRU feed at roughly half the rate of a pure burner (i.e., ATW) system.

III. DESCRIPTION OF ANALYTICAL METHODS AND MODELS

Five distinct system types: commercial ALWR, transmutation ALWR, GT-MHR, fast spectrum ADS (i.e., ATW), and advanced fast reactor (ALMR) were identified and briefly described in Section 2. In this section, the analytical techniques used to evaluate transmutation (i.e., TRU depletion and production), system performance, and detailed spent fuel characteristics are described. In addition, key assumptions employed in these analyses are explicitly identified.

No detailed computations were performed for the commercial ALWR system. Instead the ORIGEN2 computations developed in Ref. 3 were reproduced; these

standard burnup computations for "generic" reactor systems were utilized to estimate the Yucca Mountain Project (YMP) spent fuel inventory. The standard PWR benchmark composition with extended cooling time (25 years) was utilized as the feed material for previous ATW system studies. For this study, the extended burnup PWR benchmark was utilized with an assumed cooling time of 10 years before the commercial ALWR fuel is processed for insertion into the transmutation system. This feed composition is compared to the YMP inventory and the separated plutonium feed in Table 1.

Table 1: Commercial Spent Fuel Feed Specification for AAA System Studies

Isotope	YMP Inventory[a]	ALWR[b] TRU	ALWR[b] Pu
U235	0.004	0.002	
U236	0.002	0.002	
U238	0.478	0.325	
NP237	5.023	6.641	
PU238	1.272	2.749	3.184
PU239	53.196	48.652	56.349
PU240	21.534	22.980	26.616
PU241	3.782	6.926	8.022
PU242	4.686	5.033	3.829
AM241	8.967	4.654	
AM242M	0.014	0.019	
AM243	0.926	1.472	
CM242	0.000	0.000	
CM243	0.002	0.005	
CM244	0.104	0.496	
CM245	0.009	0.038	
CM246	0.001	0.006	

[a]*Based on the medium burnup (33,000 MWd/MT) PWR benchmark with 25 years cooling*

[b]*Based on the high burnup (50,000 MWd/MT) PWR benchmark with 10 years cooling*

A quick and accurate modeling path was developed for estimating mass flows in the first tier ALWR transmutation system. A unit lattice (assembly) analysis approach was utilized. This is considered sufficient because the systems are assumed to employ a full-core loading of a single fuel form (e.g., 100% MOX fuel or NFF), in which case neglect of inter-assembly spectral interferences are not expected to invalidate calculated mass flows. A variety of thermal reactor lattice codes were evaluated for this application. The WIMS8 code [8] was selected because of the availability of a 172-group library constructed from JEF2.2 nuclear data library. The burnup chain models heavy metal isotopes from U-233 to Cm-245. Preliminary calculations for an OECD/NEA unit-MOX-cell benchmark problem indicated the need for a 172-group library for cases containing degraded Pu vector (i.e., with significant amounts of the higher Pu isotopes). It should be noted that when the predominant Pu isotopes are Pu-239 and Pu-240, the same study indicated that the 69-group library gives an accurate representation of the MOX cell, because the energy range containing their low energy resonances is sufficiently covered.

For all the ALWR cases, fuel assemblies are loaded in three batches and discharged at a reactivity-limited burnup of 51 GWd/t for the full-MOX cases and 510 GWd/t for the full-NFF cases; this corresponds to a core residence time of 4.5 years at about 80% capacity. By assuming a core neutron leakage of 3.5%Δk, the fresh fissile loading needed to meet the cycle specifications was derived by adjusting the TRU loading (%TRU/HM) in the MOX-fueled assembly such that the end of cycle (EOC) k_{inf} = 1.035. The EOC state was approximated in the lattice calculation by unpoisoned conditions (i.e. 0 ppm soluble boron) at 2/3 the discharge burnup (the core averaged burnup at EOC). This approach was also used for the NFF cases, except that in these cases the erbia (Er_2O_3) and heavy metal weight fractions in the fuel were varied, and compensated by the zirconia (ZrO_2) amount. Fission-product and heavy-metal masses at this discharge burnup are additionally modified in a zero power WIMS8 calculation to account for the cooling interval, which is assumed to be seven years for MOX fuel (PUREX) recycle or two years for pyro (remote) processing.

Finally, detailed mass flow data were generated for each case using the ORIGEN2 code. The WIMS8 run tracks all the pertinent heavy-metal nuclides and the fission products (about 100 of them) that account for 99% of the reactivity impact of the fission products. The ORIGEN2 run tracks a larger number of heavy-metal and fission product isotopes as well as light and structural elements. In the ORIGEN2 run, one of the existing cross-section libraries (for a plutonium recycle core) is employed however. The heavy-metal and fission product nuclides from the WIMS8 run replace those in the ORIGEN2 output; and the fission product masses are constrained to ensure that the total fission-product mass is consistent with the total heavy-metal consumed. Once these isotope masses have been replaced, the radioactivity and thermal decay power parameters are then recalculated.

The reference results for the GT-MHR Tier-1 options were based on detailed three-dimensional calculations with the Monte Carlo depletion code MONTEBURNS [9], which couples the Monte Carlo transport code MCNP with ORIGEN2.1. The model represents a GT-MHR system operating at 600MWt with 108 "fuel elements" in three annular rings with inner and outer rings of reflector blocks. Each fuel element was modeled explicitly with 216 fuel channels, and interspersed coolant passages; no burnable poison was assumed. Each fuel channel contained coated-particle fuel in a graphite matrix at a packing fraction of 2.5%, with discrete particles assumed to be arranged in a body-centered cubic array. The resultant total fuel loading in the core is 515kg.

The analyses assumed a single fuel batch; earlier studies show modest improvements (e.g., slightly higher burnup) are achievable with multi-batch fuel management. Burnup calculations were performed with MONTEBURNS until the value for k-eff dropped below 1.0; this defined the end-of -cycle burnup. MCNP is used to calculate one-group fluxes and cross sections for significantly contributing actinides and fission products in the middle of each of 5-6 burn steps and passes that information to ORIGEN2.1. ORIGEN2.1 completes the burnup and decay calculations for each burn step using those cross sections (along with default thermal reactor ones for isotopes

not tracked by MCNP) and fluxes. The detailed isotopics and decay parameters were obtained directly from the ORIGEN computations; and the calculation extended in decay mode for 2, 5, 10, 15 and 20 years.

Analysis of the fast spectrum ATW system has focused on the equilibrium fuel cycle. Transmutation performance characteristics were calculated using the REBUS-3 fuel cycle analysis code [10]. In the fuel cycle model, the charged fuel contains the transuranics recovered via recycle from the discharged ATW fuel, supplemented by either the Tier I discharge (in the double tier cases) or the commercial ALWR discharge TRU (single tier cases) to make up for the TRU consumed by fission. The external cycle model explicitly accounts for 0.1% loss of TRU in a 2-year turnaround processing time. In addition, 5% of the rare-earth fission products are carried over with the recycled fast system TRU. A TRU-40Zr fuel alloy was employed in the interior region, and "enrichment" zoning was achieved by utilizing a higher TRU fraction (~70%) in the outer region. An 8-batch scheme with semiannual refueling, staggered reloading of neighboring assemblies and no fuel shuffling is employed for the outer zone; the fuel residence time in the inner blanket zone is reduced to 7 cycles to limit the fluence.

Multigroup cross section data were generated in several spatial regions for a 21 energy group structure based on ENDF/B-V data using the MC2-2 code [11]. The burnup chain models actinides ranging from U-234 to Cm-248. The TRU mass loading was determined using the REBUS-3 enrichment search techniques to yield a targeted subcriticality level of 0.97 at BOEC; the fuel volume fraction was varied to achieve the desired TRU-40Zr inner zone composition. REBUS-3 also computes both batch-dependent and batch-averaged compositions at BOEC and EOEC for each specified depletion region. In this study, five (equal length) axial depletion zones were consistently used; in the planar dimension, depletion zones consisted of individual fuel assemblies or groups of neighboring assemblies with similar reaction rates. Irradiation swelling of the metal fuel was modeled in the depletion calculations as a uniform 5% axial expansion of the fresh fuel, based on experiments for ternary metal (uranium-based) fuel.

Homogeneous (eigenvalue) neutronic calculations performed using the hexagonal-Z nodal diffusion option of DIF3D [12] were employed as a basis for assessing the transmutation performance. Sensitivity studies in Ref. 13 demonstrated that the transmutation parameters agree well with more detailed source-driven computations. Inhomogeneous (source) calculations would however be required to accurately predict the power peaking of the ATW systems, particularly at high source strength (to compensate low EOEC neutron multiplication). Detailed isotopic mass flow data were generated using the ORIGEN-RA code. The actinide one-group cross sections and fluxes are obtained from the REBUS-3 depletion calculation.

For the transmutation fast reactor, the computational techniques are nearly identical to those described for the ATW fast spectrum system. The only differences of significance are 1) cross section data appropriate for a fertile fuel system were utilized, 2) the close-packed fuel pin lattice employed in the ALMR burner design [7] was retained, and 3) in the enrichment search, the fuel volume fraction was not varied, but rather the TRU to uranium ratio was changed to assure criticality throughout the equilibrium cycle.

The material flows for the fuel separations of the different cases were derived from the simulated spent fuel compositions and a consistent set of assumptions. For the processing of ALWR and MOX fuels, calculations were based on the use of aqueous solvent extraction technologies. The material flows for all other separations assumed pyroprocessing technologies. Uranium and transuranic recoveries of 99.9% via solvent extraction or pyroprocessing are considered achievable and were adopted in the calculations. The transuranic losses were distributed equally to the metal waste form (containing the cladding and hardware) and glass (from solvent extraction) or ceramic waste form (from pyroprocessing).

IV. BASIC MASS FLOW AND TRU CONSUMPTION PERFORMANCE

Three primary fuel cycle approaches were evaluated in this study; double tier systems with plutonium separation, double tier systems without plutonium separation, and single tier (fast spectrum) systems. The double tier strategies utilize either the transmutation ALWR or GT-MHR designs described in Section II for the initial irradiation; furthermore, both MOX and NFF forms were considered for the ALWR. This results in three specific cases with plutonium separation in the first tier, where the minor actinides are diverted to the second tier in the initial processing. Another three cases were considered without plutonium separation where all TRUs enter the first tier system. In each case, a second tier fast spectrum system with repeated recycle was employed to destroy the remaining TRU. A fast spectrum ATW design was considered in all cases; and a fast reactor design was evaluated for comparison purposes for the MOX fuel with plutonium separation case. Finally, the single tier strategies utilize either the ATW or fast reactor designs described in Section II; in this approach, the TRUs enter directly into the fast spectrum recycle system without prior thermal reactor irradiation.

The technology and fuel form combinations for the nine specific cases noted above are shown in Table 2, and the key transmutation performance results are also summarized. For the double tier approaches, the nonfertile options destroy more than 50% of the TRU mass in a single pass, whereas, the fertile MOX fuel only destroys ~20% per stage (net destruction of 1/3 for the double pass utilized in this study). The first tier irradiation also gives a significant increase in higher actinide content; for example, the Cm-244 content increases by a factor of 3-5 as a result of Tier 1 irradiation. These buildups are exacerbated in the second approach (without plutonium separation) because the minor actinides are included in the system feed. It was also observed that the first tier is a very

Table 2. Summary of System Mass Flows and Transmutation Performance

Parameter		Approaches								
		Double Tier With Plutonium Separation				Double Tier Without Pu Separation			Single Tier	
		ALWR			GT-MHR	ALWR		GT-MHR		
Tier 1	Fuel Form	MOX	MOX	NFF	TRISO	MOX	NFF	TRISO		
	Reactor thermal power (MWt)	3000	3000	3000	600	3000	3000	600		
	Cycle length, EFPD	443.5	443.5	436.8	500.0	443.5	436.8	400.0		
	Fuel form	MOX	MOX	NFF	TRISO	MOX	NFF	TRISO		
	BOC heavy-metal inventory (t)	76.94	76.94	6.38	0.61	76.94	6.38	0.61		
	Charge per batch	Stage 1	Stage 2							
	Heavy metal (HM) (t)	26.10	26.10	2.57	0.61	26.10	2.57	0.61		
	Fissile in HM (%)	6.33	7.69	64.37	64.37	10.76	55.64	55.64		
	Consumption per batch	Stage 1	Stage 2							
	TRU (%)	**21.7**	**16.4**	**51.6**	**57.4**	**13.9**	**51.8**	**46.0**		
	Fissile (%)	43.3	31.0	74.0	84.6	27.0	78.3	78.1		
	Discharge burnup (MWd/kg)	51	51	510	591	51	510	470		
Tier 2	System Type	ATW	F-Rx	ATW	ATW	ATW	ATW	ATW	**ATW**	**F-Rx**
	Reactor total power (MWt)	840	840	840	840	840	840	840	840	840
	TRU in charge heavy metal (%)	98.7	98.7	98.5	98.6	99.0	97.4	97.5	98.5	32.4
	BOEC HM inventory (kg)	3401	13807	3662	**4070**	2825	3555	3666	2709	13894
	TRU Destruction Rate (kg/y)	232	**140**	232	232	232	230	230	231	**128**
	Burnup reactivity loss (%k)	2.77	1.84	2.53	**2.09**	3.93	2.91	2.71	**4.14**	2.34
	Batch TRU consumption (%)	*24.0*	*17.5*	*22.5*	***20.5***	*28.2*	*23.2*	*22.6*	***29.2***	*18.6*

effective consumer of the fissile material (up to 85% consumed in a single pass) resulting in discharged fuel with a factor of 2-3 lower fissile content than TRU contained in typical LWR spent fuel.

As shown in Table 2, the reduced fissile content of the first tier discharge impacts transmutation performance in the second tier. At reduced fissile content, more TRU must be loaded into the fast spectrum system leading to a lower discharge burnup compared to the single tier approach. This is detrimental because more recycle passes (with associated losses) will be required to achieve complete destruction of the remaining TRU inventory. Conversely, the low fissile inventory has some benefits for second tier performance; in particular, the burnup reactivity loss rate decreases from 4%Δk to as low as 2%Δk.

Results in Table 2 also illustrate the reduced TRU destruction rate (for a fixed system capacity) when fertile fuel is utilized. Compared to the ATW system, the fast reactor (F-Rx) single tier approach yields a TRU destruction rate about half of the nonfertile fuel case, as expected. This implies that the fertile-fuel system will require twice the dedicated capacity to achieve the same TRU destruction rate, but would produce about twice the energy output in the process. Furthermore, conventional fuel burnup (and fluence) limits constrain the net consumption of TRU to less than 20% before the material must be recycled, as compared to 21-29% per pass destruction for the nonfertile fuel cases. This implies that the fertile-fuel system will require additional fuel separations steps to transmute a given amount of TRU.

V. COMPARISONS OF SYSTEM TRANSMUTATION PERFORMANCE

The fuel cycle performance of the nine specific scenarios identified in Table 2 was evaluated in detail as described in Section III; this included detailed tracking of the material composition and properties throughout the entire transmutation fuel cycle (from commercial spent fuel to final waste forms). Based on this data, each scenario was assessed against a variety of criteria: radiotoxicity reduction, long-term dose reduction, long-term heat load, plutonium reduction, waste mass and volume reduction, system support ratio, and worker exposure. The key modeling assumptions and performance objectives for these criteria are discussed in more detail in a companion paper [1]. In this paper, the waste (and plutonium) mass reduction, radiotoxicity, and support ratio results are given.

Table 3. Comparison of Once-Through and Transmutation Waste Inventories

Parameter	Once-Through ALWR Spent Fuel	Approaches								
		Double Tier With Plutonium Separation				Double Tier Without Pu Separation			Single Tier	
		ALWR			GT-MHR	ALWR		GT-MHR		
Tier 1 Fuel Form	Enriched UO2	MOX	MOX	NFF	TRISO	MOX	NFF	TRISO		
Tier 2 System	-	ATW	F-Rx	ATW	ATW	ATW	ATW	ATW	**ATW**	**F-Rx**
Estimated Annual Waste Mass, MT										
Transuranics	**1.49E-2**	6.97E-5	8.56E-5	5.14E-5	5.13E-5	7.33E-5	4.58E-5	5.04E-5	5.09E-5	7.94E-5
% Reduction	-	99.5	99.4	99.7	99.7	99.5	99.7	99.7	99.7	99.5
Plutonium	**1.29E-2**	5.28E-5	6.75E-5	3.84E-5	3.84E-5	6.15E-5	3.61E-5	4.03E-5	4.25E-5	6.93E-5
% Reduction	-	99.6	99.5	99.7	99.7	99.5	99.7	99.7	99.7	99.5
Fission Products	**5.43E-2**	6.38E-2	6.88E-2	5.79E-2	5.78E-2	6.01E-2	5.77E-2	5.77E-2	5.79E-2	7.42E-2
% Increase	-	17.5	26.8	6.6	6.4	10.7	6.3	6.2	6.7	36.7

V.1 ANALYSIS OF WASTE INVENTORY

The basic approach of the transmutation scenarios is to recycle the TRU materials for eventual destruction by fission. However, at each recycle/refabrication step a small amount (assumed at 0.1% in this study) is not recovered and lost to the waste. Thus, given the burnup levels for each batch ranging from 20-50%, one would expect 0.2-0.5% of the TRUs to escape transmutation. Furthermore, additional fission products are being produced by TRU fission in each of the transmutation systems. The estimated waste mass of transuranics, plutonium, and fission products for each case is compared to the ALWR once-through fuel cycle in Table 3; these results are normalized to an input stream of 1 MT of ALWR spent fuel.

This table indicates that all of the transmutation scenarios have been quite effective in achieving their primary transmutation goals. More than 99.5% of the TRUs have been transmuted, which will result in severely decreased waste toxicity (see Section V.2) and also results in improved repository performance [2]. Furthermore, the proliferation concerns associated with the repository will also be improved as the plutonium content of the waste has been reduced to less than 0.5% of the once-through fuel cycle.

It is important to note that the fission product mass estimates for the transmutation scenarios in Table 3 do not include any of the fission product gases; they are released as part of the spent fuel processing. Conversely, these fission products are included in the once-through cases where they would remain inside the fuel pins that are directly disposed. Even with this reduction in fission product mass, the transmutation scenarios contain at least 6% more fission product mass than the once-through cycle. The mass increase would be more on the order of 30-75% (reflecting the power capacity of the transmutation systems, see Section V.3) if the fission product gases were included in the transmutation scenario waste estimates. The fertile fuel cases show much larger increases in fission product waste mass; this is attributed to their lower TRU destruction rates (as shown in Table 2) which imply that more power producing systems will be required to achieve the same net TRU destruction.

V.2 ANALYSIS OF RADIOTOXICITY

Based on the detailed isotopic waste inventories for each scenario, the waste radiotoxicity was evaluated and compared to the once-through ALWR fuel cycle. Isotopic ingestion toxicity factors were derived from the most recent ICRP equivalent commited dose data [14]. All results are normalized to the radiotoxicity of the natural uranium ore required to produce one metric ton of initial ALWR fuel; 7.5 metric tons of ore are required to produce one metric ton of 4.2% U-235/U low enrichment fuel. The radiotoxicity is explicitly evaluated for the entire spectrum of TRU and fission products discharged to the waste, as summarized in Table 3. The contribution of the uranium discharged to an alternate (low level) waste stream in the initial separations was neglected.

The key trends in the radiotoxicity results are illustrated in Fig. 1; results are shown for the double tier case without plutonium separation where non-uranium fuel is utilized in the transmutation ALWR (with a second tier ATW). The toxicity of the transmuted waste falls below the level of the natural ore after roughly 400 years; at this point in time, the radiotoxicity of the once-through spent fuel is ~200 times greater. This toxicity reduction quantifies the benefit of the significant reduction in the TRU inventory, as shown in Table 3. Also shown in Figure 1 is the toxicity of the material following irradiation in the thermal spectrum Tier 1 system. This curve is only slightly lower than the once-through spent fuel toxicity;

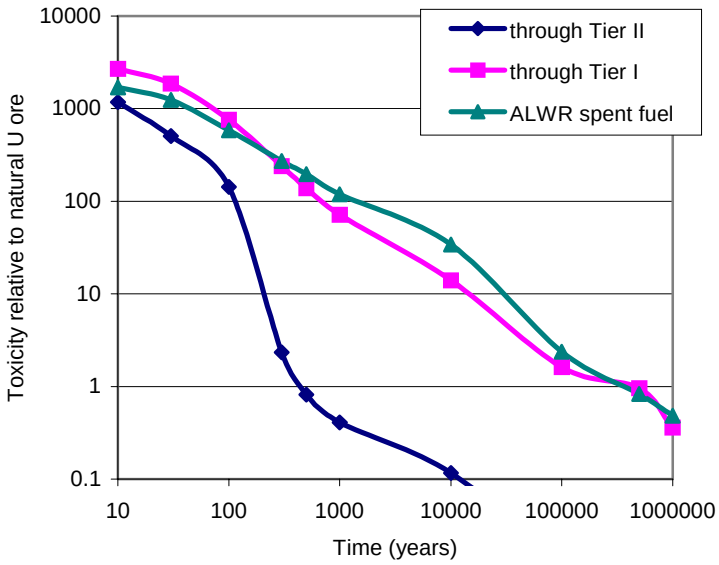

Figure 1. Radiotoxicity Comparison of ALWR Spent Fuel and Transmuted Wastes

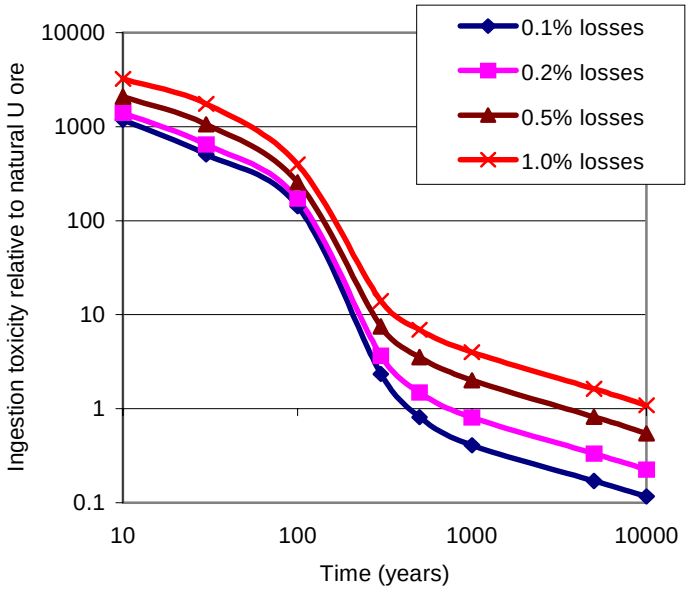

Figure 2. Impact of Loss Fraction on Waste Radiotoxicity

thus, the Tier 1 irradiation has provided little progress towards reducing the radiotoxicity. This behavior is attributed to the buildup of higher actinides during irradiation in a thermal spectrum; although nearly half of the TRUs have been consumed in the first tier, additional higher actinides (which tend to be more radiotoxic) have been created. This behavior is inhibited in a fast spectrum where direct fission (avoiding capture to higher actinides) is more likely.

All of the transmutation scenarios identified in Table 2 show behavior similar to the results in Fig. 1. The radiotoxicity falls below the level of the natural uranium ore after 400-700 years of cooling, given the 0.1% processing loss rate. The vital importance of the fuel processing/fabrication loss fraction can be illustrated by comparing waste radiotoxicity for a parametric variation of loss fraction; results are shown in Fig. 2. At higher loss fraction, additional TRUs (beyond the waste inventory given in Table 3) are discharged to the waste, increasing the radiotoxicity associated with the TRU. The total toxicity falls below the natural ore level at ~500 years for the 0.1% loss fraction, ~1000 years at 0.2%, and extends beyond 10,000 years at 1% loss fraction using the same transmutation system.

V.3 ANALYSIS OF SUPPORT RATIO

In this study, it was assumed that a commercial nuclear sector exists that produces transuranics (TRU) that are to be burned in both Tier 1 and/or Tier 2 nuclear systems. It was additionally assumed that the commercial sector is similar to the LWR industry currently existing in the U.S. and that the sector has a capacity of about 100 GWe, equivalent to ~300 GWt. The goal is to balance the TRU accumulated in this commercial sector (26.3 MT/year) with that destroyed in the transmutation systems in order to stabilize the TRU inventory (i.e., to attain an equilibrium mode in which no additional TRU is added to the TRU stockpile). Based on this premise, the power required in each tier was evaluated for each transmutation scenario and the results are shown in Table 4.

The support ratio has previously been defined in many different ways; the two dominant definitions were applied here. The first definition, denoted the transmutation support ratio, is the ratio of power generated in the commercial sector (300 GWt at 85% capacity factor, 255 GWt per year) to the total power required for the transmutation enterprise. As shown in Table 4, this support ratio is 3.6-3.8 for all options utilizing non-uranium fuel exclusively. In these cases, no new TRU are formed and the ratio can be verified by considering the ALWR spent fuel. At discharge the commercial spent fuel contains 5.3 weight% fission products compared to a TRU content of 1.4%; thus, the ultimate fission product ratio must be 5.3/1.4 = 3.8. When fertile fuel is utilized in either the first or second tier, additional TRUs are created requiring additional fissions for their destruction; this results in lower support ratios, particularly when fertile fuel is utilized in the repeated recycle Tier 2 system. However, one would expect the power produced in these Tier 2 systems to be more reliable and economical than a nonfertile ADS system, so having a *high support ratio is less crucial for the Tier 2 fast reactor* cases.

An alternate definition for the support ratio, denoted the Tier 2 support ratio, is defined as the power generated in thermal reactor systems (commercial and Tier 1) to the power required in the final fast spectrum transmutation phase (Tier 2). This ratio quantifies the relative capacity of the final Tier 2 transmutation systems required to complete the mission and is typically used when one expects power production in the Tier 2 system to be much more expensive than either commercial systems *or* TRU-fueled thermal reactors. Thus, this definition is a key design parameter for ADS Tier 2 systems, but is less

Table 4. Comparison of Power Requirements and Support Ratios

Parameter	Approaches								
	Double Tier With Plutonium Separation				Double Tier Without Pu Separation			Single Tier	
	ALWR			GT-MHR	ALWR		GT-MHR		
Tier 1 Fuel Form	MOX	MOX	NFF	TRISO	MOX	NFF	TRISO		
Capacity Factor	0.85	0.85	0.85	0.85	0.85	0.85	0.85		
TRU loading, MT/y	1.62	2.48	1.83	0.38	3.33	1.82	0.47		
Power Required in Tier, GWt	36.1	18.4	31.9	30.8	20.2	37.0	28.5		
Tier 2 System	ATW	F-Rx	ATW	ATW	ATW	ATW	ATW	**ATW**	**F-Rx**
Capacity Factor	0.75	0.85	0.75	0.75	0.75	0.75	0.75	0.75	0.85
TRU fraction to Tier 2	0.712	0.712	0.551	0.499	0.860	0.481	0.538	0.999	0.996
Makeup TRU loading, MT/y	0.233	0.144	0.232	0.232	0.233	0.230	0.231	0.232	0.128
Power Required in Tier, GWt	50.8	93.3	39.4	35.7	61.4	34.7	38.7	71.6	146.3
Transmutation Support Ratio	**2.4**	**1.7**	**3.6**	**3.8**	**3.1**	**3.6**	**3.8**	**3.6**	**1.7**
Tier 2 Support Ratio	**6.1**	**3.3**	**7.3**	**8.0**	**4.5**	**8.4**	**7.3**	**3.6**	**1.7**

applicable to fast reactor systems (if costs can be achieved comparable to thermal systems). As shown in Table 4, the Tier 2 support ratio is a factor of ~2 higher for the double tier cases since roughly 50% of the TRU was consumed in Tier 1 thermal reactor systems. These results quantify the effectiveness of the double tier strategy in reducing the number of Tier 2 systems required. Once again, the high burnup nonfertile fuel cases results in the highest support ratios. The highest observed support ratio is 8.4 in the case using the GT-MHR without plutonium separation. With a deeper burnup in Tier 1 it may be possible to increase the support ratio to as high as ~10. However, this clearly demonstrates that *a significant transmutation enterprise is necessary* to support a nuclear future assumption; at a support ratio of 10, the Tier 2 systems would have a power capacity of ~30 GWt for a constant nuclear power production scenario.

Differing definitions and interpretations of the support ratio arise because the cost differences between the diverse nuclear units are not explicitly accounted by simple measures such as the ratio of power requirements. In a study of this nature, a cost benefit analysis of the integrated transmutation fuel cycle is the appropriate basis for differentiation between options. *It is evident that in the absence of such evaluations, a better measure of the support ratio should include cost factors that differentiate between the transmutation systems.* This item should be addressed in future systems studies.

V. CONCLUSIONS

A variety of double and single tier transmutation systems, including thermal and fast spectrum systems using conventional and nonfertile fuel forms and their associated fuel processing, were evaluated in a consistent manner. Given clean fuel processing (0.1% losses), all scenarios were capable of reducing the TRU and plutonium losses to the waste to less than 0.6% of the once-through fuel cycle waste. This reduction in TRU content was observed to significantly impact the waste toxicity, reducing it to the level of natural ore in less than 1,000 years. It was demonstrated that the toxicity reduction is very sensitive to the processing loss fraction. Furthermore, the Tier 1 irradiation was shown to have little impact on the toxicity, confirming the need for a final tier fast spectrum system to complete the transmutation campaign.

The utilization of a first tier thermal spectrum system was demonstrated to increase the Tier 2 support ratio; this would reduce the number of ATW systems required to complete the transmutation mission. However, even in the double tier scenarios the Tier 2 capacity requirements are large (~1/10 the total nuclear generating capacity), indicating that a significant infrastructure will be required to achieve the transmutation goals. No significant differences were observed between ALWR and thermal spectrum GT-MHR system performance; this is attributed to the comparable burnup levels achieved in the non-uranium fuel options addressed in this study.

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