COMPARATIVE FUEL CYCLE ANALYSIS OF CRITICAL AND SUBCRITICAL FAST REACTOR TRANSMUTATION SYSTEMS

RADIOACTIVE WASTE MANAGEMENT AND DISPOSAL

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Fuel cycle analyses are performed to evaluate the impacts of further transmutation of spent nuclear fuel on high-level and low-level waste mass flows into repositories, on the composition and toxicity of the high-level waste, on the capacity of high-level waste repositories, and on the proliferation resistance of the high-level waste. Storage intact of light water reactor (LWR) spent nuclear fuel, a single recycle in a LWR of the plutonium as mixed-oxide fuel, and the repeated recycle of the transuranics in critical and subcritical fast reactors are compared with the focus on the waste management performance of these systems. Other considerations such as cost and technological challenges were beyond the scope of this study. The overall conclusion of the studies is that repeated recycling of the transuranics from spent nuclear fuel would significantly increase the capacity of high-level waste repositories per unit of nuclear energy produced, significantly increase the nuclear energy production per unit mass of uranium ore mined, significantly reduce the radiotoxicity of the waste streams per unit of nuclear energy produced, and significantly enhance the proliferation resistance of the material stored in high-level waste repositories.

I. INTRODUCTION

There is a substantial worldwide research and development activity devoted to the transmutation of spent nuclear fuel (SNF) (e.g., Refs. 1 through 4). The objective of this activity is to technically evaluate the possibility of reducing the requirements for long-term geological repositories for the storage of high-level radioactive waste (HLW) from SNF, by neutron fission of the plutonium and higher actinides remaining in the spent fuel discharged from fission power reactors. Repeated recycling of the transuranics from SNF in special purpose fast spectrum reactors could reduce the toxicity of the spent nuclear fuel by a factor of ~100, limited by safety and criticality constraints.[1] These constraints could be relaxed if the reactors (fast or thermal spectrum) could be operated subcritical, which would require a neutron source. There is a general consensus that significantly higher rates of net actinide destruction can be achieved by repeated recycling of the transuranics from SNF in subcritical reactors driven by an external neutron source. An accelerator-spallation neutron source has been extensively studied for this application (e.g., Refs. 1 through 7), and deuterium-tritium (D-T) fusion neutron sources have recently received increased attention for this purpose (e.g., Refs. 8 through 13).

An HLW repository at Yucca Mountain, Nevada, is currently being developed by the U.S. Department of Energy (DOE), Office of Civilian Radioactive Waste Management, which is charged with disposing of all SNF from commercial nuclear reactors and HLW resulting from atomic energy defense activities.[14] The once-through fuel cycle (OTC) is the baseline scenario for the proposed repository. In this scenario, the SNF discharged from light water reactors (LWRs) would be placed in specially designed containers and stored intact in a repository after a cooling period.

The "Integrated Data Base Report"[15] gives a summary of the U.S. SNF inventories and projections. The current inventory of discharged SNF has an average burnup of ~33 GWd/MTU, which has been consistently increasing over recent years and is projected to increase further in the future. The current installed capacity of LWRs is ~100 GW(electric), approximately two-thirds of which are pressurized water reactors (PWRs) and the remaining one-third are boiling water reactors. Over time, the LWRs have operated at increasing

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efficiencies and with higher initial enrichments, and have thus produced SNF with increasing discharge burnup. This trend will likely continue, resulting in a continued evolution of the composition of the discharged SNF.

The inventory of discharged SNF is estimated to be over 47 000 metric tonnes of initial uranium (MTU) by the end of 2002. Ongoing operation at the current nuclear power production level will produce over 2000 MTU of additional SNF each year. The proposed repository at Yucca Mountain has a statutory limit of 70 000 metric tons of heavy metal (MTHM), which includes 63 000 MTU of commercial SNF. At current levels of production, the discharged SNF will exceed this limit around 2010. Either legislation will be required to increase the legal capacity, or a second repository will be required. Even the most pessimistic predictions about the future of nuclear power in the United States project this statutory limit being exceeded by a large amount early in the century. Even if statutory limits are removed, Yucca Mountain has a finite capacity, and the limit will be exceeded in the not too distant future.

At current levels of nuclear power production, a new repository with the statutory capacity of Yucca Mountain would be required every 34 yr. If the United States were to exit the commercial nuclear power business, the SNF could be stored above ground for extended periods of time and eventually be placed in one or two repositories. On the other hand, a steady or growing level of nuclear power production will ultimately require a large number of HLW repositories and eventually deplete the currently very cheap supply of fissile ^{235}U and future supplies of ^{239}Pu from excess nuclear weapons. Transmutation is a potential solution to both of these problems that deserves consideration.

The waste management solution in other industries that produce large quantities of hazardous waste materials is typically a combination of solutions including recycle, incineration, and immobilization. Solely immobilization is relied upon in the OTC scenario. Expanded nuclear fuel cycles could reduce the quantity of hazardous wastes by recycling and reduce the hazard and uncertainty by "incinerating" the hazardous and problematic components of the waste. Additionally, the chemical separation of the waste streams would allow for a more effective immobilization of the residual waste. Unlike nonradiological hazardous wastes, radiological wastes are only hazardous for a finite amount of time. Therefore, immobilization can be effective in eliminating some or all of the hazardous material, particularly short-lived components. Nonradiological hazardous wastes will eventually reenter the environment, and immobilization attempts to ensure that the rate is sufficiently slow to preclude harm. In the case of radiological waste, reducing the concentration of the long-lived isotopes in the waste increases the likelihood that the rate at which these wastes are reintroduced into the environment is sufficiently slow to be acceptable.

The transmutation portion of the nuclear fuel cycle is for waste management purposes and is akin in function to a hazardous waste incinerator. As with the hazardous waste incinerator, the primary goal of transmutation is to significantly reduce the hazard, real or perceived, of the feed material. An important secondary goal is to utilize the resulting energy to offset some of the cost of the incinerator.

Transmutation is generically the conversion of problematic isotopes to less problematic isotopes. The reasons isotopes are considered problematic are varied, but generally all transuranic isotopes, along with certain long-lived fission product (FP) isotopes, may be considered problematic. Fissioning of the transuranic isotopes essentially converts them into FP, most of which are short lived. Further transmutation of long-lived FP isotopes would convert them to other less problematic isotopes.

Radioactive waste management involves both the HLW streams that will be disposed in geologic repositories, as well as the low-level waste (LLW) streams that are disposed in near-surface burial facilities. Transmutation systems would change the composition and quantities of materials that are disposed in both types of disposal systems. In addition, any transmutation system would include a substantial chemical separation system to support the recycle of the materials being transmuted. Thus, with transmutation, the final waste form(s) could be tailored to more effectively immobilize the radioactive waste. By incorporating recycle, incineration, and immobilization, the waste management system for the nuclear fuel cycle would be very different and presumably superior as a result of separation and transmutation.

In order to evaluate a transmutation system, measures of the performance must be defined. All components of any nuclear fuel cycle are required to satisfy all regulatory requirements. The difficulty of satisfying these regulatory requirements will directly affect the cost of the system and hence its relative attractiveness. Figures of merit (FOMs) have been developed to enable a quantitative comparison of different transmutation systems with the OTC.

There are many technical and nontechnical issues associated with the disposal of radioactive waste. In order to cover a wide range of issues and provide a broad set of indicators, the FOMs were chosen to address issues related to separations, HLW disposal, LLW disposal, repository performance, shipping, proliferation, public perception, and cost. In this study, the parameters evaluated relate to the technical issues (e.g., mass flow, energy production, toxicity, repository requirements, and risk of proliferation from HLW). This analysis and its conclusions focus on the waste management performance of these systems. The issues of cost, public perception, implementation, etc., are not evaluated, and no conclusions are drawn about these very important considerations.

The waste management FOMs that are evaluated in this study focus on four areas: (a) size, (b) waste toxicity, (c) repository performance, and (d) proliferation resistance of the HLW. The overall size and time frame of the varied systems are quantified in terms of mass flow and energy production. Toxicity is a simple measure commonly used to evaluate the radiological impact of transmutation systems, but toxicity does not necessarily determine which waste streams will be more difficult to effectively isolate. Therefore, FOMs were evaluated that attempt to quantify the impact on repository performance. The repository performance FOMs are related to the heat load and dose to the public from dissolution of the waste at distant future times. In addition, concerns have been raised about proliferation of materials from waste placed in repositories, and a number of parameters are evaluated to assess the impact transmutation will have on the potential for proliferation of waste materials.

Based on these FOMs, the radioactive waste disposal characteristics of the following different fuel cycles were compared:

1. the reference OTC with SNF stored intact in an HLW repository
2. a single recycle in an LWR of the plutonium from the SNF as a mixed-oxide (MOX)
3. a repeated recycle of all transuranics from the SNF in liquid-metal–cooled, metal fuel fast reactors.

Three different fast reactors were considered:

1. a sodium-cooled critical reactor with fuel containing transuranics and fertile ^{238}U [Integral Fast Reactor (IFR)]
2. a lithium lead-cooled subcritical reactor with fuel containing only transuranics driven by a fusion neutron source [Fusion Transmutation of Waste Reactor (FTWR)]
3. a sodium-cooled, subcritical reactor with fuel containing only transuranics driven by an accelerator spallation neutron source [Accelerator Transmutation of Waste Reactor (ATWR)].

The fuel cycles that are evaluated are described in more detail in Sec. II, and the methodology used to evaluate the equilibrium fuel cycles is given in Sec. III. A comparison of the mass and energy parameters is given in Sec. IV. The toxicity for both the HLW and for all waste streams (HLW and LLW) is discussed in Sec. V. A comparison of the impact on the design of the HLW repository is provided in Sec. VI, and the proliferation attributes of the HLW are discussed in Sec. VII. More detailed information on the three fast reactors is provided in Appendixes A, B, and C.

II. FUEL CYCLES

The focus of this study is on the waste management performance of the transmutation system. In order to evaluate the different transmutation systems, the entire nuclear fuel cycle must be analyzed. Nuclear fuel cycles based on an initial irradiation of low-enriched uranium in commercial LWRs were evaluated. Figure 1 shows the material flow for the fuel cycles considered in this paper. In most cases, a wide variety of technologies exist that can perform each step in this diagram. For example, the transmutation systems can be based on critical or subcritical reactors utilizing a variety of coolants and fuel designs and with different neutron spectra. The flow of nuclear materials, uranium, and its nuclear reaction products are shown with a number of decision points about how to treat major components of this material indicated.

The materials are divided into several major groups. All begin with the uranium ore, which for the purposes of this analysis is the natural uranium (NU) in equilibrium with its radioactive daughter products, and all nonradioactive components of the ore are neglected. If the analysis is expanded to include nonradioactive components, the grade of the ore will become significant. Mill tails are defined in this analysis as the radioactive daughter products in equilibrium with the NU that is mined from the earth. The NU is separated into the enriched uranium (EU) and depleted uranium (DU). The EU is used to produce the commercial LWR fuel. The DU is mostly LLW with a small fraction used in some fuel cycles. The SNF is the irradiated EU and its daughter

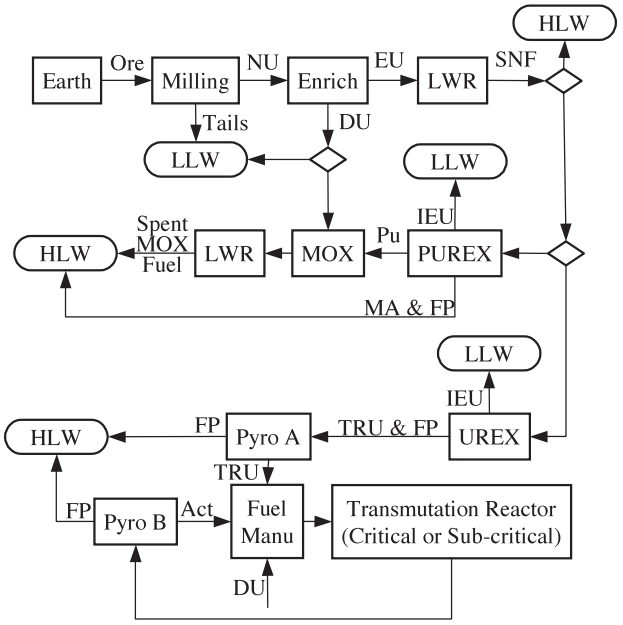

Fig. 1. Fuel cycle material flow diagram.

transmutation products that are discharged from commercial LWRs. The SNF can be chemically separated into a number of streams, and for this study the material streams included the residual irradiated enriched uranium (IEU), plutonium, minor actinides (MA), and FP. The Pu and MA are not separated from each other in some fuel cycles in which all transuranic elements (TRU) are handled together as a group. The FP streams are essentially all waste materials that are not or cannot be recovered and will include small amounts of other materials such as TRU. Any material not sent for further processing will be sent to an HLW repository or an LLW facility.

The treatment of the FP is simplified in this analysis. The FP are treated as a single stream that will be placed in an HLW repository. In fact, there will be multiple streams containing FP. Since the focus is on the aggregate effects of transmutation and specifically on the HLW repository, this treatment results in the maximum quantity of FP being placed in the repository and neglects FP that are sent to the LLW or released to the environment. In addition, activated components, which have the potential to be classified as HLW, are not included in this analysis. This is an area that clearly will require consideration before any final conclusion can be drawn.

The SNF from commercial LWRs is the basis for all subsequent fuel cycles. Since commercial LWRs will produce an evolving composition of SNF, it was necessary to make assumptions about the composition of the SNF that will feed the transmutation systems. Two different options are typically evaluated. The first uses the SNF representative of the average SNF that has been discharged and is currently in temporary storage. This is a relatively low burnup SNF with a fairly long cooling time. The second uses SNF that estimates the eventual average properties of the SNF discharged from commercial LWRs after their performance has evolved to some future optimum operation. This is a higher burnup SNF with a minimum cooling time. These constitute two significantly different SNF feed streams. The waste placed in Yucca Mountain will be of this first type, and it might be expected that the oldest SNF at shutdown reactors would be processed first. Eventually, the large backlog of discharged SNF would be processed, and over time the feed stream would evolve from the first type to the second. *A SNF composition representative of the current inventory of SNF in temporary storage was chosen for the analysis of this paper.* This decision has a relatively small impact in fast neutron spectra but would be more significant in the single MOX recycle case.

The OTC scenario, as indicated in the first horizontal line in Fig. 1, involves the irradiation of slightly enriched uranium oxide fuel in LWRs and the disposal of the intact SNF in an HLW repository after discharge from the LWRs. In the other, "MOX" or "transmutation" fuel cycles, this SNF will provide the feed stream for the MOX or transmutation fuel cycles.

The OTC scenario begins with the uranium ore being mined and separated (assumed perfectly) into an LLW stream containing the mill tails and the NU. The NU is then processed (no losses assumed) to produce an EU stream for irradiation in the LWR and an LLW stream of DU (0.2% ^{235}U).

In order to begin transmuting the backlog of SNF as quickly as possible, utilizing existing commercial reactors in the transmutation mission would seem to be a logical part of the nuclear fuel cycle. MOX fuel is used in many countries, and the United States is currently developing a program to partially destroy, degrade, and secure surplus weapons-grade plutonium (WGPu) by irradiation in MOX fuel in commercial LWRs. By recycling the Pu in the SNF in existing LWRs, the bulk of the TRU would be reused, producing more energy, which would offset the production of additional Pu because less EU fuel would be used in the production of a given quantity of energy. Plutonium recycle would have other effects, such as the production of more MA, and would constitute a significantly different feed stream to any subsequent transmutation system.

The first variant of the OTC scenario that was analyzed was a single recycle of Pu from the LWR SNF back to the LWRs as MOX fuel as shown in the second horizontal line of Fig. 1. The spent MOX fuel would then be disposed in an HLW repository. In this MOX fuel cycle scenario, the representative SNF is separated into three streams using a chemical process such as plutonium-uranium extraction (PUREX). The three streams are

1. The residual IEU is recovered and sent to an LLW facility.

2. The FP and MA are recovered and sent to an HLW repository.

3. The Pu is recovered and blended with DU to produce the MOX fuel.

The separations for this fuel cycle are assumed to be perfect.

In order for transmutation to have a dramatic impact on the waste management from the nuclear fuel cycle, all TRU would need to be nearly completely destroyed. Only a partial reduction of the TRU inventory results from recycling Pu as MOX fuel in an LWR. Complete transmutation systems that repeatedly recycle all TRU to ultimately fission all but the small fraction of TRU (which leaks into the waste streams) are required. These systems can be either "nonfertile" systems that contain essentially zero ^{238}U or "fertile" systems containing ^{238}U but designed for conversion ratios substantially less than unity. The actinide composition for these systems, whether fast or thermal, will be very different from existing reactors because of the much higher concentration of TRU. A much higher fraction of the TRU will be MA, and

there will be a much lower concentration of the conventional fissile isotopes such as ^{235}U, ^{239}Pu, and ^{241}Pu.

Although transmutation systems may be based on thermal or fast neutron spectrum reactors, the probability of fission (hence actinide destruction) per neutron absorbed is generally larger in a fast spectrum.[16] Liquid-metal–cooled, metal-fueled fast reactor systems were chosen for comparison with the FTWR that we have developed.[12,13] Fuel cycles were analyzed based on the three transmutation systems (FTWR, ATWR, and IFR) that would completely transmute the TRU in the SNF discharged from the LWRs by repeated reprocessing and recycling. The FTWR, ATWR, and IFR use metallic actinide/zirconium fuel and liquid-metal coolants and operate with fast neutron spectra. *The FTWR and ATWR are subcritical reactors using nonfertile fuel, and the IFR is a critical reactor using a fertile fuel with a low conversion rate in a critical reactor.* The IFR fuel cycle utilizes a small fraction of the DU to produce the fertile fuel.

These FTWR, ATWR, and IFR fuel cycles all use the same processing/separation technology.[17] Figure 1 shows the chemical separation systems for the transmutation systems. LWR SNF is used as the feed material to make up the fissioned TRU. The performance of the SNF processing was taken from Ref. 17. The SNF is first processed in an aqueous uranium extraction (UREX) process that is assumed to recover 99.995% of the IEU, which is sent to an LLW facility. All residual materials are assumed to be sent to a pyrochemical processing facility (Pyro A) for purification of the FP from the TRU, which includes the 0.005% IEU. The Pyro A process recovers 99.9% of the actinides (including uranium), removes 95% of the rare earth FP, and removes 100% of all other FP. The separated FP along with the 0.1% of actinides leaking from the Pyro A process are sent to an HLW repository. The discharged fuel from the transmutation reactors is sent to a separate pyrochemical processing facility (Pyro B) designed to process the metal fuel. Pyro B is assumed to operate with the same performance parameters as Pyro A.

III. EQUILIBRIUM CYCLE CALCULATION METHODOLOGY

Two thermal light-water–cooled systems and three fast liquid-metal–cooled systems were analyzed for this paper. The techniques used to predict the equilibrium mass flow and isotopic composition of the systems are described in this section.

The depletion of the LWR fuel, both the EU and MOX fuels, was simulated using the SAS2H Module of the SCALE 4.4 code package.[18] The SAS2H Module performs one-dimensional neutron transport analyses of the reactor fuel assembly using the larger unit-cell (assembly) within an infinite lattice. In the SAS2H Module, time-dependent nuclide cross sections are used in a point-depletion to determine the burnup-dependent fuel composition used for the next spectrum calculation. The 33-group ENDF/B-V library was used, and the cross sections of all isotopes (128 heavy isotopes and 879 FP) were updated after each transport calculation.

The composition of this SNF depends upon many parameters including fuel design, power density, fuel enrichment, and burnup. The representative SNF used in this study was PWR fuel irradiated at a power density of 39.8 MW/MTU to 33 GWd/MTU. The reference SNF composition used in this analysis is representative of the current inventory of discharged SNF evaluated for the Yucca Mountain Project[19] (YMP).

The PWR fuel was a standard 17×17 fuel design with 264 fuel pins and 25 guide tubes. The fuel pins have an outer diameter of 0.950 cm, a clad thickness of 0.057 cm, a fuel radius of 0.410 cm, and a pitch of 1.260 cm (Refs. 15 and 20). The fuel is uranium dioxide with the enrichment (3.15%) and postdischarge cooling time (25 yr) adjusted to produce SNF with a composition consistent with the YMP SNF inventory. The TRU feed, after processing, is shown in Table I under the column "Representative SNF."

TABLE I

Compositions of Transuranic Feed from Spent Nuclear Fuel*

Isotope	YMP Inventory[a] (%)	Representative SNF (%)	ALWR SNF[b] (%)
^{235}U	0.004	0.004	0.002
^{236}U	0.002	0.002	0.002
^{238}U	0.419	0.423	0.325
^{237}Np	5.601	4.313	6.641
^{238}Pu	1.725	1.236	2.749
^{239}Pu	52.172	53.901	48.652
^{240}Pu	21.085	21.231	22.980
^{241}Pu	3.540	3.870	6.926
^{242}Pu	4.623	4.677	5.033
^{241}Am	9.431	9.184	4.654
^{242m}Am	0.019	0.007	0.019
^{243}Am	1.199	1.021	1.472
^{243}Cm	0.003	0.002	0.005
^{244}Cm	0.156	0.116	0.496
^{245}Cm	0.019	0.013	0.038
^{246}Cm	0.002	0.001	0.006

*Note: 0.005% of uranium and 99.9% of transuranics from spent nuclear fuel; YMP Inventory = Yucca Mountain Project average SNF; Representative SNF = SNF used in this analysis; ALWR SNF = advanced light-water reactor SNF.

[a]Reference 19.

[b]Reference 4.

The representative SNF composition is compared with the YMP composition, shown in Table I under the column YMP SNF. The YMP SNF inventory is a composite of many reactor designs, fuel enrichments, and discharge burnups. The representative SNF used in this analysis represents the major actinides isotopes reasonably well. The representative SNF contains less ^{237}Np and ^{238}Pu and more ^{239}Pu than the YMP SNF, which has an effect on the performance of the reactors. The differences in TRU feed are not sufficient to significantly impact the conclusions of this study.

The initial composition and the reference MOX LWR fuel cycle parameters were taken from the Working Party on Physics of Plutonium Recycle Benchmark study,[21] and the latter are given in Table II. The MOX fuel containing 5.6% Pu is irradiated at a power density of 38.3 MW/MTHM to the discharge burnup of 50 GWd/MTHM. The calculated concentration of the major actinide isotopes present in the spent MOX fuel were within the ranges calculated by the different contributors to the benchmark study, with only a few isotopes differing significantly from the average concentrations. The total uranium content and TRU content differed slightly from the average, which suggests that the SAS2H model used in this analysis will accurately predict the mass flows for the MOX fuel cycle. The required Pu loading in the MOX fuel is sensitive to the discharge burnup and storage time for the SNF and to the discharge burnup of the MOX fuel. In this scenario, we use the parameters specified in Ref. 21, which utilize a Pu composition consistent with the representative SNF.

The production of 1 MTU was estimated to require 5.77 MT of NU. The radioactive component of the uranium ore used in this analysis was calculated from the natural isotopic abundance of the uranium isotopes in equilibrium with all radioactive daughter products. The equilibrium concentration of all radioactive daughters for the decay chains of the two primordial uranium isotopes, ^{238}U and ^{235}U, were calculated. The half-lives and branching ratios for the uranium decay chains were taken from the Table of Nuclides.[22]

The FTWR, ATWR, and IFR fuel cycles will repeatedly recycle the TRU. As a result the fuel composition will evolve over time. The analysis of the FTWR, ATWR, and IFR fuel cycles focused on the equilibrium fuel cycle (EqFC). The initial fuel cycles would be expected to be designed to perform similarly to the EqFC, and any differences would have little impact on the overall performance of the transmutation systems. The FTWR, ATWR, and IFR EqFCs were analyzed using the REBUS-3 fuel cycle analysis code,[23] in which the performance of the external cycle is explicitly modeled.

The FTWR is a subcritical reactor driven by a fusion neutron source (Appendix A). The power level in the reactor was maintained at a constant value of 3000 MW(thermal). The fusion neutron source strength (power level) was adjusted to compensate for fuel depletion effect on reactivity. The end of cycle (EOC) was reached when the fusion neutron source strength reached 150 MW of fusion power. The design limit for the maximum neutron multiplication factor was 0.95, which did not prove to be limiting for the five-batch fuel cycle. A dispersion fueled TRU-10Zr/Zr fuel design was used with a maximum loading of 45 wt% of actinides.

The ATWR is an 840-MW(thermal) subcritical reactor driven by a spallation neutron source (Appendix B). The calculations were performed using critical (eigenvalue) neutronics calculations. Sensitivity studies demonstrated that the transmutation parameters agree well with the more detailed source-driven calculations.[4] The

TABLE II

Fuel Cycle Parameters

	Design				
	OTC	MOX	FTWR	ATWR	IFR
Reactor power level (MW)	3000	3000	3000	840	840
Cycle length (effective full-power days)	276	435	564	140	310
Fuel batches	3	3	5	7/8	7
BOC neutron multiplication factor			0.925	0.970	1.021
EOC neutron multiplication factor	1.000	1.000	0.836	0.927	1.000
BOC heavy metal loading (MT)	75	75	24.32	2.44	13.89
BOC TRU loading (MT)	0.35	3.88	23.27	2.39	3.78
Reactor heavy metal discharge burnup (%)	3.3	5.3	29.0	31.0	12.7
Charge TRU enrichment (%)	0	5.60	96.3	98.4	28.0
Reactor discharge TRU burnup (%)	N/A[a]	25.4	29.0	31.0	18.0

[a]N/A = not applicable.

fuel volume fraction was adjusted using the REBUS-3 enrichment search techniques to yield a target beginning-of-cycle (BOC) eigenvalue of 0.97.

The IFR is an 840-MW(thermal) critical reactor (Appendix C). The TRU-to-DU ratio in the charged fuel was determined using the REBUS-3 enrichment search techniques to yield the target EOC eigenvalue of 1.0.

The same techniques were used to produce the multigroup cross sections and perform the neutronics calculation for the FTWR, ATWR, and IFR. The neutronics calculations were two-dimensional discrete ordinates (S8) transport calculations using the DANT code[24] with material-dependent multigroup cross-section libraries. The transport calculations were source-driven calculations for the FTWR and eigenvalue calculations for the ATWR and the IFR. Material-dependent multigroup libraries based on the ENDF/B-V.2 nuclear data library processed using the MCC-2 (Ref. 25) and SDX (Ref. 26) codes for a 34-group energy structure were created for each of the reactors. The reactors were modeled using *r-z* geometry models.

The tritium production cross sections for the FTWR calculations were taken from the JEF-2.2 cross-section set.[22] The tritium production material-dependent group cross sections were collapsed using the spectrum calculated by MCC-2 for the region in which the lithium was located.

IV. ENERGY PRODUCTION AND MASS FLOW

The energy production and mass flows in the different fuel cycles are quantified in this section. The capacity factor and thermal-to-net electrical conversion efficiency will have a significant effect on the performance and net cost of the transmutation system, if electricity is sold. The FTWR and ATWR will both require additional electricity (relative to LWRs and the IFR) for the systems required to operate the neutron source. The higher operating temperatures in the liquid-metal–cooled FTWR, ATWR, and IFR should improve conversion efficiency relative to water-cooled reactors. The capacity factors initially would be greater for LWRs than for IFRs, which in turn would be greater than for FTWRs or ATWRs. Rather than introduce assumptions about these uncertain parameters, the analysis is based on effective full-power operation and thermal energy generation.

Table III summarizes a number of parameters related to energy production and mass flow. These parameters express the performance in different manners, but are related to a few primary variables. These parameters are the TRU content and discharge burnup of the LWR SNF, the chemical separation efficiency, the fractional discharge burnup of the recycled TRU in a single pass through the reactor, and conversion rate.

The reactor energy production is the energy generated in the specific reactor normalized to the initial fuel loading in the LWR. The system energy production is the integral energy production resulting from 1 MTU initial irradiation in the LWR during all phases of the fuel cycle. This quantity is used to normalize the HLW quantities to determine the rate of discharge. Even though the FTWR and ATWR fission essentially the same mass of TRU, differences in neutron spectra and fuel cycles result in different equilibrium fuel compositions and fission rates for the various isotopes. As a result, the ATWR produces slightly more fission energy per gram than the FTWR, which produces small differences in a number of parameters normalized to system energy production. The repeated recycle of the TRU in the fertile-fuel IFR increases energy production by roughly two and one-half times the increase in the FTWR or ATWR because of the

TABLE III

Energy Production and Mass Flow

	Fuel Cycle				
	OTC	MOX	FTWR	ATWR	IFR
Reactor energy production [GW(thermal)·yr/MTU]	0.0903	0.0250	0.0298	0.0315	0.0749
System energy production [GW(thermal)·yr/MTU]	0.0903	0.1153	0.1202	0.1219	0.1653
System TRU concentration (g/MTU)	11 005	8 264	38	37	62
System TRU discharge rate [g/GW(thermal)·yr]	121 805	71 666	316	303	374
System TRU burnup (%)	N/A[a]	24.9	99.65	99.67	99.44
SNF feed rate [MTU/GW(thermal)·yr]	11.1	40.1	33.5	31.7	13.3
TRU energy generation (MWd/g)	N/A	3.33	0.96	0.99	2.46
Support ratio [GW(thermal) LWR/GW(thermal)]	N/A	3.64	3.05	2.88	1.21
First core SNF requirements [MTU/GW(thermal)]	N/A	118	705	259	409

[a]N/A = not applicable.

fast fission of ^{238}U and the subsequent fission of transuranics produced by neutron capture in ^{238}U. The energy produced by the repeated recycle of the TRU in the IFR will be nearly as large as the energy produced in the LWR when the SNF was created.

Table III also includes the system TRU concentration and the system TRU discharge rate. The system TRU concentration is the mass of TRU in the HLW resulting from 1 MTU initial irradiation in the LWR during all phases of the fuel cycle. The system TRU discharge rate is the system TRU concentration normalized by the system energy production. The TRU concentration in the waste from a transmutation fuel cycle is a function of the separation efficiency and the discharge TRU burnup in each pass through the transmutation reactor. The single recycle of Pu in MOX fuel would reduce the TRU concentration by nearly 3 kg/MTU, but 8 kg/MTU would remain. At the separation efficiencies assumed in this analysis, only a small fraction of the TRU would ultimately end up in an HLW repository. The IFR EqFC HLW has a higher concentration of TRU than the ATWR or FTWR EqFCs because the IFR operates at about half the discharge TRU burnup of the FTWR and ATWR. This results in a higher fraction of the TRU being recycled after each pass through the IFR, resulting in more TRU leaking into the waste stream. When the increased energy production of the IFR EqFC is taken into account, the system TRU discharge rates from the IFR, FTWR, and ATWR EqFC are very similar.

Table III also includes the system TRU burnup, which is defined as the reduction in TRU concentration relative to the TRU concentration in the SNF feed. The single recycle of Pu in MOX fuel results in a system TRU burnup of 25%. If separations were perfect, repeated recycle of the TRU would result in a 100% system TRU burnup, regardless of reactor design. This parameter is sensitive to the separations efficiencies for the EqFCs, but the system TRU burnup of the fertile fuel will always lag because of a lower TRU discharge burnup after each pass through the reactor.

Table III also includes the TRU energy generation, which is the energy generated per unit reduction in TRU mass. The average fission energy for actinides is ~1 MWd/g. This limits the maximum rate of TRU reduction, which is the inverse of the energy generated, to ~1 g for each MWd of thermal energy, regardless of reactor design. Utilization of a fertile fuel will increase the energy generated and reduce the rate of TRU reduction. The nonfertile ATWR and FTWR both operate at the maximum rate, with a small difference resulting primarily from differences in the fission rates of the various isotopes and a few other reactions that contribute to the thermal energy balance. Transuranic reduction in the IFR or in an LWR using MOX fuel will produce 2.46 MWd/g or 3.33 MWd/g, respectively, which reduces the TRU inventory at significantly below the maximum rate.

The IFR and the MOX LWR are operating at conversion rates of 0.6 and 0.7, respectively. For this discussion, the conversion rate is defined as the change in mass of uranium from fission and capture to the total change in the mass of the actinides. This gives a small difference relative to including only capture and conversion to TRU and is more appropriate for energy and mass balance for transmutation systems. Reduction of the conversion rate to 0.5 is probably achievable. At a conversion rate of 0.5, the energy generated would be 2 MWd/g for transuranic reduction. This corresponds to 0.5 g of TRU reduction per MWd of thermal energy produced or half the rate of the nonfertile FTWR and ATWR.

The inventory of discharged SNF is estimated to be over 47 000 MTU by the end of 2002. Over 14 000 GW(thermal)·yr of operation will be required to destroy the TRU inventory in the accumulated inventory of discharged SNF. The current discharge rate for SNF is over 2000 MTU/yr. This would require over 60 GW(thermal) of transmutation reactor capacity to stop the growth in the discharge SNF inventory. Operations using fertile fuel with a conversion rate of 0.5 would double these requirements.

A common way to express the efficiency of a transmutation system is the ratio of the LWR thermal power capacity to the required transmutation reactor thermal power capacity to transmute all TRU from LWR SNF. This "support ratio," included in Table III, would need to be adjusted for the relative capacity factors. The ATWR and FTWR will support LWRs producing three times the FTWR or ATWR thermal power. The IFR would support LWRs producing 1.2 times its thermal power. Reducing the conversion rate to 0.5 would increase the IFR support ratio to 1.5, which is still half that of the nonfertile ATWR and FTWR. The transmutation reactors would be a very large fraction of the nuclear generating capacity, ~25% for FTWR and ATWR systems and ~40 to 45% for the IFR system.

Table III also includes the feed rate, which is the rate SNF from the LWR is processed by the transmutation systems and is primarily a function of the TRU concentration in the SNF, the conversion rate, and system TRU burnup. There is only a small difference between the nonfertile FTWR and ATWR systems. The FTWR and ATWR EqFCs would process the LWR SNF at a rate of 34 MTU/GW(thermal)·yr and 32 MTU/GW(thermal)·yr, respectively. The fertile IFR would process SNF at the much lower rate of 13.3 MTU/GW(thermal)·yr. Utilizing existing technology to recycle Pu once in MOX fuel would result in the highest processing rate, 40 MTU/GW(thermal)·yr, allowing the SNF that has been accumulating to move into the transmutation system most rapidly, but a significant quantity of MA and spent MOX fuel would accumulate and would need to be sent to another transmutation system. For essentially total actinide destruction, the nonfertile FTWR and ATWR would process the SNF at the highest possible rates, but their

operation would probably require the longest lead times for deployment and achievement of high-capacity operation.

Table III also includes an estimate of the SNF that would need to be processed to produce the fuel for the first core. The first core loading represents a significant logistics problem because the initial startup of the transmutation systems would require a large quantity of SNF to be processed to produce fuel for at least the first full-core loading and first reload. After recycled material from the previously irradiated fuel is available, a much smaller quantity of SNF will be required for makeup of the fissioned TRU. The equilibrium TRU loading can be used to estimate the quantity of SNF that will need to be processed for the first core loading. The actual value would be somewhat less than the equilibrium loading because the SNF TRU has a higher concentration of fissile isotopes than the recycled transmutation reactor fuel, and there will not be any FP present from the fuel that was in the reactor during previous cycles. The FTWR has an equilibrium BOC TRU loading of 7.8 MT/GW(thermal), requiring 705 MTU/GW(thermal) of SNF for the initial core loading. The equilibrium ATWR operates with a much lower TRU loading of 2.9 MT/GW(thermal), requiring 259 MTU/GW(thermal) of SNF for the initial core loading. The equilibrium IFR operates at an intermediate TRU loading of 4.5 MT/GW(thermal), requiring 409 MTU/GW(thermal) for the initial core loading. The MOX fuel can be interspersed with EU fuel, and the entire first core of MOX fuel is not required if existing reactors are utilized.

If the MOX fuel is assumed to be loaded into existing reactors that would have otherwise produced SNF, the use of MOX fuel offsets the production of new sources of Pu. One MTU of SNF produces enough Pu for ~0.2 MT of MOX fuel. In other words, reprocessing 1 MTU of SNF offsets the production of 0.2 MTU of SNF. If the MOX fuel reduces the TRU inventory by 25% and the EU loading is reduced by 17%, the effective TRU reduction from a single MOX recycle is ~38%. Under these conditions, MOX recycle would reduce TRU inventories without increasing nuclear energy production.

The use of fertile fuel increases the required capacity of the transmutation system and the quantity of TRU in the waste streams. The use of MOX fuels makes modest reductions in the TRU inventory and could offset new production of TRU. Nonfertile fuels transmute TRU at nearly the same rate regardless of the system. Differences in discharge burnup, electrical efficiencies, capacity factor, and ultimately cost will be distinguishing factors between these systems.

V. TOXICITY

The toxicity is defined as the quantity of water required to dilute the waste to the maximum permissible concentration. The toxicity was calculated using the values from the SCALE 4.4 code package,[18] which considers only the radiotoxicity without considering any chemical toxicity effects. The toxicity is one of the simplest measures of the radiological benefit of transmutation and is used frequently to measure the effectiveness of transmutation systems. Toxicity is a measure based solely on the isotopic composition of a material at any given time. It is primarily a measure of the radioactivity of a material and is not a good measure of the risk posed by a material placed in a geologic repository. In general, the short-lived isotopes are the most toxic but easiest to contain for sufficient time, while the long-lived isotopes are the least toxic but most difficult to contain for sufficient time.

The toxicity of the various waste streams will be very time dependent, with changes in toxicity that will vary over many orders of magnitude. Isotopes with short half-lives that decay to stable isotopes rapidly decline in toxicity. Isotopes with very long-half lives have relatively low toxicities in the pure form. If the very long-lived isotopes have long decay chains, the toxicity will slowly increase and eventually be dominated by the radioactive daughter. For example, a sample of ^{238}U has a toxicity two orders of magnitude less than it does at equilibrium with its radioactive daughters.

Figure 2 shows the toxicity of the representative SNF from the LWRs used in this evaluation. The radioactive decay will result in a continual evolution of the composition of the SNF. In order to determine the materials that need to be transmuted today to affect the toxicity at some point in the future, the time-dependent toxicity attributed to each isotope or group is the toxicity of all isotopes present as a result of decay from the original isotope or group.

For example, the individual isotope ^{241}Pu is shown separately in Fig. 2. The ^{241}Pu has a half-life of 14 yr, and there is only a very small concentration of parent isotopes for ^{241}Pu in the SNF. Therefore, the actual mass of ^{241}Pu will decay away nearly completely in 200 yr. The ^{241}Pu will decay to ^{241}Am, 430-yr half-life, and then to ^{237}Np, which has a two million year half-life. The ^{241}Pu toxicity curve shows that initially it constitutes a very small fraction of the toxicity, but at a few hundred years, its daughter products will constitute a very large fraction of the toxicity. After the ^{241}Am decays away in a few thousand years, the ^{241}Pu contribution to toxicity is again small, but eventually the radioactive daughters of ^{237}Np controlled by the 150 thousand-year half-life of ^{233}U will build to secular equilibrium with ^{237}Np, and the 14-yr half-life isotope ^{241}Pu will contribute a small but significant fraction of the toxicity of the SNF at one million years as shown in Fig. 2. Therefore, the original ^{241}Pu present in the SNF must be fissioned in order to destroy the toxicity of the ^{237}Np and its daughters present at one million years that were initially ^{241}Pu.

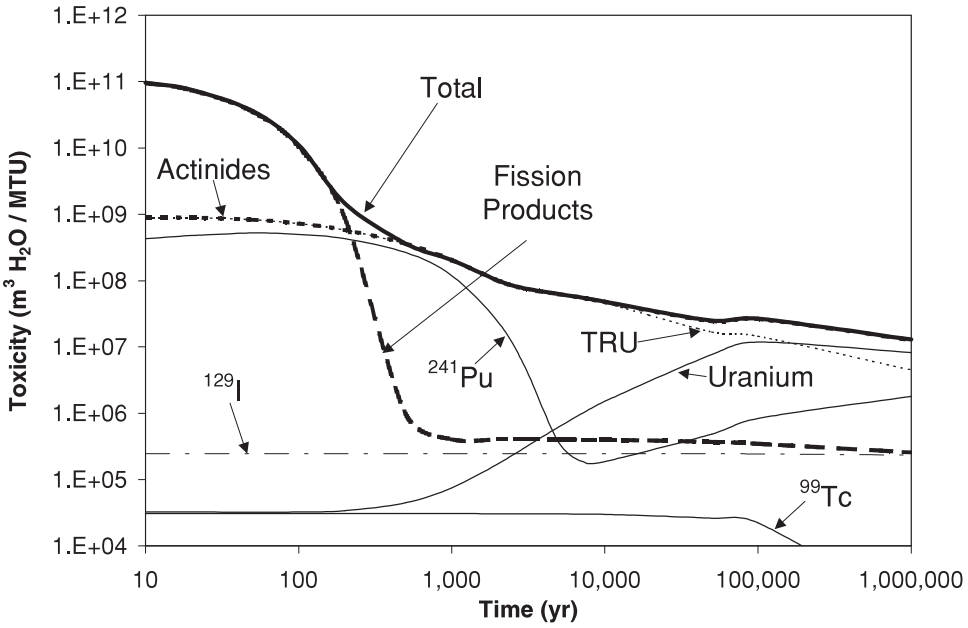

Fig. 2. Toxicity of representative SNF. (Note: All groups are for parent isotopes at discharge and include all daughter products that accumulate over time.)

Figure 2 shows that the short-term toxicity is dominated by FP that decay away in a few hundred years. After the FP have decayed away, the toxicity is dominated by the TRU for ~100 000 yr. Beyond 100 000 yr, the residual IEU will be the largest contributor to toxicity. By one million years, only a few TRU isotopes have half-lives sufficiently long to remain in any significant quantities, and all other TRU isotopes will have decayed away, many of them back to ^{238}U and ^{235}U. The long-lived FP represent an extremely small fraction of the toxicity.

The time-dependent toxicity of the unirradiated EU from which the LWR fuel is fabricated will be used as a benchmark of the HLW toxicity. The uranium ore mined for fuel fabrication will be used as a benchmark for the toxicity of all waste streams. The uranium ore is at equilibrium and would not vary over the one million year time frame if left in the ground. However, the uranium ore is split into the mill tails and NU. The NU is further divided into the EU and DU streams. Initially, nearly all of the toxicity is in the mill tails, which remains true for nearly 100 000 yr. Slowly the daughter products will build to equilibrium levels, and the toxicity will be determined by the ^{238}U concentration, the bulk of which is in the DU stream.

Figure 3 shows the toxicity concentration in the HLW from the OTC SNF, from the single recycle of the Pu from the SNF in MOX fuel, and from repeatedly recycling TRU from the SNF in an FTWR, ATWR, or IFR EqFCs. The unirradiated EU is shown to illustrate the effect of irradiation and the separation and transmutation. The unirradiated EU will increase in toxicity by more than two orders of magnitude as its radioactive daughters build up, reaching a maximum at ~100 000 yr, and then decline from the peak toxicity as the ^{234}U concentration falls from its enriched levels. The toxicity of the HLW from the OTC and from the single recycle of Pu in MOX fuel is reduced to the level of the unirradiated EU toxicity after ~100 000 yr. The single recycle of Pu in MOX fuel has no significant effect on toxicity up to ~1000 yr and causes only a small reduction at later times. Recycling the TRU repeatedly in the FTWR, ATWR, and IFR EqFCs reduces the toxicity of the HLW below that of the unirradiated EU in ~6000 to 8000 yr. Beyond 100 000 yr, the separation of the IEU from the SNF has a significant effect on the HLW toxicity because the recovered uranium is sent to an LLW facility.

In the short-term, the FTWR, ATWR, and IFR transmutation systems increase the FP concentrations in the HLW, which increases the toxicity for a few hundred years. The toxicity in the several hundred to 100 000-yr timeframe is dominated by the medium-lived TRU isotopes and their daughters. By reducing the TRU concentration dramatically, the transmutation systems reduce the toxicity by approximately two orders of magnitude relative to the untreated OTC SNF at 1000 yr. The MOX fuel cycle destroys a large fraction of the long-lived Pu isotopes and increases many of the medium-lived MA isotopes, which tend to cancel each other in this time frame.

The HLW from the IFR EqFC has a long-term toxicity that is nearly double that of the HLW from the

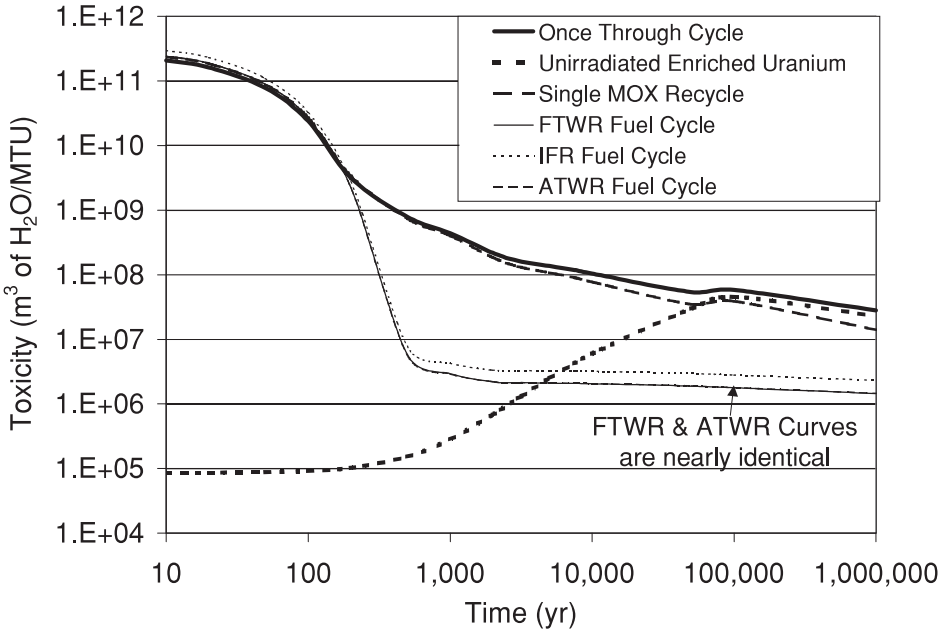

Fig. 3. Toxicity of waste sent to repository per unit mass of LWR SNF.

FTWR and ATWR EqFCs because the lower system TRU burnup of the IFR EqFC results in a larger number of imperfect separations and hence a larger concentration of TRU in the HLW. The increased fissions in the IFR EqFC increase the short-term toxicity far more than in the FTWR and ATWR EqFCs.

The previous discussion was about the toxicity sent to a repository as the result of 1 MTU irradiated in an LWR and then processed and irradiated according to the given fuel cycle. However, in the process, additional energy will be produced. A more relevant comparison might be the rate toxicity would be sent to the HLW repository as a function of system energy production for each fuel cycle. Figure 4 shows the HLW toxicity normalized to the total thermal energy generation. The differences in toxicity between the HLW from the IFR and the ATWR and FTWR EqFCs are reduced because of the much larger energy production of the IFR. The rate of production of the short-term toxicity in the HLW is nearly the same for the FTWR, ATWR, and IFR EqFCs. Yet, the rate of production of the long-term toxicity in the HLW from the IFR EqFC is still significantly higher than that from the ATWR and FTWR EqFCs. The IFR HLW will have significantly higher levels of ^{238}U because of the fertile fuel and a greater concentration of TRU because of the lower system TRU burnup. The single MOX recycle shows a significant improvement over the OTC in terms of toxicity per unit thermal energy production.

Figure 5 shows the toxicity of all waste streams, including both the HLW and LLW streams. For comparison, the toxicity of the uranium ore is also shown. The toxicities of all waste streams from the FTWR, ATWR, or IFR EqFCs approach the toxicity of uranium ore after ~500 yr. For the FTWR, ATWR, and IFR EqFCs, once the FP have decayed, nearly all the toxicity will be in LLW facilities. The medium-lived TRU from the OTC and MOX fuel cycle produce a longer tail that increases the toxicity for tens of thousands of years. The toxicity of all waste streams from the OTC and MOX fuel cycle differ only slightly and approach the toxicity of uranium ore after ~100 000 yr.

Complete transmutation of the TRU reduces the toxicity beyond a few hundred years in the HLW that will be sent to the repository. All fuel cycles analyzed only fission, a very small fraction of the original NU; the maximum is just over 1% in the IFR EqFC. Therefore, the long-term toxicity beyond 100 000 yr is essentially unchanged. The FTWR, ATWR, or IFR transmutation systems shift a large fraction of the long-term toxicity from the HLW repository to the LLW facility by separating the residual uranium in the LWR SNF.

VI. REPOSITORY IMPACTS

The repeated recycling of TRU in the FTWR, ATWR, or IFR EqFC would have a dramatic impact on the design requirements for an HLW repository, relative to the requirements for a repository designed for intact LWR SNF. Transmutation would change the isotopic composition of the waste by converting actinides to FP, most of which are relatively short lived. The only waste sent to the HLW repository as a result of the repeated recycle of

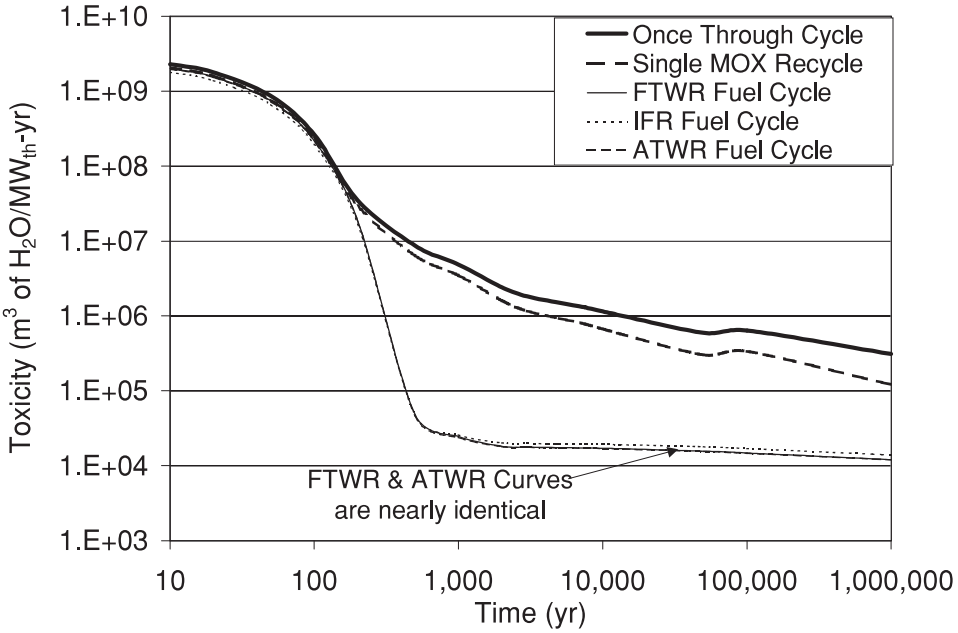

Fig. 4. Toxicity of waste sent to repository per unit thermal energy.

all transuranics in a transmutation system would be the FP and the small fraction of TRU that would inherently leak into the HLW stream. This would change the radioactive source term, the heat source, heat profile, and volume of the waste sent to the repository. We note that with the availability of separation and processing capability for repeated recycling in a transmutation reactor, it would be logical to prepare the waste in a vitrified or other form for improved performance relative to intact storage of SNF; however, we do not consider this in our analysis. In addition, the volume of waste would be a function of the final waste form and would be comprised mostly

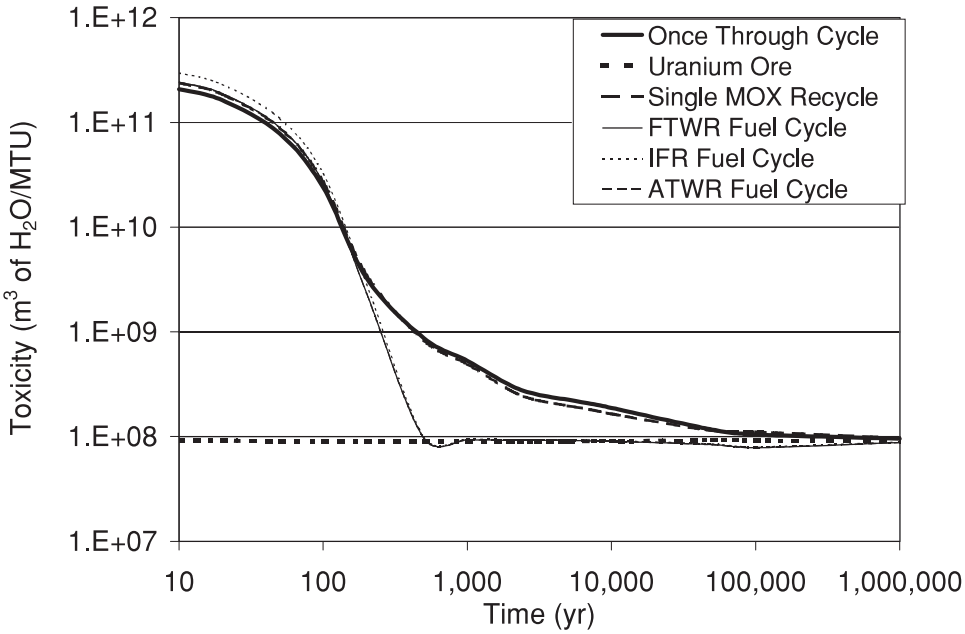

Fig. 5. Toxicity of all waste streams from 1 ton of enriched uranium.

of the benign waste matrix, which is unknown. Therefore, volume was not considered, but it is anticipated to be a secondary effect.

The quantity of the six "repository" isotopes in the HLW of the different fuel cycles was evaluated. These repository isotopes were identified in the Yucca Mountain Viability Assessment[27] as contributing the largest fraction to predicted dose rates at 10 000, 100 000, and 1 000 000 yr for scenarios involving waste dissolution and transportation via groundwater. Other analyses have identified other isotopes including activation products that may contribute significantly. The six repository isotopes include two long-lived FP (LLFPs), ^{99}Tc and ^{129}I, and four actinides ^{237}Np, ^{234}U, ^{239}Pu, and ^{242}Pu. The individual repository isotopes include all parent isotopes that would decay to these isotopes on the timescale of the repository. For example, the repository isotope ^{234}U includes ^{238}Pu, but not ^{238}U. The relative importance of these isotopes, and other isotopes not included, depends on many factors, including the waste form and assumptions about conditions in the repository and future climatic conditions. Therefore, large uncertainties exist about the actual dose rates that would occur, but significant reductions in these isotopes would be expected to translate into significant reductions in the predicted dose rates.

In Sec. IV, the fuel cycle performance was assessed in terms of TRU mass, and all TRU isotopes were treated equally. In this section, only the actinide isotopes that are one of the four repository isotopes or included parents are considered. For example, neutron capture in ^{237}Np produces ^{238}Pu, which is a parent to and is included in the ^{234}U total, thereby reducing the ^{237}Np mass and increasing the ^{234}U mass. On the other hand, neutron capture in ^{239}Pu produces ^{240}Pu, which is not included as a repository isotope, and thus results in a net reduction in the repository isotopes without fission occurring. The different neutron spectra and fuel cycles result in different total concentrations of TRU as well as of the individual isotopes. Differences in the mass of repository isotopes are a combination of the system TRU burnup and variations in the concentrations of individual isotopes.

Figure 6 shows the concentration of the repository isotopes in the HLW from the reference OTC SNF, from the single recycle of the Pu from the SNF in MOX fuel, and from repeatedly recycling TRU from the SNF in an FTWR, ATWR, or IFR EqFC. As a benchmark, the ^{234}U concentration in the unirradiated EU is also included.

The effect of the FTWR, ATWR, and IFR EqFCs is to convert actinides into FP, thus reducing the concentration of the TRU isotopes and increasing the concentration of the FP isotopes. The increase in LLFPs is roughly proportional to the fission rate with small differences resulting from neutron spectra and fuel cycle.

The MOX fuel cycle has minimal impact on the ^{237}Np, while the FTWR, ATWR, and IFR EqFCs transmute nearly all the original ^{237}Np. The ^{237}Np concentration in the HLW is reduced by $>$99.6% in the FTWR, ATWR, and IFR EqFCs, relative to the OTC SNF.

The original unirradiated EU has a significant concentration of ^{234}U, which is increased by 14% in the OTC. In the MOX, FTWR, ATWR, and IFR EqFCs, 46% of the repository isotope ^{234}U in the SNF is separated and sent to LLW facilities. The fraction of the ^{234}U that is not sent to an LLW facility is the parent TRU isotopes

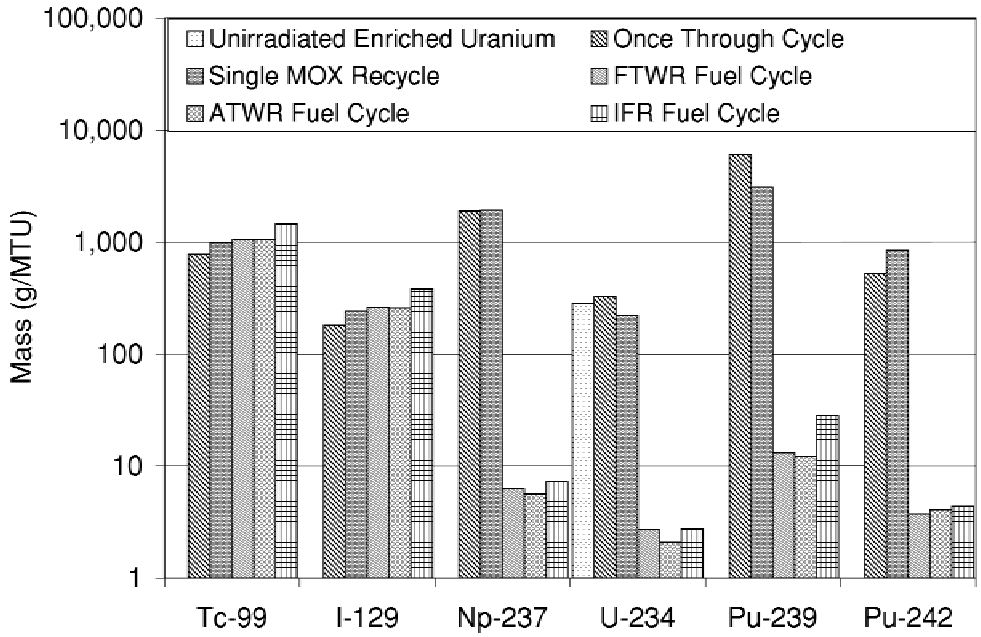

Fig. 6. Concentration of repository isotopes in HLW.

(e.g., ^{238}Pu) that have not yet decayed. The ^{234}U concentration is increased by 37% in the spent MOX fuel relative to the fresh MOX fuel. Excluding the ^{234}U, which is separated and sent to the LLW facility, the ATWR EqFC reduces the ^{234}U concentration in the TRU feed by 98.5%, which is a slightly larger than the FTWR and IFR EqFCs. The larger reduction in the ^{234}U concentration in the ATWR EqFC relative to the FTWR EqFC exists despite the nearly identical total quantities of actinides in the HLW streams. The combination of neutron spectra and fuel cycle differences results in a lower fraction of ^{234}U (^{234}U, ^{238}Pu, ^{242}Cm, and ^{242m}Am) in the HLW from the ATWR EqFC relative to the FTWR EqFC. Nearly the same concentration of ^{234}U is in the HLW of the IFR as in the HLW of the FTWR, despite the much larger concentration of actinides in the HLW from the IFR EqFC.

The single recycle of Pu in MOX fuel achieves a modest 48% reduction in the ^{239}Pu, but increases the ^{242}Pu concentration by 62%. Because of the large ^{239}Pu source term resulting from the ^{238}U neutron capture in the fertile IFR fuel matrix, nearly three times as much ^{239}Pu will leak into the HLW streams from the IFR EqFC than the technologically equivalent FTWR or ATWR EqFCs, which still represents a 99.5% reduction in ^{239}Pu.

Figure 7 compares the rate that the repository isotopes will be sent to the HLW repository per unit thermal energy production. The LLFPs ^{99}Tc and ^{129}I would be sent to the repository at rates of 9 g/MW(thermal)·yr and 2 g/MW(thermal)·yr, respectively. The small difference in rates for the LLFPs is a result of differences in fission yields and in situ transmutation. The IFR, in general, appears to be the most effective at reducing the rates for the actinide repository isotopes, with the exception of a significantly higher rate for ^{239}Pu. Taken in aggregate, the OTC SNF would send the four actinide repository isotopes to the HLW repository at a rate of 97 g/MW(thermal)·yr, the MOX fuel cycle at 53 g/MW(thermal)·yr, the IFR EqFC at 0.26 g/MW(thermal)·yr, the FTWR EqFC at 0.22 g/MW(thermal)·yr, and the ATWR at 0.20 g/MW(thermal)·yr. The significance of the differences between the FTWR, ATWR, and IFR EqFCs can only be determined by detailed analysis of the repository, including the final waste forms.

The heat source from the HLW is a major factor in the design of the repository. The decay heat will determine how the HLW is managed and the design of the containers used for shipping, storage, and disposal. The waste will be stored above ground for some period of time, and after emplacement the repository will remain open for an additional period of time. This time period will allow a large fraction of the FP to decay with the heat vented to the atmosphere. After the repository has been sealed, the waste will be well insulated, and the temperature of the waste and surrounding repository will increase as a result of the heat load. This affects the behavior of the groundwater as it moves through the repository, the dissolution rates of materials in contact with the groundwater, and the quantity of material that can be placed in the repository. In order to evaluate the impact of the heat loading, two parameters were calculated. The first parameter was the instantaneous power from the decay heat of the HLW. The second parameter was the integral decay energy of the HLW after closure

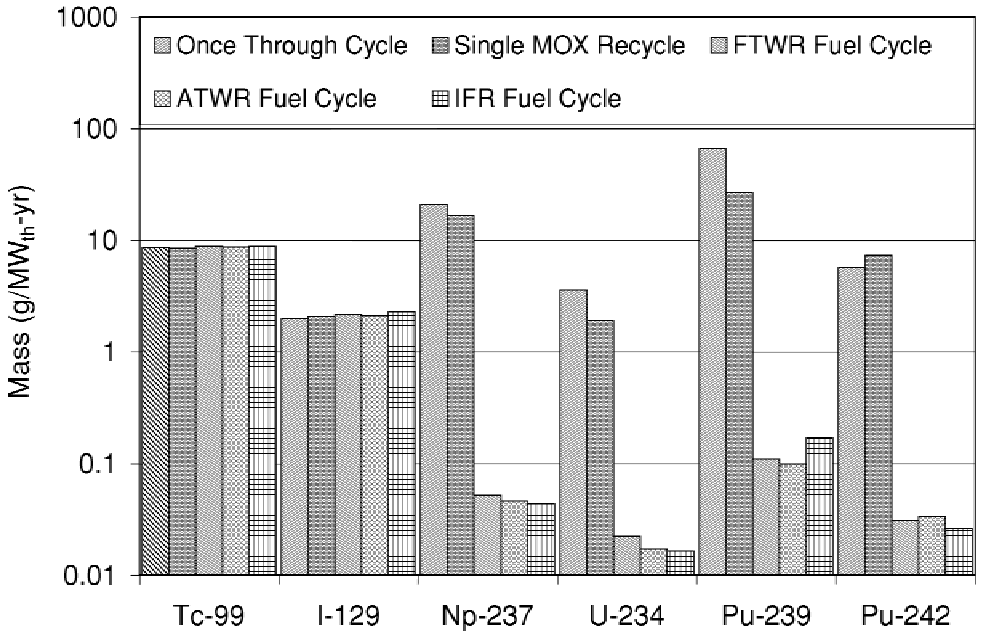

Fig. 7. Rate of production of repository isotopes in HLW.

of the repository. At the time of repository closure, the HLW was assumed to have decayed for 100 yr.

Figure 8 compares the decay heat production rates for the different fuel cycles. Over the first 100 yr, there is very little difference because all systems are dominated by the FP, and the production of FP is roughly proportional to energy production. Beyond 100 yr, the medium-lived TRU isotopes will dominate the heat source, and the HLW from both the OTC and MOX fuel cycle will still contain very large concentrations of these isotopes. Therefore, the heat sources from the HLW from the OTC and MOX fuel cycles will drop at a much slower rate than the heat sources from the HLW from the FTWR, ATWR, and IFR EqFCs, which have similar heat source time profiles. Thus, transmutation significantly reduces the repository heat removal requirement.

Figure 9 shows the rate of production of the integral decay energy beyond 100 yr, the assumed time to closure, for the different fuel cycles. The medium-lived actinides present the most significant heat load beyond 100 yr. The HLW from the single recycle of Pu in MOX fuel reduces the energy deposited in the repository from 100 to 1000 yr by 23% relative to the OTC SNF. The energy deposited in the repository from 100 to 1000 yr after repeated recycle of the TRU in the FTWR, ATWR, or IFR EqFCs is reduced by 96% relative to the OTC SNF due to the highly reduced level of TRU in the HLW. Transmutation will allow for a much lower heat load design or a more tightly packed repository than with the OTC SNF.

The repository for the HLW from the FTWR, ATWR, or IFR EqFCs will be loaded with significantly different waste than the OTC SNF. The waste will contain far lower concentrations of uranium and TRU and higher concentrations of FP for a given quantity of SNF. The initial heat load will be increased by the higher concentration of FP. Elimination of the TRU causes the heat load to fall rapidly beyond 100 yr. Therefore, the heat deposited in the repository after closure will be dramatically reduced. The transmuted waste will be in a tailored waste form. All of these changes will almost certainly allow for a significant increase in the capacity of the repository without exceeding dose and heating limits.

VII. PROLIFERATION RESISTANCE OF HLW REPOSITORY WASTE

One concern that has been raised about the disposal of the intact SNF is that the large quantity of TRU will present a proliferation risk after the radiation barrier has fallen to levels where the SNF is no longer self-protecting. Repeatedly recycling the TRU in the FTWR, ATWR, or IFR EqFC would result in a far more dilute TRU waste with a much higher concentration of FP, although the very existence of separation systems will raise a different type of proliferation concern. Several parameters were calculated to evaluate the relative proliferation risk of the SNF from the OTC, spent MOX fuel from the single recycle of Pu in MOX fuel, and the HLW from the repeated recycle of TRU in an FTWR, ATWR, or IFR EqFC. These parameters are related to technical and physical

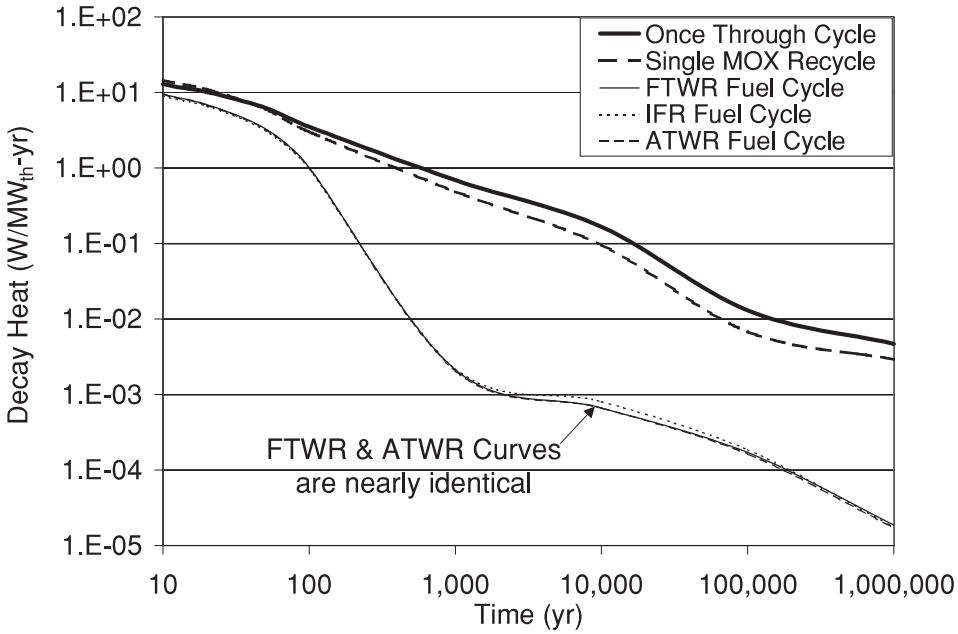

Fig. 8. Rate of production of decay heat.

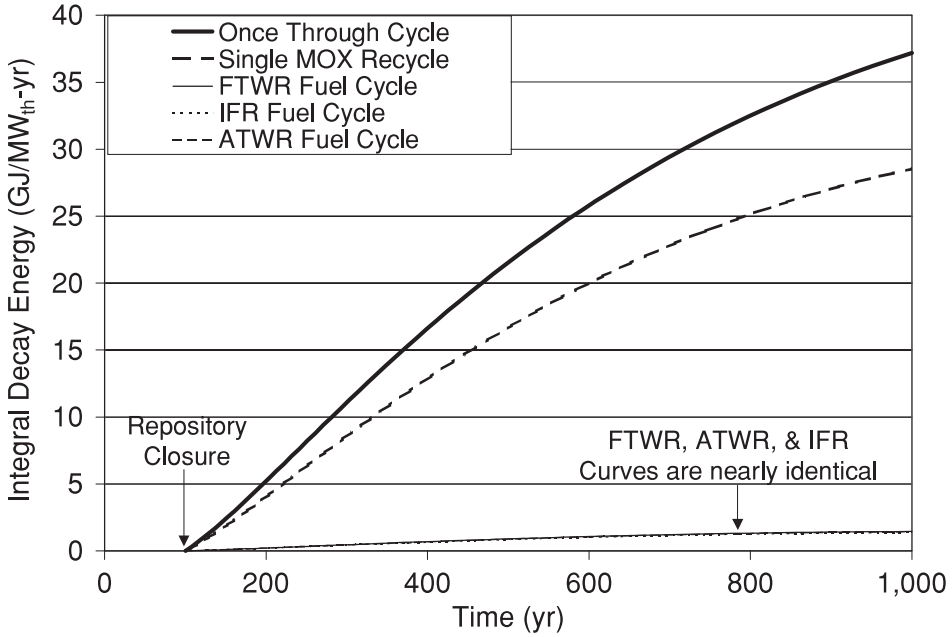

Fig. 9. Rate of accumulation of integral decay energy (after repository closure at 100 yr).

barriers to proliferation and are based on the analysis of Ref. 28.

The technical barrier parameters provide a measure of the relative difficulty of achieving a significant nuclear yield from an explosive device. The first parameter is the bare critical mass (BCM). The BCM is the minimum unreflected quantity required to produce a critical nuclear device. A critical configuration can be created from nearly all TRU isotopes. The BCM was calculated in two ways: (a) assuming separation of pure Pu and (b) assuming separation of all TRU en masse. The BCM is time dependent because short-lived isotopes decay, changing the isotopic composition of the remaining Pu and TRU. Other technical parameters are the decay heat (DH) and spontaneous fission neutron source (SNS) of one BCM. The higher the levels of DH and SNS of the explosive device, the more technically challenging it is to produce a significant nuclear yield. These quantities are a function of the BCM and isotopic composition of the material.

The physical barrier parameters provide a measure of the relative difficulty of acquiring sufficient material in a form pure enough to produce a nuclear explosive. The two physical barrier parameters analyzed are related to the quantity of waste containing one BCM and the radiological hazard associated with that quantity of waste. The mass of radioactive waste (MRW) containing one BCM provides a measure of the relative amounts of radioactive waste, actinides, and FP that would need to be handled and processed to recover sufficient material to produce an explosive device. The MRW does not account for the additional dilution and tailoring of the waste from the transmutation systems that would increase the barrier to proliferation. The unshielded dose rate (UDR) at 1 m provides a measure of the relative radiological hazard of separating sufficient material to produce an explosive device.

Figure 10 shows the BCM of the Pu and TRU in the HLW from the OTC SNF, from the single recycle of the Pu, from the SNF in MOX fuel, and from repeatedly recycling TRU from the SNF in an FTWR, ATWR, or IFR EqFC. For reference, the BCM (10.7 kg) for WGPu is also shown. Transmutation increases the BCM by increasing the concentrations of isotopes with large BCM such as ^{242}Pu, which in pure form has a BCM of 92 kg. The BCMs remain fairly constant for $>$10 000 yr, and then slowly increase as the ^{239}Pu decays with a 24 110-yr half-life. At one million years, ^{242}Pu and ^{237}Np are the only TRU isotopes present in significant quantities.

Figure 11 shows the DH of one BCM of the Pu and one BCM of the TRU in the HLW from the OTC SNF, from the single recycle of the Pu from the SNF in MOX fuel, and from repeatedly recycling TRU from the SNF in an FTWR, ATWR, or IFR EqFC. For reference, the DH of one BCM of WGPu (24.6 W) is also shown. Transmutation increases the DH by increasing the concentrations of the relatively short-lived alpha-emitting isotopes ^{238}Pu and ^{240}Pu. Initially, one BCM of Pu from the OTC SNF generates eight times more heat than one BCM of ^{239}Pu. The short-lived Pu isotopes that are producing the heat decay away and reduce the DH. Even so, the DH of the Pu remains significantly higher than that of WGPu for tens of thousands of years. Even though the MA have a small effect on the BCM, if they are not separated, the

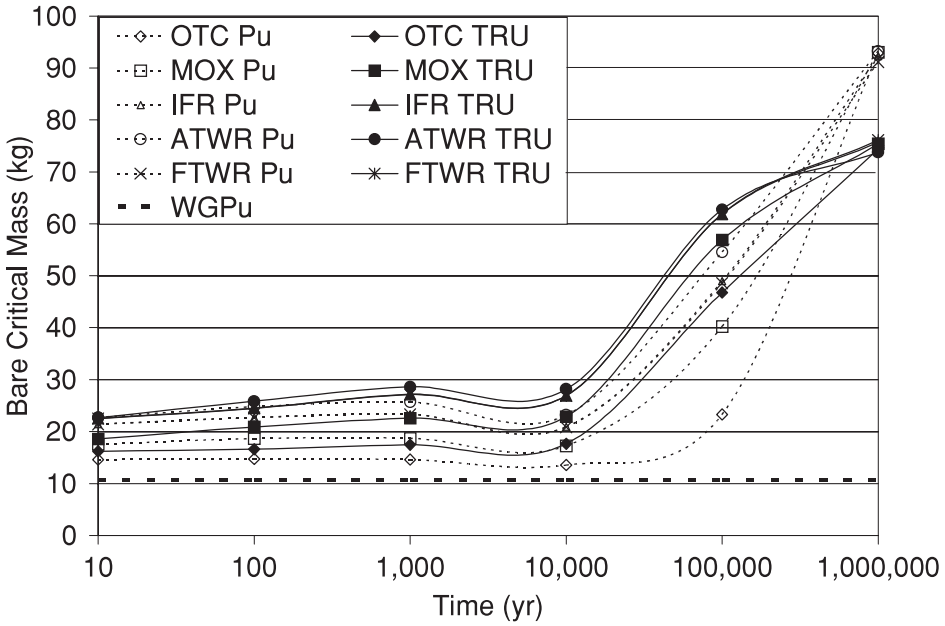

Fig. 10. BCM (dashed lines = plutonium only; solid lines = all transuranics).

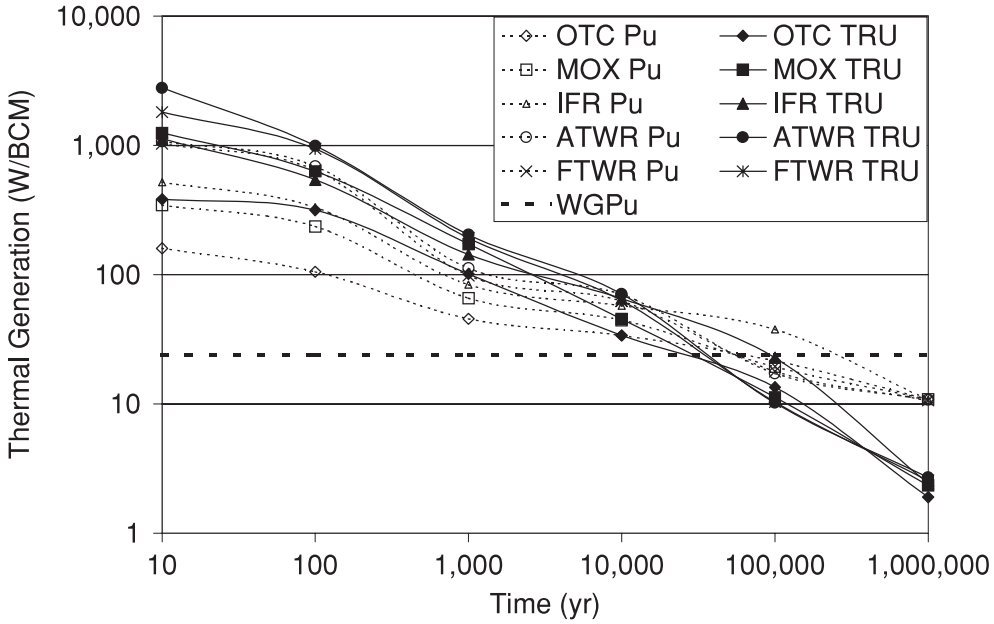

Fig. 11. Decay heat from one BCM (dashed lines = plutonium only; solid lines = all transuranics).

DH of one BCM of TRU is roughly double the DH of one BCM of Pu from the same fuel cycle. The increased DH of the TRU relative to Pu falls significantly with time.

Figure 12 shows the SNS of one BCM of the Pu and TRU in the HLW from the OTC SNF, from the single recycle of the Pu from the SNF in MOX fuel, and from repeatedly recycling TRU from the SNF in an FTWR, ATWR, or IFR EqFC. For reference, the SNS of one BCM of WGPu (6.0×10^5 n/s) is also shown. The OTC Pu has an SNS an order of magnitude higher than WGPu. Transmutation increases the SNS by increasing the even mass Pu isotopes, which have SNSs that are orders of magnitude greater than ^{239}Pu and ^{241}Pu. The SNS of the TRU remains significantly above WGPu for all fuel cycles. The very small concentrations of a few isotopes

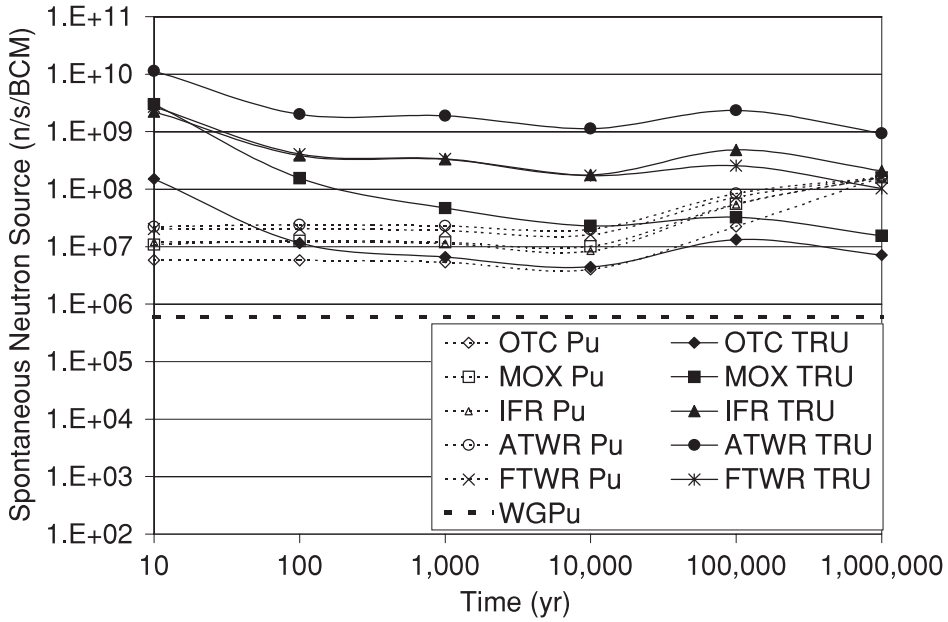

Fig. 12. Spontaneous neutron source from one BCM (dashed lines = plutonium only; solid lines = all transuranics).

with very high spontaneous fission rates produce the variation in SNS from the TRU in the different fuel cycles. The concentration of these isotopes is relatively uncertain and differences in SNS for the TRU may not be real.

Figure 13 shows the MRW containing one BCM of the Pu and one BCM of the TRU from the OTC SNF, from the single recycle of the Pu from the SNF in MOX fuel, and from repeatedly recycling TRU from the SNF in an FTWR, ATWR, or IFR EqFC. A relatively small MRW, <2 MT, of OTC SNF would have to be processed to recover one BCM of Pu. By concentrating the Pu in the MOX fuel, the MRW of spent MOX fuel is reduced to less than one-third that of the OTC SNF. The IFR, ATWR, and FTWR EqFCs increase the MRW

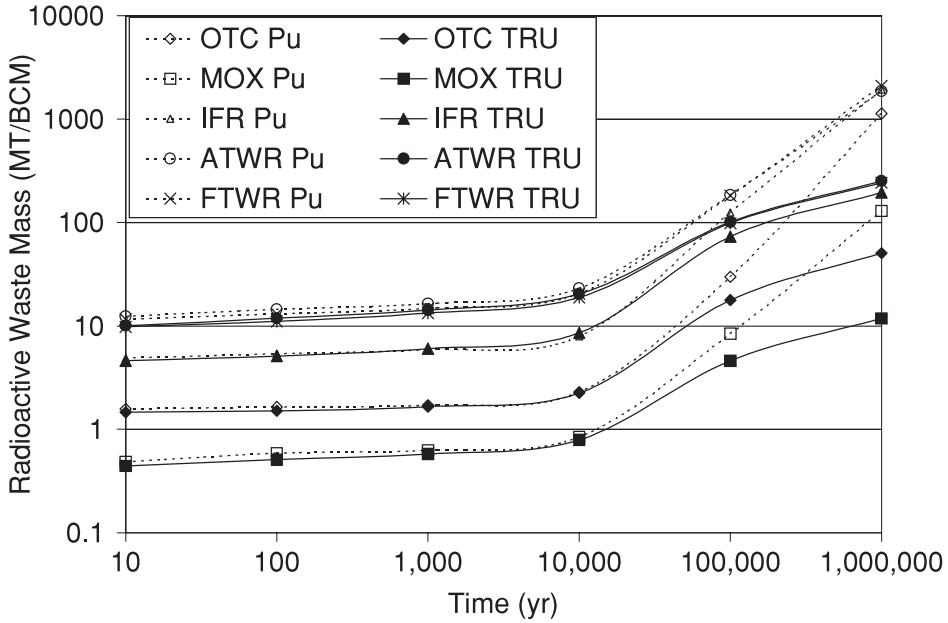

Fig. 13. Mass of radioactive waste containing one BCM (dashed lines = plutonium only; solid lines = all transuranics).

significantly. The actual mass that will need to be processed will be significantly larger than the MRW from the FTWR, ATWR, and IFR EqFCs because the HLW will be further diluted in the final waste form. Over time, the MRW rises as significant fractions of the Pu decay, and at one million years, the MRW is over 1000 MT for all but the spent MOX fuel, which is over 100 MT. Inclusion of the MA has little effect on the MRW until well beyond 10 000 yr.

Figure 14 shows the radiation barrier as measured by the *UDR* from the HLW containing one BCM of the Pu and one BCM of the TRU from the OTC SNF, from the single recycle of the Pu from the SNF in MOX fuel, and from repeatedly recycling TRU from the SNF in an FTWR, ATWR, or IFR EqFC. The repeated recycle of the TRU in the FTWR, ATWR, and IFR EqFCs produces HLW streams that will present a significantly larger radiation barrier than the OTC SNF or spent MOX fuel for nearly one million years because the Pu and TRU are distributed in a very large quantity of FP. The radiation barrier falls significantly over time for all fuel cycles. The minimum radiation barrier of the OTC SNF and spent MOX fuel occurs in the 1000 to 10 000-yr time frame, when many hours would be required to receive a lethal dose, hundreds of rem, even neglecting self-shielding and any other shielding that might be provided.

The Pu or TRU from any of the fuel cycles can theoretically be used to produce a nuclear explosive. The Pu from the OTC SNF is significantly degraded relative to WGPu. The proliferation effects of the MOX fuel are mixed relative to the OTC; there is a higher concentration of lower quality material in the spent MOX fuel. Transmutation in an FTWR, ATWR, or IFR will result in a very small quantity of even lower quality material that will be diluted in a large concentration of FP. The HLW from the FTWR, ATWR, and IFR EqFCs would provide a very significant barrier to the proliferation of the repository waste at any time in the future.

VIII. SUMMARY AND CONCLUSIONS

Detailed fuel cycle analyses were performed to evaluate the impacts of further transmutation of SNF on mass flows into HLW and LLW facilities per unit nuclear energy produced, on the potential radiological hazard of the repository wastes, on the capacity of the HLW repositories, and on the proliferation resistance of the material stored in HLW repositories. An SNF composition representative of the current inventory of SNF discharged from LWRs in the OTC was taken as the base case against which the impact of further transmutation was compared.

Relative to the OTC, the impacts on waste management of a fuel cycle based on a single recycle of the Pu from the SNF as MOX fuel in an LWR and of fuel cycles based on repeated recycling of the transuranics from the SNF as metal fuel in liquid-metal–cooled fast reactors were evaluated. Equilibrium fuel cycles corresponding to three fast transmutation reactor concepts were evaluated:

1. a subcritical reactor with a nonfertile transuranics metal fuel and PbLi eutectic coolant, driven by a tokamak D-T fusion neutron source (FTWR)

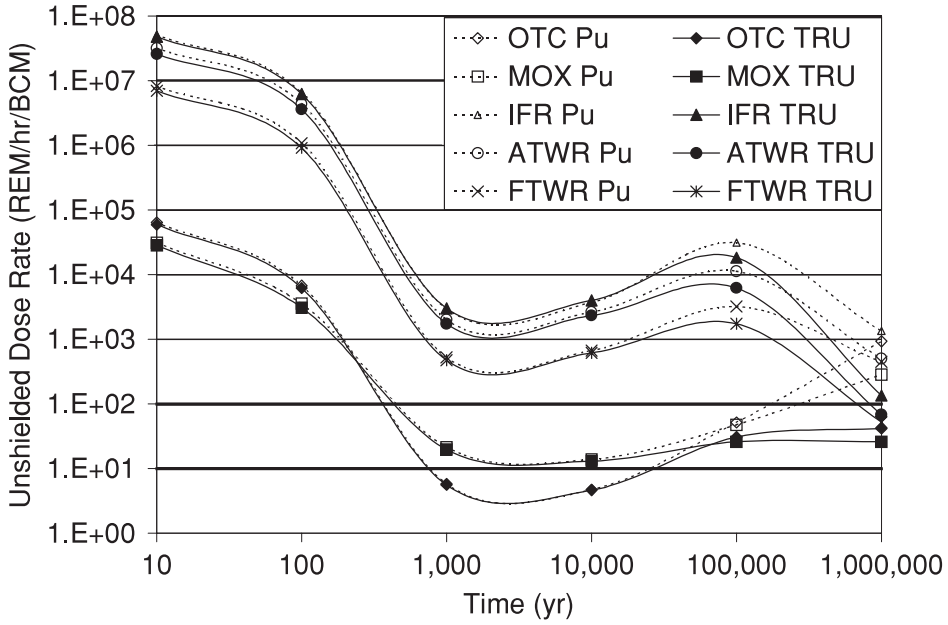

Fig. 14. Radiation barrier of waste containing one BCM (dashed lines = plutonium only; solid lines = all transuranics).

2. a subcritical reactor with a nonfertile transuranics metal fuel and Na coolant, driven by an accelerator spallation neutron source (ATWR)
3. a critical reactor with a fertile transuranics plus uranium metal fuel and Na coolant (IFR).

This study emphasized transmutation fuel cycles that would reduce the transuranic inventory in the HLW, by fissioning the transuranics into FP, most of which are relatively short lived. A single recycle of the Pu in SNF as MOX fuel in an LWR would reduce the HLW transuranic inventory by 25%, and repeated recycling in any of the fast transmutation reactors would eventually reduce the HLW transuranic inventory by >99%. The ATWR and FTWR would be capable of a net transuranic destruction rate that is 2 to 3 times larger than for the IFR because of the production of transuranics in the fertile fuel in the IFR.

The destruction of transuranics by fission also produces nuclear energy, of course, with 1 g of fissioned transuranics producing ~1 MWd of thermal energy. The U.S. inventory of discharged SNF by the end of 2002 would produce ~14 000 GW(thermal)·yr if burned as nonfertile fuel in a fast transmutation reactor, and 2 to 3 times this much if burned as fertile fuel. In an equilibrium system in which there were enough transmutation reactors to recycle the annual discharge rate of LWR SNF, the transmutation reactors would produce a large fraction of the total nuclear power. The nonfertile fuel ATWRs or FTWRs would produce ~25% of the total nuclear power, of which some fraction will be required to drive the neutron source. The fertile fuel IFRs would produce ~40 to 45% of the total nuclear power.

The destruction of the transuranics by repeated recycling in fast transmutation reactors reduces the radiotoxicity of the HLW after the first 100 yr or so, during which many of the short-lived FP decay. The toxicity of all waste streams from the fast transmutation reactors approaches the toxicity of uranium ore in ~500 yr. On the other hand, a single recycle of the Pu from SNF as MOX fuel only slightly reduces the toxicity from all waste streams for LWR operation, and the toxicities of both the OTC and MOX fuel cycles remain above the toxicity of uranium ore for almost 100 000 yr.

Transmutation has a dramatic impact on the HLW composition, of course. With respect to the six troublesome repository isotopes identified in the Yucca Mountain Viability Assessment,[28] the concentrations in HLW from the fast transmutation reactors of the isotopes ^{237}Np, ^{234}U, ^{239}Pu, and ^{242}Pu are all reduced by >99% relative to the OTC SNF, while the concentrations of ^{99}Tc and ^{129}I are increased significantly.

The decay heat source from the HLW is a major determinant in the design of an HLW repository. Repeated recycling of transuranics in fast transmutation reactors results in an HLW decay heat source, after 100 yr, that is about two orders of magnitude lower than for the OTC SNF. This should allow a significant increase in the capacity of the repository. A single recycle of the Pu from SNF as MOX fuel, on the other hand, only achieves a small reduction in decay heat source and would not significantly impact the HLW repository capacity.

The transuranics in the SNF that would be stored intact in an HLW repository in the present OTC scenario could conceivably be a proliferation risk after the radiation has decreased to levels where the SNF is no longer "self-protecting." Repeatedly recycling the transuranics from the SNF in fast transmutation reactors not only destroys a large fraction of the transuranics but also increases the inventory of highly radioactive FP in the waste that is deposited in the HLW repository. The BCM of Pu in the HLW going to the repository in the ATWR, FTWR, or IFR EqFCs is ~50% greater than the BCM of the Pu in the OTC SNF. Furthermore, because the transuranics are more dilute in the HLW from the fast transmutation reactors, the mass of HLW that must be processed to obtain a BCM of Pu is about three times greater for the HLW from the IFR EqFC, and about eight times greater for the HLW from the ATWR and FTWR EqFCs than from the intact OTC SNF. On the other hand, while the BCM of the Pu in the HLW from once-recycled MOX fuel is ~20% greater than the BCM of Pu from the OTC SNF, the mass of spent MOX fuel that must be processed to obtain this BCM is less than one-third the mass of OTC SNF that must be processed, because the Pu is concentrated in making the MOX fuel.

High decay heat and spontaneous fission neutron sources are impediments that enhance the proliferation resistance of nuclear material because they can lead to thermal decomposition of high explosives and premature nuclear detonation, respectively. The decay heat sources from one BCM of Pu from the HLW of the ATWR, FTWR, IFR, and MOX fuel cycles are 7, 6, 3, and 2 times, respectively, larger than the decay heat source from one BCM of Pu from OTC SNF. The SNS for one BCM of Pu is about a factor of 2 larger for Pu from MOX and IFR HLW than for Pu for OTC SNF, and another factor of ~2 larger for Pu from ATWR and FTWR HLW.

The gamma dose rate would act as a deterrent to chemical separation of HLW by technologically unsophisticated parties. The unshielded dose gamma rate from the HLW containing one BCM of Pu from ATWR, FTWR, or IFR HLW is 1 to 2 orders of magnitude greater than the dose rate from the OTC SNF or spent MOX fuel containing one BCM of Pu. The time required for an unshielded worker handling the material to receive a lethal dose never falls below 1 h for the HLW from the ATWR, FTWR, or IFR EqFCs.

The overall conclusion from the above results is that the repeated recycling of the transuranics from SNF would significantly increase the capacity of HLW repositories per unit of nuclear energy produced, significantly increase the nuclear energy production per unit mass of uranium ore mined, significantly reduce the radiotoxicity

of combined HLW and LLW streams per unit of nuclear energy produced, and significantly enhance the proliferation resistance of the material stored in HLW repositories.

While these studies indicate some advantages (e.g., larger net TRU destruction rate per unit power) for subcritical fast transmutation reactors, these advantages were largely associated with the use of nonfertile transuranic fuel in the subcritical reactors and fertile transuranic plus uranium fuel in the critical reactor. The need to use fertile fuel in critical reactors arises from the need to have ^{238}U to provide a significant negative Doppler temperature coefficient of reactivity. The larger margin to prompt critical in a subcritical reactor may be a real advantage in this respect, but this issue is beyond the scope of the present paper. We note current efforts[29] to develop a design for a critical transmutation reactor with nonfertile fuel.

APPENDIX A

FTWR

The FTWR (Refs. 12 and 13) is a 3000-MW(thermal) subcritical reactor driven by a D-T tokamak neutron source. The design of the FTWR was an adaptation of the heavy-metal–cooled, metal-fueled subcritical reactor being studied for ATWR systems.[4,30] The goal of the design was to use nuclear and processing technologies that either exist or are being developed for the ATWR, to the maximum extent possible. The nuclear design analysis supporting the FTWR is presented in Ref. 13.

The fusion neutron source is a D-T tokamak with a major radius of 3.1 m and a minor radius of 0.89 m capable of generating up to 150 MW of fusion power and a neutron source of 5.3×10^{19} s^{-1} (Ref. 12). The fusion source design constrains the geometry of the subcritical reactor. Figure A.1 shows the arrangement of the major components of the FTWR.

The subcritical reactor is a 40-cm-thick by 2.3-m high annulus centered on the midplane and located just outboard of the tokamak plasma (Fig. A.1). The subcritical reactor contains 360 hexagonal fuel assemblies and 180 half-assemblies with a 16.1-cm pitch, grouped into 90 reactor segments aligned with the first-wall segments and circumscribing the tokamak plasma. The arrangement of two of these reactor and first-wall segments is shown in Fig. A.2. The fuel is a transuranic zirconium alloy (TRU-10Zr) dispersed in a zirconium matrix and clad with a steel similar to HT-9. There are 217 pins, 210 fuel pins and 7 structural pins per assembly, with a triangular pitch of 1.1 cm. Most of the volume within the toroidal field coils not occupied by the plasma neutron source and the reactor is occupied by the reflector and shield. The design of the FTWR is given in Ref. 12.

The requirement for tritium self-sufficiency necessitates the incorporation of lithium into the reactor and/or

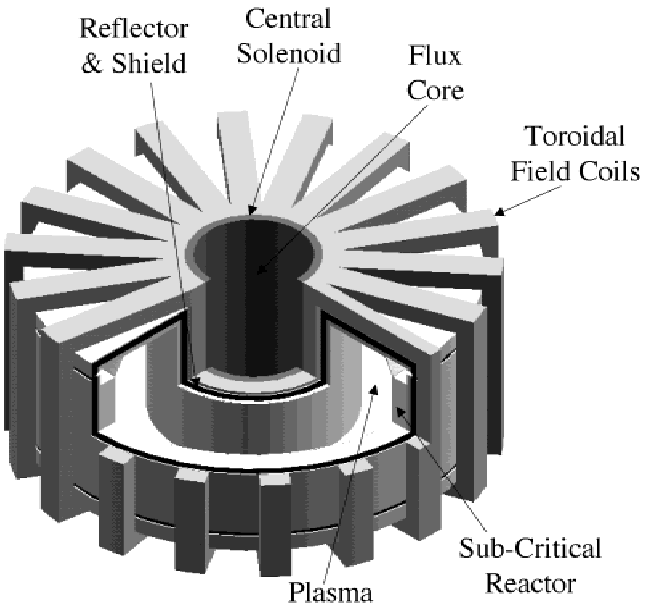

Fig. A.1. FTWR schematic.

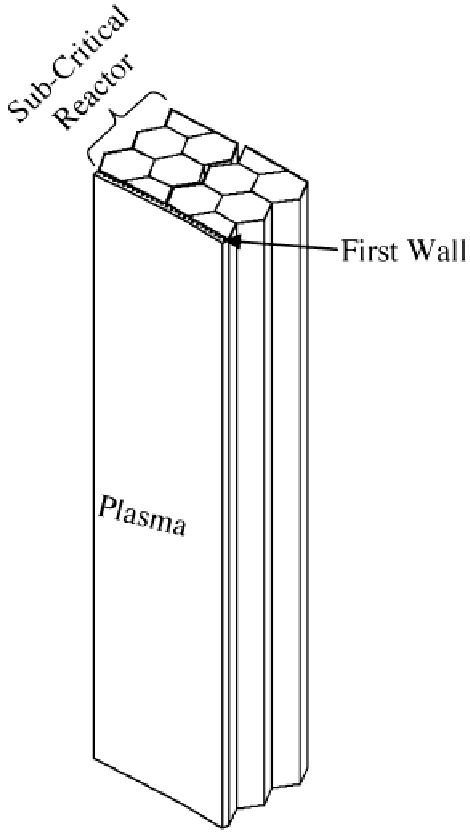

Fig. A.2. Transmutation reactor configuration outboard of plasma chamber.

reflector-shield system. This can be done in either solid or liquid form. Design concepts have been developed for a lithium lead (Li17-Pb83) coolant-breeder design and for a lead bismuth eutectic (LBE) plus solid (Li_2O) breeder design. Both designs performed similarly. The Li17-Pb83 design results are discussed in this paper.

The FTWR operates on a 564-day cycle with a five-batch refueling scheme. The BOC fuel loading is 24.3 MT of heavy metal, of which 23.3 MT are TRU and the rest a small quantity of uranium, mostly ^{234}U that has accumulated as a result of radioactive decay. The LWR SNF feed used in this analysis was the representative SNF given in Table I.

APPENDIX B

ATWR

The ATWR (Refs. 4 and 30) is an 840-MW(thermal) sodium-cooled, metal-fueled, subcritical reactor driven by an accelerator spallation neutron source being studied as part of the DOE Advanced Accelerator Applications Program. Multiple ATWRs will be operated by a single large proton accelerator.

The ATWR design consists of a central LBE target/buffer surrounded by 132 hexagonal fuel assemblies. The subcritical reactor is an annulus with an inner radius of 37 cm, an outer radius of 105 cm, and a fuel height of 113 cm. The fuel is the TRU-40Zr metal alloy clad with a steel similar to HT-9. Additional details of the ATWR design are provided in Ref. 30.

The ATWR design and fuel cycle were developed for SNF feed from an advanced light-water reactor (ALWR) operated to 50 GWd/MTU. The composition of the ALWR SNF is given in Table I. For the purposes of this paper, the representative SNF given in Table I was used in this analysis. No other changes were made to the reactor design or fuel cycle. The ATWR operates on a 140-day cycle with two fuel zones operating on a seven- and eight-batch refueling scheme.

The BOC fuel loading for a single ATWR with the representative SNF feed is 2.44 MT of heavy metal, of which 2.39 MT are TRU and the rest a small quantity of uranium, mostly ^{234}U that has accumulated as a result of radioactive decay. The heavy metal loading for the ATWR given in Ref. 4 was 2.71 for the ALWR SNF feed. Other parameters were very similar for the two SNF feeds. The discharge TRU burnup calculated in this analysis was 31.0% compared with 29.2% for the ALWR feed. The burnup reactivity loss calculated in this analysis was 4.30% compared with 4.14% for the ALWR feed. The TRU charge enrichment was 98.4% in this analysis compared with 98.5% for the ALWR feed. Overall, the results suggest that differences in the SNF feed will have some impact on the design of the ATWR and/or the fuel cycle, but these would not be expected to be dramatic or impact the conclusions of this study.

APPENDIX C

IFR

The IFR (Refs. 4 and 31) is an 840-MW(thermal) sodium-cooled, metal-fueled, critical reactor. The IFR is a cylindrical reactor with a radius of 161 cm and a fuel height of 48 cm. The height of the IFR is reduced to increase leakage and reduce the conversion rate. The fuel is the U/TRU-10Zr metal alloy with a charge of ~72% U and 28% TRU. The fuel is clad with a steel similar to HT-9. Additional details of the IFR design are provided in Ref. 31.

The IFR design and fuel cycle were developed for SNF feed from an ALWR operated to 50 GWd/MTU. For the purposes of this paper, the representative SNF given in Table I was used in this analysis. No other changes were made to the reactor design or fuel cycle. The IFR operates on a 310-day cycle with a seven-batch refueling scheme.

The BOC fuel loading for a single IFR is 13.89 MT of heavy metal of which 3.78 MT are TRU. The heavy metal loading for the IFR given in Ref. 4 was 13.89 for the ALWR SNF feed. The discharge TRU burnup calculated in this analysis was 18.0% compared with 18.6% for the ALWR feed. The burnup reactivity loss calculated in this analysis was 2.08% compared with 2.34% for the ALWR feed. The TRU charge enrichment was 28.0% in this analysis compared with 32.4% for the ALWR feed. In this analysis 20% less TRU is fissioned in each pass through the reactor than was calculated for the ALWR feed. The conversion rate in this analysis is 0.59 compared with 0.51 for the ALWR feed. The TRU reduction is 99.44% for this analysis compared with 99.5% for the ALWR feed. Overall, there are some significant differences with the results calculated in Ref. 4 because of the different SNF feeds, but better optimization for the SNF feed used in this analysis should not significantly affect the conclusions of this study.

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