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Co-Extraction Impacts on LWR and Fast Reactor Fuel Cycles

Nuclear Engineering Division

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ABSTRACT

A systematic investigation of the impact of the co-extraction COEX™ process on reactor performance has been performed. The proliferation implication of the process was also evaluated using the critical mass, radioactivity, decay heat and neutron and gamma source rates and gamma doses as indicators.

The use of LWR-spent-uranium-based MOX fuel results in a higher initial plutonium content requirement in an LWR MOX core than if natural uranium based MOX fuel is used (by about 1%); the plutonium for both cases is derived from the spent LWR spent fuel. More transuranics are consequently discharged in the spent fuel of the MOX core. The presence of U-236 in the initial fuel was also found to result in higher content of Np-237 in the spent MOX fuel and less consumption of Pu-238 and Am-241 in the MOX core. The higher quantities of Np-237 (factor of 5), Pu-238 (20%) and Am-241 (14%) decrease the effective repository utilization, relative to the use of natural uranium in the PWR MOX core. Additionally, the minor actinides continue to accumulate in the fuel cycle, even if the U-Pu co-extraction products are continuously recycled in the PWR cores, and thus a solution is required for the minor actinides.

The utilization of plutonium derived from LWR spent fuel versus weapons-grade plutonium for the startup core of a 1,000 MWT advanced burner fast reactor (ABR) increases the TRU content by about 4%. Differences are negligible for the equilibrium recycle core. The impact of using reactor spent uranium instead of depleted uranium was found to be relatively smaller in the fast reactor (TRU content difference less than 0.4%).

The critical masses of the co-extraction products were found to be higher than that of weapons-grade plutonium and the decay heat and radiation sources of the materials (products) were also found to be generally higher than that of weapons-grade plutonium (WG-Pu) in the transuranics content range of 0.1 to 1.0 in the heavy-metal. The magnitude of the differences is higher with the presence of the minor actinides. The critical mass of (Pu,U) MOX fuel derived from the co-extraction process is a factor of ~2.4 greater than that of low enriched uranium (20% U-235); that for the 50-50% Pu-U product of COEX is a factor of ~12 lower. The decay heat from (Pu,U) MOX fuel is 20% less than that of WG-Pu metal for the same mass of material; its neutron and gamma source rates and contact dose are, however, at least 40% higher.

1.0 INTRODUCTION

The Global Nuclear Energy Partnership (GNEP) reference scenario proposes to use the UREX+1a process for the group separation of transuranics (TRU) from light water reactor spent nuclear fuel (LWR SNF). The recovered transuranics would be used in fuels for advanced recycling fast reactors that are proposed for consuming the highly radioactive and radiotoxic transuranics. The continuous recycling of the TRU in the advanced fast reactors limits the amount of these hazardous materials to be passed to a repository (process losses only) and consequently provides significant benefits to the repository as the effective repository utilization is increased per unit energy produced. The group separation of the transuranics also provides some intrinsic proliferation barriers due to the high radiation fields arising from the presence of higher plutonium isotopes and minor actinides (Np, Am, and Cm). (The fuel would still require physical protection, particularly as the post-irradiation cooling time increases.)

The group separation process however is new and has not been used in any commercial fuel cycle and thus comes with economic penalties and technical uncertainties. The use of remote fabrication required for the TRU containing fuels will incur additional cost to the fuel cycle. The TRU fuel qualification has technical uncertainties that could potentially delay the utilization of the advanced recycling reactors in the GNEP reference scenario. Consequently, alternative fuel cycles are also being evaluated in addition to the reference one. [1]

One such alternative fuel cycle is the use of the uranium-plutonium co-extraction process for the recovery of the actinides. The COEX^{TM} approach that has been proposed by AREVA is an example of a co-extraction process. [1,2] In the COEX process, two major products are pure uranium and a mixture of plutonium and uranium derived from the LWR SNF. These streams are then subsequently used in making MOX fuel for recycle in existing or advanced light water reactors. A primary attraction of the COEX process is that coupling it to the use of (U,Pu) MOX fuels in LWR could provide a stop-gap measure, or bridging mechanism by providing a consumption path for U-Pu fuel pending the introduction of the advanced recycling reactors. For the U.S., since there is an existing fleet of LWRs, and new advanced LWRs (ALWRs) are anticipated in the near-term, the consumption path could accept large quantities of (U,Pu) material, pending the deployment of fast reactors provided these reactors were licensed to use

this fuel. [1] Additionally, MOX fuels are routinely used in LWRs in Europe and Japan, indicating the feasibility of this approach. Advantages of the COEX process include: (1) Early LWR spent fuel separations at commercial-scale and reduced technical risk associated with initial fuel and separations process; and (2) Lower costs for initial driver fuel qualification and separations front-end process development. [1]

A potential shortcoming of COEX is that the minor actinides (MA) in the spent fuel would however not be recovered and would be dumped as waste. Since the minor actinides constitute a significant fraction of the hazard in spent nuclear fuel, a solution would have to be found for them in order to achieve effective repository utilization. Suggestions have included using the MA as targets in fast reactors [1] or even in thermal reactors. The economic penalties associated with target development, fabrication, and management (handling and utilization) in the fuel cycle requires evaluation. The use of MA targets will also necessitate additional physical protection requirements since some of these materials (e.g., Np-237) are of proliferation concern.

This co-extraction alternative (without targets) is also being promoted as a proliferation-resistant separation process that could be used in advanced nuclear fuel cycles. This assertion is however debatable since the separation of the co-extracted uranium and plutonium involves an additional chemical process that is known to most fuel chemistry experts. However, the use of such a separation process in a fuel cycle state under proper safeguards makes it a plausible alternative. Under this premise, an assessment of the impact of the COEX process on PWR and sodium cooled fast core performance has been completed, along with the evaluation of the proliferation indices that could be used to infer proliferation resistance.

In chapter 2, the impacts of using fuels derived from spent uranium and plutonium (COEX products) in a PWR fuel cycle is presented, by comparing reactivity balances and fuel compositions following irradiation of the fuels in a MOX-fueled core. Fast reactor studies in which the spent uranium and plutonium materials are used as fuels have been done to assess the impacts of these fuels on core performance. The results of the studies are summarized in Chapter 3.

The critical mass, radioactivity, decay heat, neutron and gamma sources, and gamma doses arising from the co-extracted plutonium-uranium products have been evaluated and compared to those from weapons-grade plutonium, in order to provide indication of the level of proliferation resistance of the materials. Results are summarized in Section 4. Conclusions from the study are presented in Section 5.

2.0 IMPACT OF COEX PROCESS ON PWR FUEL CYCLE

A study of the impact of the COEX process on the neutronic performance of a PWR MOX core has been performed. The recycled product of the COEX process is assumed to include co-extracted uranium and plutonium in a 50-50 mixture. To make MOX fuel for the PWR, the Pu-U mixture would be diluted using the recovered reactor-spent uranium from the COEX process; in this case, the MOX fuel contains reactor-grade plutonium and reactor-spent uranium. The impact of the reactor-spent uranium on core reactivity and material balances has been estimated by comparing results to those for a MOX core using the reactor-grade plutonium and natural uranium or depleted uranium. For delineation of cases and clarity, the MOX fuel derived from the COEX path is designated MOX-CX (MOX using reactor-spent uranium), that using natural uranium is designated MOX-NU, and that using depleted uranium is designated MOX-DU. The results of the study are provided in Sections 2.1 to 2.4.

In Section 2.1, a description of the PWR assembly used in the study is presented along with input fuel vectors and associated modeling assumptions. The linear reactivity model used to represent the core reactivity balance is discussed in Section 2.2. Core reactivity balance results are presented in Section 2.3, and core material mass composition data are provided in Section 2.4. Conclusions for these sections are presented in Section 2.5.

2.1 Description of PWR Assembly and Core

A 17x17 PWR assembly with design parameters like those of a Framatome/COGEMA Fuels Advanced Mark-BW assembly was assumed in this study [3]. A homogenous core model consisting of only MOX assemblies was modeled: some parameters describing the assembly design appear in Table 1. The effect of assembly grid spacers was neglected, and the study was restricted to two-dimensional (x-y) analysis. As this was a scoping study to primarily investigate the reactivity effects of the various uranium compositions, all assemblies were loaded homogenously with one of the MOX fuels and no detailed treatment of the power peaking was performed.

For the purpose of comparing results it was assumed that the MOX assembly has a discharge burnup of 45 GWd/t. [3] The core is assumed to have a power rating of 3,000 MWt and to contain 194 assemblies. A three-batch fuel management is also assumed for the core.

Table 1. MOX Assembly Design Parameters.

Assembly size	17 x 17 pins
Number of fuel pins	264
Number of guide tubes	24
Number of instrumentation tubes	1
Fuel rod pitch (cm)	1.2598
Inter-assembly gap (cm)	0.08
Fuel pellet radius (cm)	0.4096
Clad inner radius (cm)	0.4178
Clad outer radius (cm)	0.4750
Smeared fuel density (g/cm^3) (pellet at 95% T.D., 1.2% pellet dishing)	9.88
Fuel mass (kg/assembly)	523.4
Fuel mass (kg HM/assembly)	461.3
Zircaloy-4 clad density (g/cm^3)	6.5
GT/IT inner radius (cm)	0.5715
GT/IT outer radius (cm)	0.6121
Specific power density (MW/MTIHM)	33.69*
Fuel temperature (K)	900.0
Cladding temperature (K)	581.0
Bulk coolant temperature (K)	581.0
Coolant density (g/cm^3)	0.72

*Based on reactor loading of 193 assemblies and total core power of 3,000 MWt.

The initial vectors of uranium and plutonium for the MOX-NU, MOX-DU and MOX-CX fuels are summarized in Table 2, along with the reactor-grade plutonium vector. The spent

uranium vector and the reactor-grade plutonium vector were derived from a PWR-UO_2 spent nuclear fuel with a burnup of 50 GWd/t that is cooled for ten years and reprocessed to recover the plutonium. [3] It is additionally assumed that the fuel is loaded into the MOX-containing core two years after fabrication. In this period, some of the Pu-241 has beta decay into Am-241. This post-fabrication buildup of Am-241 is modeled by the Am-241 content of the plutonium vector.

Table 2. Uranium and Plutonium Vectors in MOX-CX, MOX-NU and MOX-DU.

Uranium Vector (%)				Plutonium Vector (%)	
Nuclide	MOX-NU	MOX-DU	MOX-CX*	Nuclide	
U-234	0.0054		0.02	Pu-238	3.136
U-235	0.711	0.2	0.701	Pu-239	56.379
U-236	--		0.585	Pu-240	26.626
U-238	99.2836	99.8	98.694	Pu-241	7.290
				Pu-242	5.832
				Am-241	0.736

*The MOX-CX uranium vector was obtained from Ref. 3. Other sources have indicated that the U-235 content in spent fuel uranium could be as high as 0.85%. This would make MOX-CX more reactive.

The primary differences between the three uranium vectors include:

1. Compared to natural uranium (MOX-NU), the MOX-CX fuel (spent uranium) contains a significant fraction of U-236 (predominantly) and U-234. These isotopes of uranium are neutron absorbers in PWR cores. The MOX-CX fuel also contains a significant fraction of U-235, which is the product of enriched U-235 in the original fuel of the PWR UO_2 core that has been depleted by neutron irradiation (i.e., used for making power).

2. The MOX-DU fuel (depleted uranium), recovered from fuel enrichment operations in the front end of the fuel cycle, contains U-238 primarily and a small fraction of U-235. Its U-235 content is a factor of 3½ times less that in MOX-NU or MOX-CX.

2.2 Fuel Cycle Analysis Results

A linear reactivity model [4] using assembly-level WIMS9 [5] calculations was utilized to estimate the performance of a PWR core loaded entirely with MOX fuel. In the model, the whole-core leakage and non-assembly absorption are assumed to give a reactivity penalty of

~3.5% Δk/k: that is, the critical burnup is defined as the burnup at which the k-infinity of the assembly model is equal to 1.035. The linear reactivity model gives the relationship between the predicted assembly discharge burnup B_d and calculated critical burnup B_c:

$$B_d = \frac{2n}{n+1} B_c, \qquad (1)$$

where *n* denotes the number of fuel management batches. A three-batch fuel management has been assumed for the PWR calculations in this study.

2.3 Core Reactivity Balance

Two sets of reactivity balance calculations were performed. The first was a simple study in which the three MOX (MOX-NU, MOX-DU, MOX-CX) assemblies were loaded with the same plutonium oxide (PuO_2) content of 8.5% in the heavy-metal. Results for the reactivity letdown curves for the different fuels are given in Figure 1. It is seen that the MOX-NU (natural) case is the most reactive due to its higher U-235 content and no U-236 in the fuel. The reactivity letdown behaviors of the MOX-CX (spent) and MOX-NU (depleted) fuels are quite similar, although the MOX-CX fuel appears to be more reactive. The two fuels are less reactive than MOX-NU fuel.

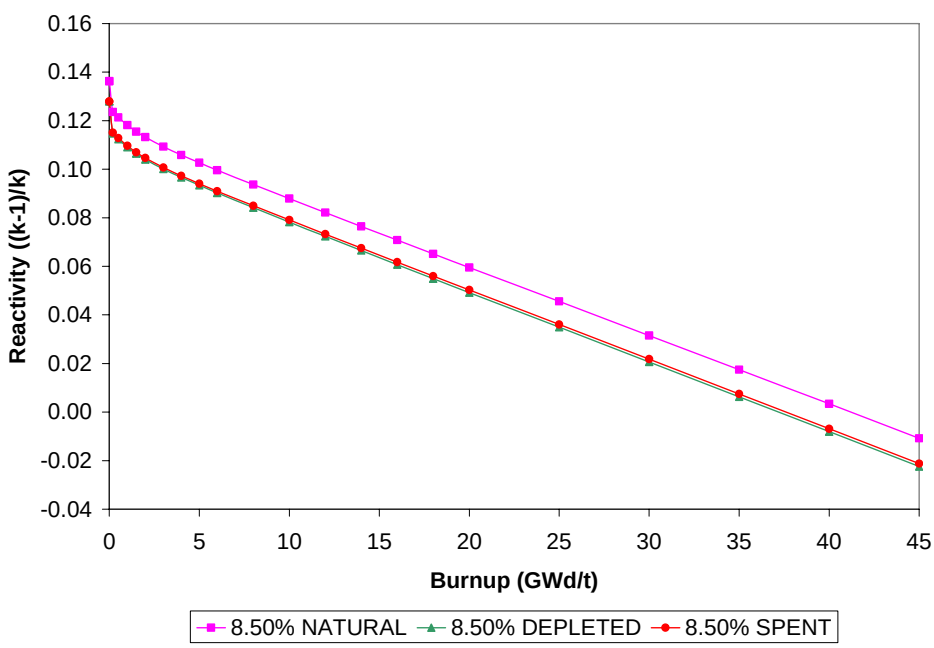

Figure 1. Reactivity versus Burnup for Three Different Uranium Vectors.

These differences in reactivity letdown are attributed to the different U-235 and U-236 contents of the fuels. It was found that the presence of U-236 in the MOX-CX fuel results in a lower reactivity than for a MOX-NU fuel having the same initial PuO_2 content in the MOX fuel. This is because U-236 is a neutron absorber. The lower U-235 content of the depleted uranium MOX fuel (MOX-DU) also leads to a lower reactivity than for MOX-NU. The slightly lower initial reactivity of the MOX-DU fuel relative to MOX-CX is associated to the trade-off between the low U-235 content and no U-236 in the MOX-DU fuel. These lower initial k-infinity values indicate shorter core cycle lengths and fuel discharge burnups (see Table 3).

Table 3. Discharge Burnups Calculated with the Linear Reactivity Model.

Core Type	MOX-NU	MOX-CX	MOX-DU
Type of Uranium Vector	Natural	Spent	Depleted
Critical Burnup (B_c) (GWd/t)	29.18	25.77	25.37
LRM Discharge Burnup (B_d) for 3 batches (GWd/t)	43.76	38.66	38.05
Cycle length per batch	433	382	376

*All cases had initial PuO_2 content of 8.50%.

For meaningful comparison, a second set of calculations were performed, designing for the same discharge burnup of 45 GWd/t in the three PWR MOX cores. *It was found that the initial PuO_2 contents of the MOX must be 9.75%, 9.7% and 8.75% when using MOX-DU, MOX-CX and MOX-NU fuels, respectively.* Thus, a higher PuO_2 content (by ~1%) is required in the PWR core using MOX-CX compared to MOX-NU. The MOX-DU fuel required the highest PuO_2 content due to its lowest fuel reactivity, compared to the other two cases.

2.4 Comparison of Mass Contents at Discharge

The discharge compositions of the three cores, normalized to the initial heavy metal metric tonnes (IHMMT), have been evaluated and compared, in order to investigate the impact of uranium vector. Table 4 is a summary of the mass-normalized initial and discharge composition of these cores and indicates some large differences in the compositions at the common discharge burnup of 45 GWd/t. These values are derived from the WIMS9 calculations for the assemblies.

Table 4. Comparison of MOX Fuels at Discharge Burnup.

Spent Fuel Mass per Initial Heavy Metal Metric Ton (g/IHMMT)						
Case	MOX-NU		MOX-CX		MOX-DU	
PuO_2 Content, %	8.75		9.70		9.75	
	Charge	Discharge	Charge	Discharge	Charge	Discharge
U-234	45.4	86.0	181.5	182.8	0.0	64.2
U-235	6556.3	3661.6	6329.7	3724.2	1803.6	1038.8
U-236	0.0	630.7	5286.1	4930.7	0.0	178.8
U-237	0.0	2.1	0.0	8.0	0.0	1.3
U-238	905853.3	878635.8	891134.1	864677.9	900640.6	873613.7
U total	912455.0	883016.2	902931.3	873523.6	902444.2	874896.8
Np-237	0.0	157.3	0.0	803.8	0.0	109.8
Np-239	0.0	64.5	0.0	62.3	0.0	63.7
Np total	0.0	221.8	0.0	866.1	0.0	173.4
Pu-238	2745.2	2493.9	3040.0	2987.8	3062.7	2772.7
Pu-239	49367.0	27494.7	54732.5	31736.6	55004.0	31085.3
Pu-240	23311.2	20904.8	25840.7	23454.0	25976.2	23474.0
Pu-241	6386.3	10733.5	7078.4	11624.8	7112.6	11667.2
Pu-242	5104.6	5934.2	5660.4	6365.3	5694.4	6471.8
Pu total	86914.3	67561.1	96352.1	76168.6	96850.0	75471.1
Am-241	646.6	1089.8	714.6	1246.5	714.6	1227.5
Am-242m	0.0	22.5	0.0	26.5	0.0	25.9
Am-243	0.0	1452.7	0.0	1513.7	0.0	1544.6
Am total	646.6	2565.1	714.6	2786.7	714.6	2798.0
Cm-242	0.0	186.0	0.0	192.9	0.0	197.4
Cm-243	0.0	7.6	0.0	7.9	0.0	8.1
Cm-244	0.0	778.6	0.0	774.1	0.0	810.0
Cm-245	0.0	94.2	0.0	93.3	0.0	98.3
Cm total	0.0	1066.5	0.0	1068.3	0.0	1113.8
Total TRU	87561	71414	97067	80890	97565	79556
Total HM	1000016	954431	999998	954413	1000009	954453

Significant differences are observed in the discharge quantity of Np-237. This arises because of the production of Np-237 from the U-236 initially present in the MOX-CX fuel. The U-236 is a precursor for creating neptunium and the lower plutonium isotopes through (n,γ) reactions. As expected, the results for the MOX-NU and MOX-DU fuels are quite similar, with the main difference being that even less neptunium is created during irradiation of MOX-DU fuel because of the lower initial U-235 content. However, the discharge fuel of the MOX-DU core contains more plutonium than that of the natural uranium MOX, due to the higher initial

plutonium content. This trend is the same for the discharge plutonium content in MOX-CX, which is higher than that of MOX-NU.

Other differences are better illustrated by comparing the fractional consumption or production ($\Delta M/M_i * 100$) of the nuclides, where M_i is the initial mass, and ΔM is the difference in mass between the discharge and initial masses for a given nuclide or group (e.g., heavy metal (HM) and TRU). Fractional consumption values for the three cases are summarized in Table 5. A positive value indicates production while a negative value indicates consumption over the irradiation period of the fuel. As the initial fuels contain no neptunium, curium, or higher isotopes of americium beyond Am-241, these isotopes are not listed, though they are produced in significant amounts over the fuel irradiation period, as shown in Table 4. The higher isotopes of each element tend to be produced as the lower-numbered isotopes are either fissioned or transmuted to the higher-numbered isotopes through absorption of neutrons.

Table 5. Actinide Consumption at Discharge Burnup (%).

Case	MOX-NU	MOX-CX	MOX-DU
PuO_2 Content, %	8.75	9.70	9.75
Discharge Burnup, GWd/t	45	45	45
U-234	89.5	0.7	--
U-235	-44.2	-41.2	-42.4
U-236	--	-6.7	--
U-238	-3.0	-3.0	-3.0
U total	-3.2	-3.3	-3.1
Pu-238	-9.2	-1.7	-9.5
Pu-239	-44.3	-42.0	-43.80
Pu-240	-10.3	-9.2	-9.6
Pu-241	68.1	64.2	64.0
Pu-242	16.3	12.5	13.7
Pu total	-22.3	-21.0	-22.1
Am-241	68.6	74.4	71.8
Am total	296.7	290.0	291.5
Total TRU	-18.4	-16.7	-18.5
Total Heavy Metal	-4.6	-4.6	-4.6

It can be seen in Table 5 that using MOX-CX (spent uranium based fuel) instead of MOX-NU (natural uranium based fuel) produces the most Am-241, but least total Am and less of the higher Pu isotopes, while the MOX-DU (depleted uranium) results tend to be in between those for the other two fuel types. The MOX-DU consumes the most Pu-238, however. The least consumption of Pu-238 is obtained for the MOX-CX case. This trend for Pu-238 is attributed to its additional production via neutron captures from the U-236 initially present in MOX-CX. It is also interesting to note that while the total heavy metal consumed is equal (as the fuels are all at equal burnups), less totals TRU (mostly Pu) is consumed in the MOX-CX. As aforementioned, the presence of U-236 significantly affects this fuel, and as both neptunium and plutonium are counted in the total TRU amount, the production of these isotopes in the spent uranium MOX (MOX-CX) reduces the total overall consumption of TRU in that fuel form. (For all the cases, the overall consumption of TRU is affected strongly by U-238 conversion to plutonium.)

Table 6 shows the differences between the plutonium compositions at discharge given the different initial uranium loading. It is seen that more plutonium remains post-irradiation per initial heavy-metal in the MOX-CX fuel than for MOX-NU, again due to the initial U-236. While the Pu-238 fraction of the plutonium in the fuel increases (from charge to discharge) using natural and depleted uranium MOX fuels, it increases more if MOX with spent fuel uranium is used. The plutonium compositions of these MOX fuels follow the same trend over burnup. In general, the discharge plutonium vectors are quite similar.

Table 6. Plutonium Compositions at Discharge Burnup (45 GWd/t).

Case	MOX-NU	MOX-CX	MOX-DU	Charged vector for all cases
Initial PuO_2 Content, %	8.75	9.70	9.75	
Burnup, GWd/t	45.08	45.08	45.03	
Plutonium Vector, %				
Pu-238	3.7	3.9	3.7	3.2
Pu-239	40.7	41.7	41.2	56.8
Pu-240	30.9	30.8	31.1	26.8
Pu-241	15.9	15.3	15.5	7.4
Pu-242	8.8	8.4	8.6	5.9
Pu-total per initial fuel, %*	77.7	79.1	77.9	100.0
Fissile (Pu239 + Pu241), %	56.58	56.93	56.65	64.15

*Pu-total at discharge as fraction of initial Pu loading.

2.5 Conclusions on Utilization of Spent Uranium in PWR MOX Fuel Cycle

The use of spent uranium instead of natural uranium in a PWR core results in the need for higher plutonium content to meet the reactivity requirement for a given fuel burnup level. This higher plutonium content and the presence of U-236 in the spent uranium fuel result in over 13% higher TRU (80890 / 71414) in the discharged fuel per initial heavy-metal. Additionally, it was observed that significantly more Np-237 (about a factor of 5 more) is contained in the spent fuel of a core using spent uranium based fuel than that using natural uranium based fuel, primarily because of the presence of U-236. Furthermore, less Pu-238 is consumed and more Am-241 is produced due to the use of spent uranium in the LWR MOX fuel cycle. These higher quantities of Np-237, Pu-238 and Am-241 (and of Pu and TRU) in the waste would decrease the effective repository utilization, relative to the use of natural uranium in the PWR MOX core; the Pu-238 and Am-241 masses at discharge are 20% and 14% higher, respectively, for the PWR MOX core using spent uranium.

There are three potential paths for the discharged PWR MOX fuel. The spent fuel could be: (1) sent to a repository; (2) recycled or multi-recycled in MOX PWRs; or (3) multi-recycled in fast reactors. The implications of these paths have not been considered in the current study, but prior AAA/AFCI studies have evaluated such paths [1,6,7,8]. Limited recycle is not expected to provide significant benefits to effective repository utilization. While continuous recycling of the material in PWRs is feasible neutronically and from a safety point of view, [9] the minor actinides would however continue to accumulate and a solution must be found for removing them from the waste stream if significant gains are to be made for effective repository capacity. Consumption in fast reactors and the use of minor actinides containing targets in advanced reactors (thermal and fast) are potential approaches for minimizing the minor actinides passing to the repository. [1]

3.0 IMPACT OF COEX PROCESS ON FAST REACTOR FUEL CYCLE

A study has been performed to evaluate the impact of using uranium and plutonium (Pu) derived from the COEX process in the reference 1,000 MWt advanced burner fast reactor (ABR) design. [10] Both metal and oxide fueled ABRs are currently being considered under the AFCI/GNEP program. For this study, metallic fuel is assumed for the ABR. Both the equilibrium recycle and startup cores of the ABR are considered. It is noted that the ABR design used in this study is the current *snapshot* of the core, as there is no official ABR design at this time. Results from the study, including core design data and modeling assumptions are given in Sections 3.1 to 3.3. Only the direct utilization of the co-extraction uranium-plutonium products in the fast reactor fuel cycle has been considered. But since a solution has to be found for the minor actinides that would buildup from thermal recycle, a potential approach might be to consume the remaining transuranics in a fast reactor. The impact of the double-tier approach was not evaluated in this study, but the results of pertinent previous studies are discussed (see Section 3.4).

3.1 ABR Core Design

A recent 1,000 MWt ABR core design [10] has been employed to evaluate the impacts of using LWR spent uranium and plutonium as fuel material for the fast reactor. This design allows a 1-year cycle length with a control system that provides sufficient shutdown margin with the reactivity worth of each single control assembly less than 1$. This was achieved by using a lower specific power density than in previous design studies. Figure 2 displays the radial configuration of the ABR core. The ABR core is assumed to use ternary metallic fuel, U-TRU-10Zr. The core consists of 180 driver, 114 reflector, and 66 radial shield assemblies, and 15 primary and 4 secondary control assemblies. The core is divided into inner and outer core zones, which are composed of 78 and 102 fuel assemblies, respectively, for the purpose of minimizing the power peaking. Two independent safety-grade reactivity control systems are used. The primary control system consists of 3 control assemblies in the fourth row and 12 control assemblies in the seventh row, and the secondary control system contains 4 control assemblies located at the core center (1) and in the fourth row (3).

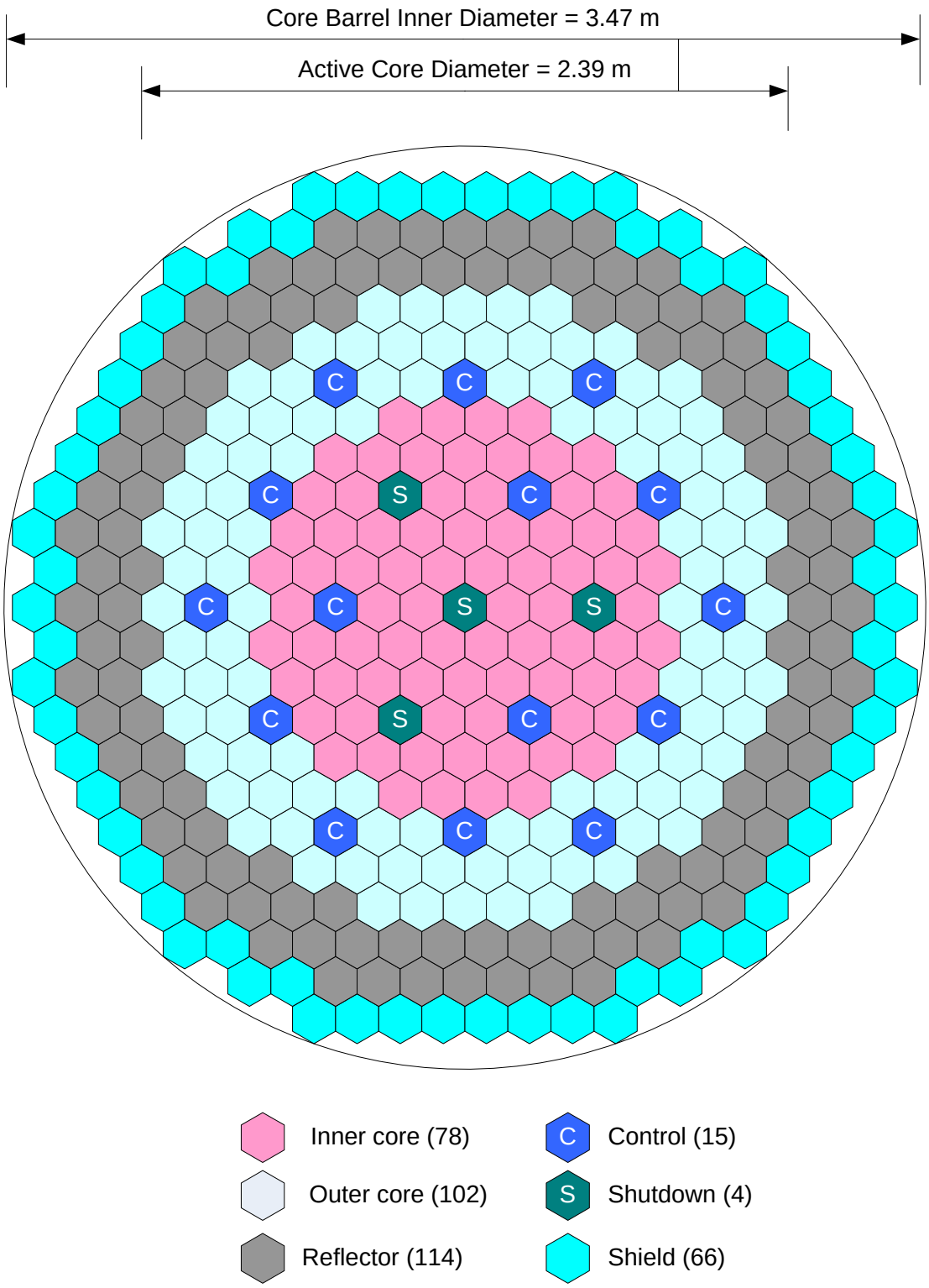

Figure 2. Planar Configuration of Reference 1,000 MWt Metal Fuel Core Concept. [10]

Figure 3 shows the schematics of fuel assembly, and the design parameters of fuel, reflector, shield, and control assemblies are provided in Table 7. The fuel assembly contains 271 fuel pins with 7.55 mm diameter and 0.56 mm cladding thickness, and the fuel volume fraction is 29.2%. Fuel pins are made of sealed cladding containing a metallic fuel column of 81.3 cm length. Sodium is used as the initial thermal bond between the fuel column and the cladding. The pin size is within the ALMR pin diameter range of 7.4 - 8.3 mm. [10] The fuel smeared density is 75%. The fuel pin is helically wrapped with wire to maintain the pin spacing so that the coolant can flow freely through the pin bundle, and the wire-wrap helical pitch is 20.32 cm. A 124.5 cm long fission gas plenum is located above the fuel slug and sodium bond.

The control assemblies consist of a B_4C absorber bundle contained in a duct. The absorber bundle is a closely packed array of seven tubes containing compacted boron carbide pellets. To minimize the single control assembly reactivity fault, two different B-10 enrichments were used for the primary control assemblies. Using natural boron (B-10 enrichment of 19.9 atom %) for the control assemblies in the fourth row, the B-10 enrichment of the control assemblies in the seventh row was determined such that the reactivity fault of the control assembly in the fourth row becomes similar to that of a control assembly in the seventh row. The resulting B-10 enrichment of the control assemblies in the seventh row was 60%. For the secondary control assemblies, natural boron was used.

The reflector assembly contains 91 solid HT-9 pins arranged in a triangular pitch array. The HT-9 pin volume fraction is 75.3% and the duct volume fraction is 9.2%, yielding a total HT-9 volume fraction of 84.5%. The shield assembly is composed of HT-9 cladding and B_4C absorber. It consists of 19 thick HT-9 tubes containing boron carbide pellets. The natural boron is used with a smeared B_4C pellet density of 81%. The resulting B_4C and HT-9 volume fractions are 43.1% and 29.7%, respectively.

The REBUS-3/DIF3D code package was used for the fuel cycle evaluations of this study. Region-dependent 21-group neutron cross-section sets generated for fast reactor cores with the ETOE-2/MC2-2/SDX code system, based on ENDF/B-V data, were employed in the calculations. The startup and equilibrium recycle flux calculations were done with the 3-D hexagonal-Z geometry option of the DIF3D-nodal module of the code package.

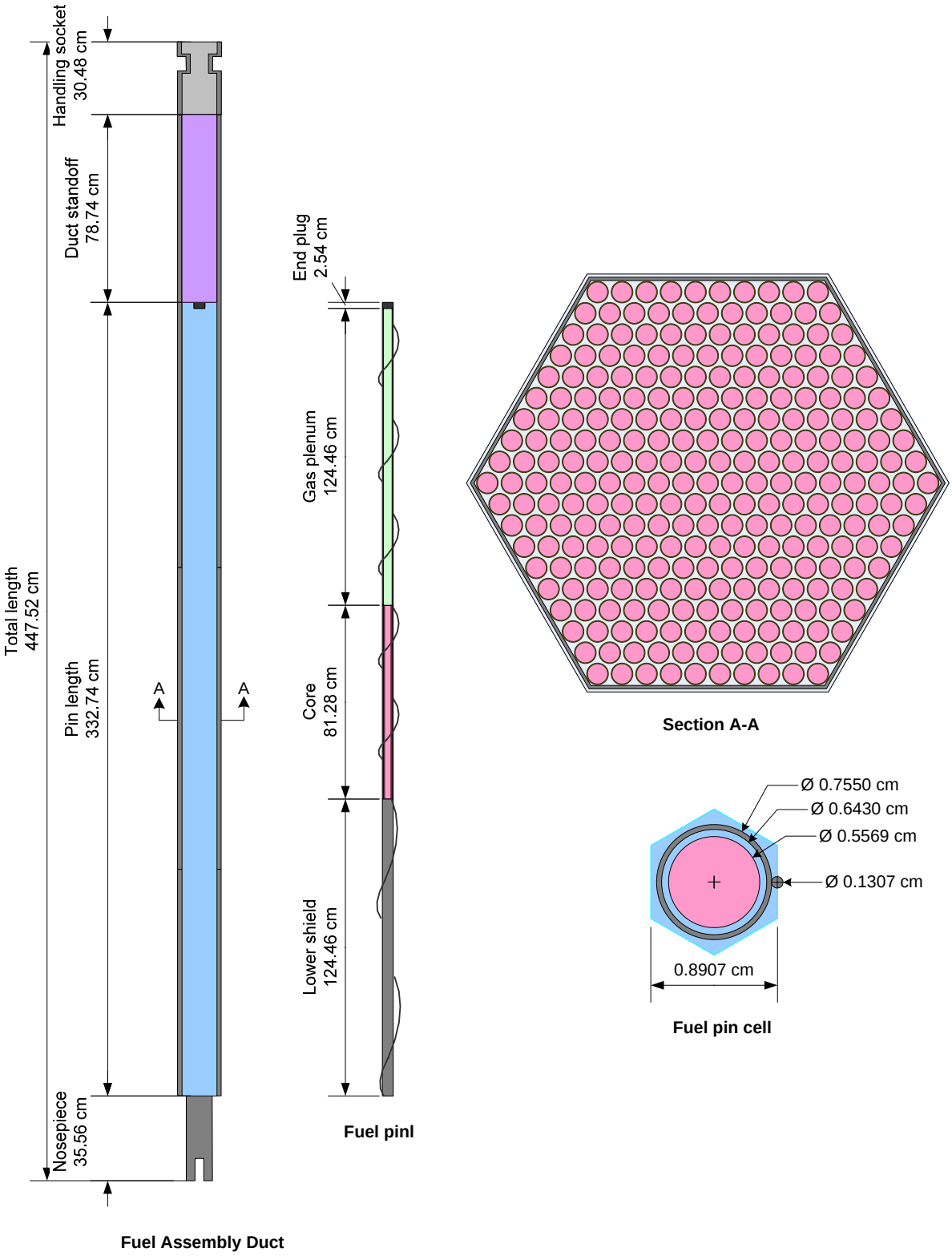

Figure 3. Schematics of Reference Metal Core Fuel Assembly. [10]

Table 7. Assembly Design Parameters of Reference Metal Core Concept. [10]

	Fuel	Reflector	Shield	Control
Assembly data				
- Number of pins	271	91	19	7
- Assembly pitch, cm	16.142	16.142	16.142	16.142
- Inter-assembly gap, cm	0.432	0.432	0.432	0.432
- Duct outside flat-to flat distance, cm	15.710	15.170	15.170	15.170
- Duct material	HT9	HT9	HT9	HT9
- Duct thickness, cm	0.394	0.394	0.394	0.394
- Gap between duct and interior duct, cm	-	-	-	0.400
- Interior duct thickness, cm	-	-	-	0.394
- Interior duct inside flat-to-flat distance, cm	-	-	-	13.334
- Overall duct height, cm	477.52	477.52	477.52	477.52
Pin data				
- Pin material and type	U-TRU-10Zr	HT9-solid pin	B_4C (natural B)	B_4C [a)]
- Bond material	Na	-	He	He
- Overall pin length, cm	332.74	332.74	332.74	86.30
- Pellet smeared density, % TD	75.0	-	81.0	85.0
- Pellet diameter, cm	0.755	1.541	2.553	4.193
- Cladding material	HT9	-	HT9	HT9
- Clad outer diameter, cm	0.557	-	3.337	4.688
- Pin pitch-to-diameter ratio	1.180	1.001	1.001	1.029
- Cladding thickness, cm	0.056	-	0.250	0.070
- Wire wrap diameter, cm	0.131	(no wire-wrap)	(no wire-wrap)	0.133
Volume fraction, %				
- Fuel or Absorber	29.2	-	43.1	42.8
- Bond	9.8	-	10.1	7.6
- Structure	25.7	84.5	29.7	20.8
- Coolant	35.3	15.5	17.1	28.8

a) Natural and 60% enriched boron were used in 4th and 7th row primary control assemblies, respectively.

3.2 COEX with ABR Fuel Cycle and Modeling Approach, and Assumptions

For this study, it was assumed that the COEX separation process is coupled with the ABR fuel cycle (designated COEX/ABR). Figure 4 illustrates the coupling as modeled in REBUS-3. In the COEX process, the LWR spent fuel is separated into three major products: pure uranium, mixture of plutonium and uranium, and others. The mixture is assumed to be 50% plutonium and 50% uranium. The equilibrium cycle calculation path of the REBUS-3 code was used for the startup core calculations. For these calculations, the mixture of plutonium and uranium is used as the more active fuel feed, while the uranium is used as the less active fuel feed without recycling the ABR spent fuel. The more and less active fuel feeds are designated class 1 and class 2 fuels, respectively, in the REBUS-3 code model, and the volume fraction of the class 1 fuel to total fuel is used as a key parameter to determine the equilibrium cycle for a given fuel management condition.

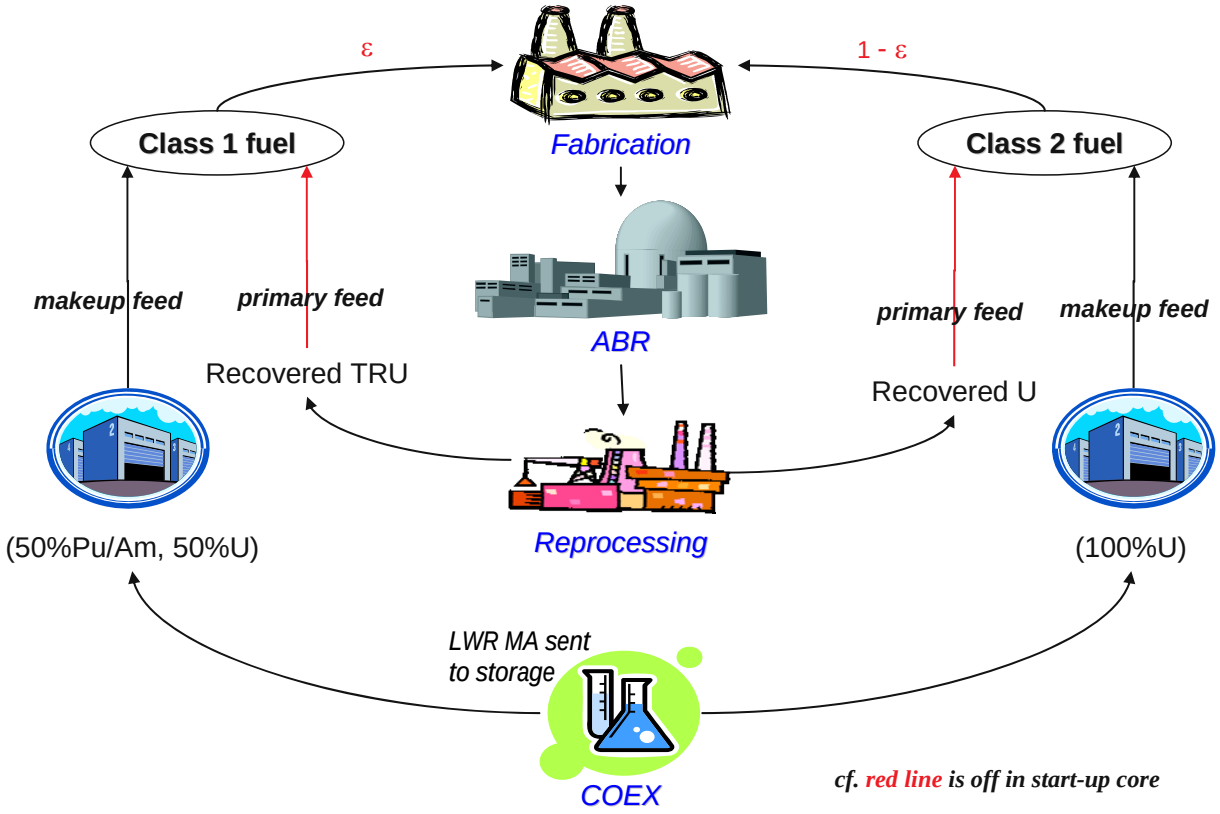

Figure 4. REBUS-3 Model of COEX with ABR Fuel Cycle.

In the equilibrium recycle core calculations, the TRU recovered from the ABR spent fuel is used as the primary class 1 fuel feed and the mixture produced in the COEX process is used as the makeup feed for class 1 fuel; the makeup feed is used when the primary feed is not sufficient to make fresh fuel assemblies. In these calculations, the uranium recovered from the ABR spent fuel is used as the primary class 2 fuel feed and the uranium produced in the COEX process is used as the makeup feed. For the startup core of the reference ABR design, weapons-grade Pu (WG-Pu) was used as the class 1 fuel feed and depleted uranium as the class 2 fuel feed, without recycling the ABR spent fuel. For the equilibrium recycle core of the reference ABR, the TRU recovered from the ABR spent fuel was considered as the primary class 1 fuel feed and the TRU recovered from the LWR spent fuel was considered as the makeup feed. The depleted uranium was also used as the class 2 fuel feed for the equilibrium recycle core. Thus, the uranium feed of the reference ABR core does not contain U-236, while the COEX/ABR fuel cycle contains U-236.

Table 8 contains the TRU vector and WG-Pu and LWR spent fuel used for the reference ABR design and the Pu vector from the COEX process (same as on Table 2). The discharge burnup of the LWR used for spent fuel isotopics is assumed to be 50.0 GWd/t burnup; a 10 year post-irradiation cooling period is assumed.

Table 8. Isotopic Composition (%) of TRU Feeds.

	LWR spent fuel	Weapons grade plutonium	Plutonium recovered in COEX
Np-237	4.79	0.00	0.00
Pu-238	2.21	0.01	3.14
Pu-239	47.71	93.81	56.38
Pu-240	22.90	5.81	26.63
Pu-241	8.42	0.35	7.29
Pu-242	6.80	0.02	5.83
Am-241	5.61	0.00	0.74
Am-242m	0.01	0.00	0.00
Am-243	1.54	0.00	0.00
Fissile	56.14	94.16	63.67

3.3 Core Performance Results

The core performance parameters of the COEX coupled with the ABR (COEX/ABR) fuel cycle have been compared to those of the reference ABR fuel cycle. Since the uranium vector is different for the reference ABR and COEX/ABR fuel cycles, equilibrium cycle analysis of the COEX/ABR fuel cycle was additionally performed using depleted uranium instead of the uranium recovered from the spent fuel to evaluate the impact. A four-batch fuel management scheme was used for both startup and recycle cores with a one-year cycle length and 90% capacity factor. A one-year cooling time and a half-year reprocessing time for ABR spent fuel were assumed in the recycle core calculations.

The main core performance parameters are compared in Table 9. For the reference ABR core design, the required TRU enrichment (TRU mass fraction in total heavy metal mass) of the startup core to achieve the desired cycle length is 15.4%, while it increases to 21.9% for the recycle core because of the reduced fissile content (from WG-Pu in the fuel of the start-up core to TRU recovered from spent fuels as feed in the recycle core). The TRU conversion ratios of the startup and recycle cores are 0.81 and 0.73, respectively, which result in TRU consumption rates of 55.5 kg/year and 81.9 kg/year, respectively. The burnup reactivity loss of the startup core is 3.6%Δk and the peak control assembly fault is about 0.7$. For the recycle core, the burnup reactivity loss is reduced to 2.2%Δk. For both cores, the heavy metal loading and the average discharge burnup are ~3.15 MT and ~93 GWd/t, respectively. In Reference 10, it was confirmed that the reference ABR core has sufficient control and thermal margins.

The core performance of the COEX/ABR fuel cycle is generally the same as that of the reference ABR core design except for the required TRU enrichment and TRU conversion ratio of the startup core. The required TRU enrichment of the startup core increases to 19.5% from 15.4%, and the increased TRU enrichment decreases the TRU conversion ratio to 0.78 from 0.81. The difference is due to the use of the reactor-grade Pu from the COEX process which is less fissile than weapons-grade plutonium. Consistent with the lower conversion ratio, the transuranics consumption rate of the core using reactor-grade plutonium is 19% higher. For the equilibrium recycle core, all core performance parameters are comparable to those of the reference ABR core design.

Table 9. Comparison of Main Core Performance Parameters.

	Reference ABR		COEX/ABR		COEX/ABR with depleted uranium	
	Startup	Recycle	Startup	Recycle	Startup	Recycle
Class 1 fuel feed	WG-Pu	Spent fuel TRU	Pu-U mixture	Spent fuel TRU	Pu-U mixture	Spent fuel TRU
Cycle length, month	12	12	12	12	12	12
Number of batches	4	4	4	4	4	4
Fuel volume fraction, %	29.2	29.2	29.2	29.2	29.2	29.2
Average TRU enrichment, %	15.4	21.9	19.3	21.6	19.7	21.4
Fissile/TRU conversion ratio	0.71 / 0.81	0.84 / 0.73	0.78 / 0.78	0.83 / 0.74	0.80 / 0.76	0.83 / 0.74
HM/TRU loading, MT	13.14 / 2.0	13.15 / 2.9	13.15 / 2.5	13.15 / 2.8	13.15 / 2.6	13.15 / 2.8
Discharge burnup (ave/peak), MWd/kg	93 / 136	93 / 138	93 / 137	93 / 138	93 / 137	93 / 138
Peak fast fluence, $10^{23}/cm^2$	4.07	4.09	4.07	4.03	4.07	4.04
Burnup reactivity loss (%Δk)	3.6	2.2	3.0	2.3	3.0	2.3
Power peaking factor, BOEC/EOEC	1.44 / 1.39	1.47 / 1.42	1.45 / 1.44	1.47 / 1.41	1.45 / 1.41	1.47 / 1.41
Average linear power, kW/m	23.3	23.3	23.3	23.3	23.3	23.3
Peak/limit linear power, kW/m	37.7 / 42.2	37.2 / 43.0	37.3 / 43.0	37.5 / 42.9	37.4 / 42.6	37.4 / 42.9
Core average flux, $10^{15}/cm^2$-sec	3.38	3.34	3.34	3.35	3.36	3.36
Fast flux fraction	0.67	0.68	0.67	0.67	0.67	0.67
TRU consumption rate, kg/year	55.5	81.9	66.1	79.0	71.5	77.8

Finally, the effect of U-236 in the spent uranium feed was evaluated by replacing the spent uranium of the COEX/ABR fuel cycle with depleted uranium (0.2% U-235 content). It was found that the required average TRU enrichment increases by ~ 0.4% for the startup core, but decreases by ~0.2% for the equilibrium recycle core. This resulted in a decrease of 0.02 in the TRU conversion ratio of the startup core (and increase in the TRU consumption rate by 8%); there was virtually no change in the results for the equilibrium recycle core**.** In Table 10, the nuclide masses at charge and discharge states are provided for the COEX/ABR fuel cycles using recycled and depleted uranium, in unit of gram per initial heavy metal metric tonne (g/IHMMT). For both the startup and recycle cores, the discharge Np-237 mass is significantly higher for the case using recycled uranium because of the presence of U-236 in its charge fuel.

Table 10. Comparison of COEX Fuel Compositions (g/IHMMT).

	COEX/ABR (with recycled uranium)				COEX/ABR (with depleted uranium)			
	startup		recycle		startup		recycle	
	charge	discharge	charge	discharge	charge	discharge	charge	discharge
U-234	161.3	231.7	602.6	541.7	0.0	113.4	0.0	86.3
U-235	5655.3	2655.8	1112.9	578.0	1606.7	756.3	1571.4	738.8
U-236	4719.5	4419.8	3162.8	2710.2	0.0	179.4	0.0	179.5
U-238	796210	719965	778818	704081	801739	724381	784147	708584
Np237	0.0	894.9	1578.7	1569.6	0.0	344.9	801.3	792.4
Pu-238	6060.5	3789.5	4691.6	3824.7	6167.2	3720.2	4108.9	3247.3
Pu-239	108955	96766	108731	95659	110873	97955	108701	95830
Pu-240	51456.3	49180.7	67330.8	61051.7	52362.0	50034.5	67005.7	60822.4
Pu-241	14088.4	9256.9	10172.1	9102.4	14336.3	9415.7	10113.5	9067.5
Pu-242	11270.7	10347.7	13098.4	11745.5	11469.0	10524.7	12945.9	11613.7
Am-241	1422.4	2161.8	4288.4	3505.8	1447.4	2191.4	4248.5	3470.8
Am-242m	0.0	108.0	285.6	287.5	0.0	110.2	282.9	284.8
Am-243	0.0	1062.9	3345.4	3345.9	0.0	1091.0	3311.3	3311.7
Cm-242	0.0	120.0	20.9	208.6	0.0	122.8	20.8	207.6
Cm-243	0.0	4.6	15.0	15.6	0.0	4.8	15.0	15.6
Cm-244	0.0	184.1	1961.2	2077.0	0.0	190.8	1946.9	2061.9
Cm-245	0.0	13.9	510.8	510.9	0.0	14.5	507.7	507.8
Cm-246	0.0	0.4	273.6	273.6	0.0	0.4	272.4	272.4
TRU	193254	173892	216304	193178	196655	175721	214282	191506
HM	1000000	901163	1000000	901089	1000000	901151	1000000	901095

3.4 Double-Tier Studies

The studies performed in this work have concentrated on the direct use of co-extraction U-Pu products in PWRs and fast reactors independently. Double-tier fuel cycle strategies have not been evaluated here since some previous studies on this item have been done. [1,6] A recent study considered the coupling of a modified co-extraction process (MCX) to single recycle in PWRs followed by the recovery and use of spent fuel transuranics in fast reactors. [1] The study concluded that the general advantage of the approach is an earlier start to recycling, the potential to use existing or new LWRs as a stop-gap measure, proven fuels technologies and more mature separations technologies, at least initially. The approach reduces the amount of LWR spent fuel in storage more quickly than the reference GNEP scenario, and could allow advanced recycling reactors to be started earlier by initially using U-Pu fuel obtained from LWR used fuel, with minor actinide targets inserted into the reactors once developed and qualified.

The study indicated that a single-pass LWR MOX recycle could reduce the required capacity of fast reactors under equilibrium conditions by 25%. Furthermore, current and advanced LWRs would need to be licensed to use U-Pu MOX fuel, but this is already underway to a certain extent as part of a U.S.–Russia weapons grade plutonium disposition program. Some additional licensing review will be required due to the higher enrichment and different isotopic composition. [1]

It was also noted in the study that the fuel fabrication costs for initial fast reactor cores (or LWR MOX fuel) to perform the initial transmutation could be reduced relative to remote fuel fabrication costs, however costs associated with minor actinide management (additional separations, target fabrication, storage, recycle) need to be taken into account as well to determine if this is a net benefit. [1]

Finally, the earlier study identified as potential disadvantages the significantly reduced repository/waste management benefits if the co-extraction process is used without modifications for minor actinide recovery and/or associated costs for storing minor actinides mixed with fission products pending development of this capability. [1]

4.0 PROLIFERATION RESISTANCE INDICES OF COEX SEPARATION PRODUCTS

The critical masses, and radiation sources and doses of co-extraction products for a given mass of material have been evaluated to obtain indications of the proliferation resistant features of the separation products. Results of these calculations are summarized in the following Sections 4.1 to 4.3.

4.1 Bare-Sphere Critical Masses

A measure of the attractiveness of a nuclear fuel material for weapons utilization is the critical mass of the material. Material quantities that are of the same amount required for weapons-grade plutonium (WG-Pu) provide an indication of potential ease of acquiring the necessary amounts and concealing the material, from the viewpoint of the low critical mass and geometric size required. Conversely, a significantly higher critical mass than for WG-Pu might be indicative of difficulties in acquisition and concealment of the material. Based on these reasoning, the bare (un-reflected) critical masses of Pu containing materials have been calculated for different contents of the element (Pu) in the co-extracted U-Pu products. For comparison, critical masses have also been calculated for U-TRU products.

The calculations were performed with the MCNP code. [12] In the calculation approach, a bare-sphere critical mass that will give a multiplication factor of 1.0 was determined iteratively. It was assumed that the density of the heavy-metal (containing U-Pu) is 19.8 g/cm^3, consistently with previous calculation [1]; a representative material density is used since the intent of the study is to be indicative and not an actual design of nuclear devices. The WG-Pu, reactor-grade plutonium and reactor-grade TRU vectors used in the calculations are provided in Table 11. The reactor spent plutonium vector contained in Table 2 (now without Am in Table 11) was used for cases containing mixtures of uranium and plutonium.

The normalized bare-sphere critical mass results are summarized in Table 12 and are shown graphically in Figure 5. It is noted that the statistical accuracy of the MCNP calculations for these cases is ~60 pcm ($\Delta k/k$). The normalization was performed by dividing the obtained bare sphere critical mass of the material by that for WG-Pu. Previous evaluation had indicated

that the critical mass of a bare sphere of weapons-grade plutonium (WG-Pu) with the assumed density is approximately 10.6 kg; this was confirmed in this study.

Table 11. Initial Pu and TRU Vectors for MCNP Calculations.

Nuclide	WG-Pu Vector (w/o)	Plutonium Vector (w/o)	TRU Vector (w/o)
Pu-238	0.02	3.159	2.758
Pu-239	93.4	56.798	48.813
Pu-240	6.04	26.824	23.056
Pu-241	0.5	7.344	6.949
Pu-242	0.04	5.875	5.050
Np-237			6.663
Am-241			4.669
Am-242m			0.019
Am-243			1.477
Cm-243			0.005
Cm-244			0.498
Cm-245			0.038

Table 12. Relative Bare-Sphere Critical Masses for (Pu,U) and (TRU,U) Materials.*

	Fraction of Pu or TRU in HM	Critical Mass (Relative to WG-Pu)
WG-Pu	1	1
Pu/(Pu+U)	0.1	158.7
	0.2	33.9
	0.3	15.1
	0.4	8.6
	0.5	5.6
	0.75	2.4
	1.0	1.3
TRU/(TRU+U)	0.1	217.8
	0.2	40.4
	0.3	17.7
	0.4	9.9
	0.5	6.3
	0.75	2.7
	1.0	1.4
Enriched U**	20w/o U-235/U	66.2

*Relative to WG-Pu.
** Results derived from Ref. 7.

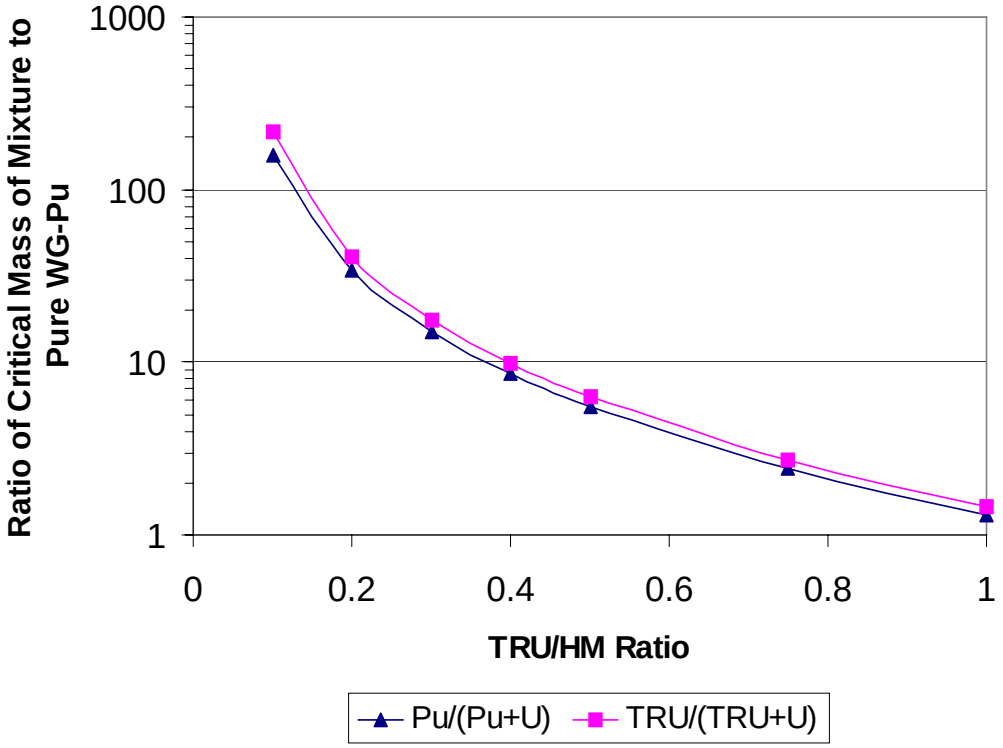

Figure 5. Critical Masses for Pu and TRU Containing Materials.

The results show that all Pu and TRU compositions have larger bare critical masses than WG-Pu, even when they are not mixed with uranium from spent fuel. This is because of the degraded plutonium vector (lower Pu-239 and higher Pu-240 and Pu-242 contents) relative to WG-Pu and the presence of U-238 (a neutron absorber) in the U-Pu mixture cases. Critical masses that are a factor of 1.3 to 220 higher than for WG-Pu are observed. Note that the value for 20 w/o enriched uranium (upper limit of low enriched uranium) is ~66 times higher than that of WG-Pu. It is found that for a plutonium fraction of 1.0 (i.e. as could be used for inert matrix type fuel), the critical mass is ~30% higher than for weapons-grade plutonium. For plutonium fraction of 0.1, similar to that required for LWR MOX fuel, the critical mass is a factor of 159 higher than that of weapons-grade plutonium (greater also than that for low enriched uranium); factor of 2.4 higher than that of low enriched uranium (20% U-235). This critical mass is significantly more than the quantity of heavy metal in a single assembly in the LWR MOX core; it is about three-assembly worth. The critical mass is a factor of 5.6 higher for a fuel with a 50-50 mixture of

Pu-U. These results confirm that fuels containing mixtures of Pu and U will have much larger critical masses than weapons-grade plutonium.

The critical mass is higher in the (TRU,U) fuel compared to the (Pu,U) fuel with the same TRU content, because the Np, Am, and Cm, present in the TRU fuel are generally fertile or less fissile. The difference between the critical masses of the (TRU,U) and (Pu,U) fuels was found to decrease as the transuranics content of the fuel increases; differences of 37% and 8% at TRU contents of 0.1 and 1.0, respectively. This trend is due to the differences in the fissile contents of the two fuels, the impact of which is exaggerated in the lower TRU content range.

4.2 Radioactivity, Decay Heat, and Neutron and Gamma Source Rates

The radioactivity, decay heat, neutron and gamma source rates of mixtures of U-Pu have been generated and compared for transuranics containing materials for the purpose of evaluating the ease of handling these materials in the fuel cycle. The quantities were calculated for 1.056 kg bare sphere of (Pu,U) or (TRU,U) oxide using the ORIGEN2 code [13]. This quantity of material was used, to be consistent with previous evaluations. [14] Similar radiation source data were calculated for WG-Pu metal of the same mass, which were used to normalize the data for the TRU or Pu mixed oxide materials. Figures 6, 7, and 8 provide comparisons of the WG-Pu results to those of Pu (or TRU) fractions of 0.1 to 1.0 in the heavy metal. Tables 13 and 14 shows the leading contributors to the decay heat, neutron source, gamma source, and radioactivity for the WG-Pu metal and (Pu,U) and (TRU,U) oxide cases.

In the tables and figures, the 0.1 case corresponds to the LWR MOX fuel material derived from the COEX process following blending with the pure uranium product, and the 0.5 case corresponds to the COEX process material (50:50 product of the separation). Generally, radiation sources for (Pu,U) oxide and (TRU,U) oxide are much higher than for the WG-Pu metal. In WG-Pu material, Pu-239 (decay heat and gamma source) and Pu-240 (neutron source) and Pu-241 (radioactivity) are the major sources of radiation (see Table 13).

Decay heats calculated for the different cases are summarized in Figure 6. A consequence of an excessively high decay heat is the possibility of malfunctions or failures in a weapon (e.g.

by melting), or reduced weapon yield. Thus, a higher decay heat level makes the material less attractive. The decay heats for the reactor-grade materials are generally higher than for WG-Pu because of the presence of Pu-238 in reactor-grade Pu and additionally Cm-244 and Am-241 in TRU; that for the (Pu,U) oxide case with Pu fraction of 0.1 (Pu-0.1) is lower.

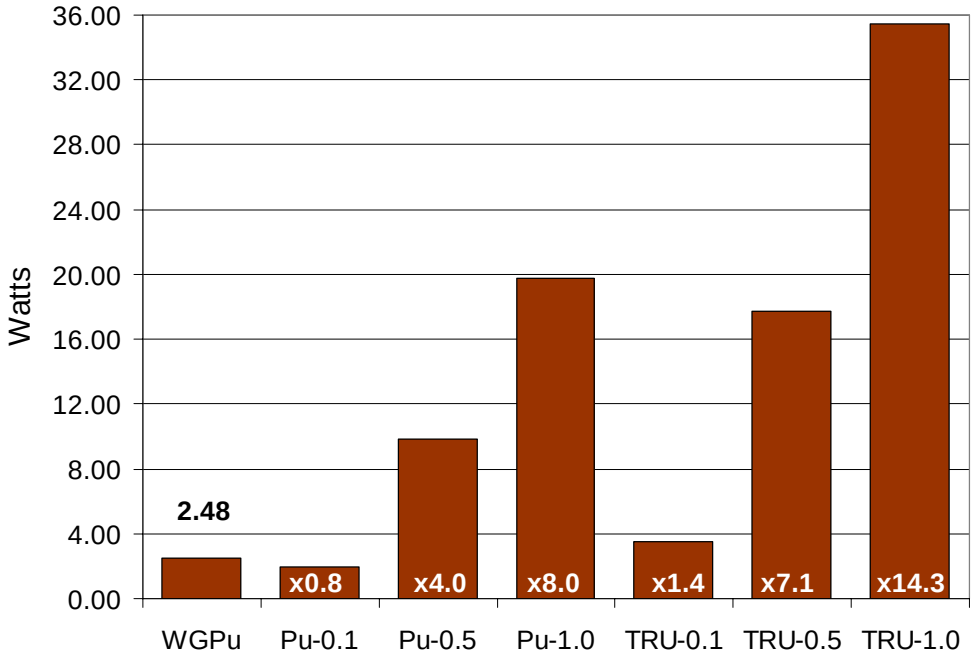

Figure 6. Decay Heats for Spheres Containing U-Pu Mixtures and WG-Pu.

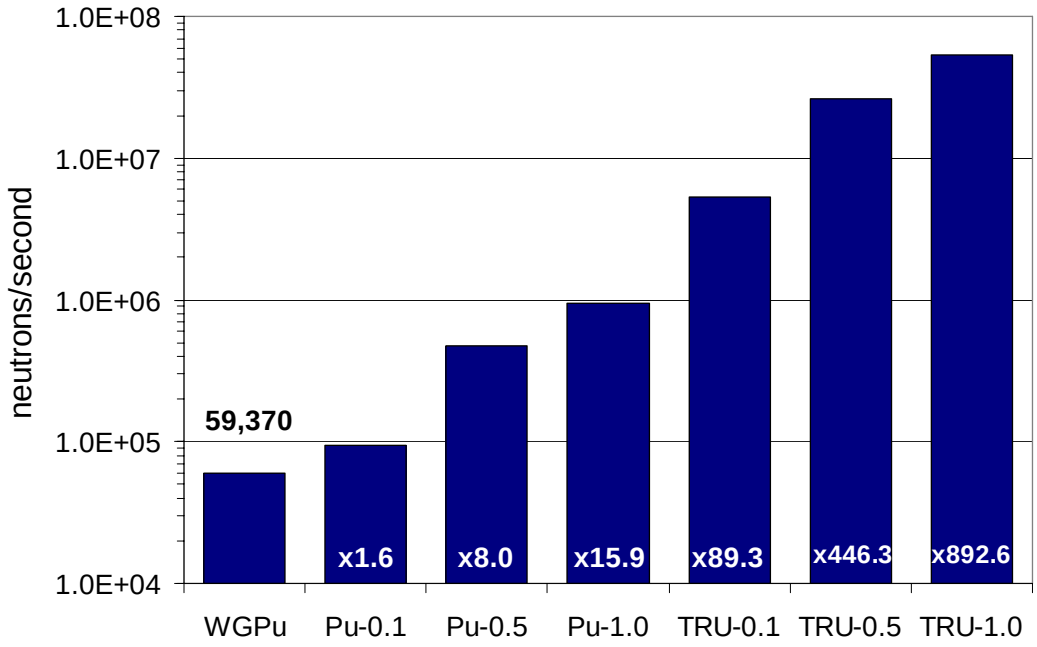

Figure 7. Neutron Source Rates for Spheres Containing U-Pu Mixtures and WG-Pu.

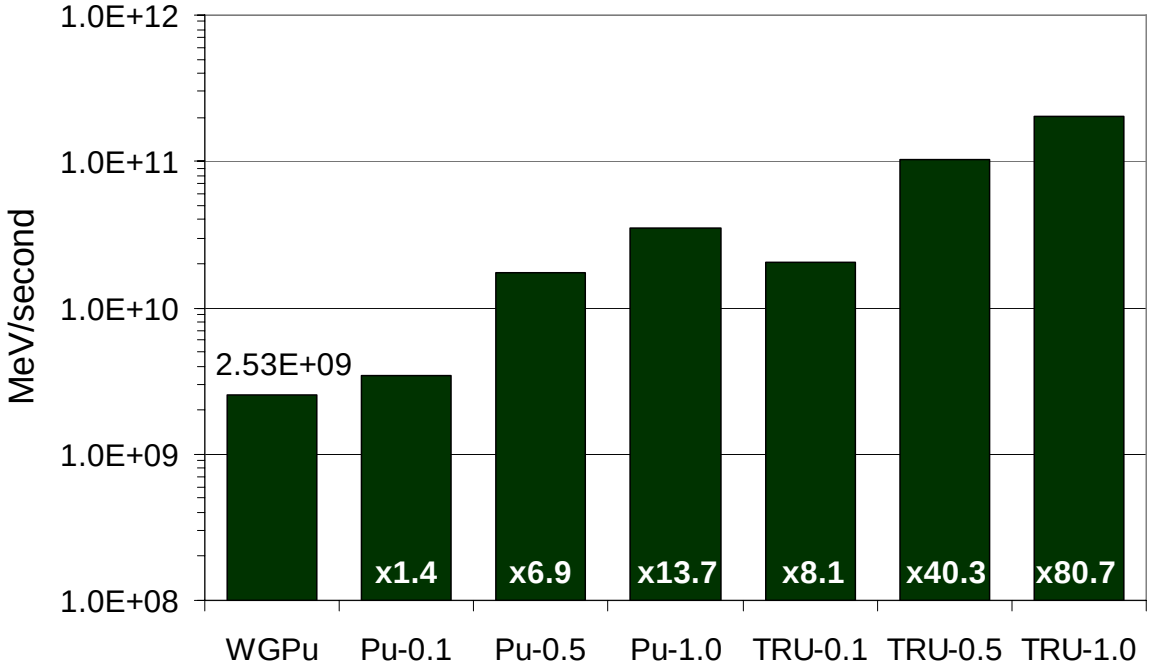

Figure 8. Gamma Source Strengths for Spheres Containing U-Pu Mixtures and WG-Pu.

Table 13. Leading Contributors to Heating and Radiation Sources, (Pu,U) Materials.

Type	WG-Pu metal		Pu/(U+Pu) = 0.1		Pu/(U+Pu) = 0.5		Pu/(U+Pu) = 1.0	
Radio-activity (Curies)	TOTAL	623.70	TOTAL	764.60	TOTAL	3823.00	TOTAL	7645.00
	PU241	544.20	PU241	705.20	PU241	3526.00	PU241	7051.00
	PU239	61.33	PU238	50.40	PU238	252.00	PU238	504.00
	PU240	14.54	PU240	5.70	PU240	28.48	PU240	56.96
	PU238	3.61	PU239	3.29	PU239	16.45	PU239	32.90
	PU242	0.00	PU242	0.02	PU242	0.10	PU242	0.21
Neutron (neutron/ second)	TOTAL	5.94E+4	TOTAL	9.45E+04	TOTAL	4.72E+05	TOTAL	9.45E+05
	PU240	5.81E+4	PU238	5.58E+04	PU238	2.79E+05	PU238	5.58E+05
	PU242	7.11E+2	PU240	2.71E+04	PU240	1.35E+05	PU240	2.71E+05
	PU238	5.61E+2	PU242	9.23E+03	PU242	4.61E+04	PU242	9.23E+04
			PU239	2.40E+03	PU239	1.20E+04	PU239	2.40E+04
Decay Heat (Watts)	TOTAL	2.48	TOTAL	1.97	TOTAL	9.86	TOTAL	19.72
	PU239	1.89	PU238	1.67	PU238	8.35	PU238	16.70
	PU240	0.45	PU240	0.18	PU240	0.89	PU240	1.774
	PU238	0.12	PU239	0.10	PU239	0.51	PU239	1.01
	PU241	0.02	PU241	0.02	PU241	0.11	PU241	0.2186
	PU242	0.0	PU242	0.00	PU242	0.00	PU242	0.006174
Gamma Source (MeV/s)	TOTAL	2.53E+9	TOTAL	3.47E+09	TOTAL	1.74E+10	TOTAL	3.47E+10
	PU239	1.42E+9	PU238	2.98E+09	PU238	1.49E+10	PU238	2.98E+10
	PU240	8.22E+8	PU240	3.22E+08	PU240	1.61E+09	PU240	3.22E+09
	PU238	2.14E+8	PU241	9.91E+07	PU241	4.96E+08	PU241	9.91E+08

Table 14. Leading Contributors to Heating and Radiation Sources, (TRU,U) Materials.

Type	WG-Pu metal		TRU/(U+TRU) = 0.1		TRU/(U+TRU) = 0.5		TRU/(U+TRU) = 1.0	
Radio-activity (Curies)	TOTAL	623.70	TOTAL	772.20	TOTAL	3861.00	TOTAL	7722.00
	PU241	544.20	PU241	667.20	PU241	3336.00	PU241	6672.00
	PU239	61.33	PU238	44.00	PU238	220.00	PU238	440.00
	PU240	14.54	CM244	37.56	CM244	187.80	CM244	375.60
	PU238	3.61	AM241	14.94	AM241	74.67	AM241	149.30
	PU242	0.00	PU240	4.90	PU240	24.48	PU240	48.96
			PU239	2.83	PU239	14.14	PU239	28.28
			AM243	0.27	AM243	1.37	AM243	2.74
			CM243	0.26	CM243	1.19	CM243	2.43
			AM242M	0.18	AM242M	0.87	AM242M	1.72
			PU242	0.02	PU242	0.09	PU242	0.18
Neutron (neutron/ second	TOTAL	5.94E+4	TOTAL	5.30E+06	TOTAL	2.65E+07	TOTAL	5.30E+07
	PU240	5.81E+04	CM244	5.20E+06	CM244	2.60E+07	CM244	5.20E+07
	PU242	7.11E+02	PU238	4.87E+04	PU238	2.43E+05	PU238	4.87E+05
	PU238	5.61E+02	PU240	2.33E+04	PU240	1.16E+05	PU240	2.33E+05
			AM241	1.43E+04	AM241	7.16E+04	AM241	1.43E+05
Decay Heat	TOTAL	2.48	TOTAL	3.55	TOTAL	17.73	TOTAL	35.47
	PU239	1.89	PU238	1.46	PU238	7.29	PU238	14.58

(Watts)	PU240	0.45	CM244	1.31	CM244	6.57	CM244	13.14
	PU238	0.12	AM241	0.50	AM241	2.48	AM241	4.96
	PU241	0.02	PU240	0.15	PU240	0.76	PU240	1.53
	PU242	0.00	PU239	0.09	PU239	0.44	PU239	0.87
			PU241	0.02	PU241	0.10	PU241	0.21
			CM243	0.01	AM243	0.04	CM243	0.09
			AM243	0.01	CM243	0.04	AM243	0.09
			PU242	0.00	PU242	0.00	PU242	0.01
			CM245	0.00	CM245	0.00	CM245	0.00
Gamma Source (MeV/s)	TOTAL	2.53E+9	TOTAL	2.04E+10	TOTAL	1.02E+11	TOTAL	2.04E+11
	PU239	1.42E+9	AM241	1.36E+10	AM241	6.78E+10	AM241	1.36E+11
	PU240	8.22E+8	PU238	2.60E+09	PU238	1.30E+10	PU238	2.60E+10
	PU238	2.14E+8	CM244	2.12E+09	CM244	1.06E+10	CM244	2.12E+10

The neutron source rates given for the (Pu,U) and (TRU,U) oxide cases are the sum of contributions from spontaneous fission neutrons and (α,n) neutron production reactions. Spontaneous neutron data only are presented for the WG-Pu metal. For the (Pu,U) fuels, the ratio of (α,n) neutrons to spontaneous fission neutrons is ~1.37. However, in the (TRU,U) fuels the spontaneous neutron rates overshadow the contribution from (α,n) reactions, which accounts for ~2 percent of the total neutron source strength. Figure 7 quantifies the neutron source rate of the oxide materials compared to WG-Pu metal. The neutron source rate affects the level of intrinsic protection in the fuel, both by constituting a radiation safety hazard at high levels and by limiting the amount of material that could be used in a weapon. A high neutron source rate provides a criticality hazard if too much fissile material is collected, as in the fabrication of a weapon, and makes a less effective weapon as there is a risk of early detonation from the neutron source. As seen in Figure 7, all of the various (Pu,U) or (TRU,U) oxides have larger neutron emission rates than WG-Pu, indicating a higher intrinsic proliferation barrier for these materials. The neutron sources for the reactor-grade materials are higher due to the presence of Pu-238, Pu-240, and Pu-242 in the Pu cases, and are even much higher due to Cm-244 in the TRU cases.

Tables 13 and 14 show that the radioactivity values (Curies) for the (Pu,U) and (TRU,U) cases are higher than those of the WG-Pu case. The primary contributor to radioactivity for all the cases is the Pu-241. The radioactivity values of the (TRU,U) and (Pu,U) materials are quite similar, though the (TRU,U) values are generally slightly higher.

Similarly to the radioactivity, decay heat and neutron source results, the gamma source rate (MeV/s) is higher for the (Pu,U) or (TRU,U) oxide relative to the WG-Pu material (see

Figure 8 and Tables 13 and 14). The gamma source is higher due to the presence of Pu-238 (predominantly) and Pu-240 in the Pu cases and Am-241 (predominantly) and Cm-244 for the TRU cases. The gammas from Am-241 have higher energies than those from Pu-238 and Pu-239; Pu-238 has the lowest energy gammas of the three isotopes.

Sensitivity Study on U-232 Content

Following the calculations presented above, it was found in Ref. 15 that the presence of U-232 in recovered uranium fuel is an important item for consideration when determining the radiation sources for uranium-containing materials. Because of the presence of plutonium and minor actinides in material mixtures being considered in this study, it was assumed that the radiation sources arising from U-232 would be overwhelmed by those of these other sources, and so could be neglected. To confirm this assumption, an evaluation of this issue was performed. A few sensitivity calculations with trace amounts of U-232 and daughter products including Th-228, Ra-224, Rn-220, Po-216, Bi-212, Pb-212 and Tl-208 were done with the ORIGEN2 code.

Typically, trace amount of U-232 is present in spent fuel uranium due to the α-decay of Pu-236 (dominant route), the electron capture process in Np-232 and β- decay in Pa-232. The content of U-232 is about 0.5 to 5E-09 gram per gram of total spent fuel uranium. Uranium-232 has a half-life of 68.9 years and decays through a series of much short-lived daughter products. It has three daughters that emit strong beta and gamma radiation: Tl-208, Bi-212, and Pb-212 (average of 3.4 MeV per disintegration) [15]. However, for the 1.056 kg sample used in the study, the total mass of U-232 and daughters is ~1.2E-06 grams. The source strength of the gamma, alpha, and neutron radiation, calculated with ORIGEN2, were compared with and without the trace amounts of U-232 and its daughters at 10-year post-irradiation. The 10-year cooling period was chosen as the peak of the activity of the U-232 and daughters between 5 and 30 years of cooling time [15]. The relative gamma source strength calculated with ORIGEN2 was ~0.1 to 1% higher when U-232 and its daughter products are included in the calculation. Dose calculations with MCNP have not been done due to the fact that the ENDF/B-V and ENDF/B-VI cross section libraries available to this work do not contain data for Tl-208, Bi-212, Pb-212, and Po-216, and because their contributions are expected to be small.

It is noted however that the issue of radiation sources from U-232 and daughters is quite important for *enriched uranium* fuel derived from spent fuel uranium. This is due to the negligible radiation sources from U-238 and U-235, and the propensity to preferentially accumulate U-232 in the enrichment process due to its lower mass than U-235.

4.3 Gamma Dose Rates

Contact gamma dose rates for bare and steel-encased spheres of Pu- and TRU-containing oxide materials have also been calculated using MCNP in order to investigate the intrinsic radiation barrier of such materials. The gamma sources for the MCNP cases were obtained from ORIGEN2 calculations. The reference sphere used for these calculations has a mass of 1.056 kg and is assumed to contain oxide mixture of plutonium and uranium from the COEX process with a density of 10 g/cm^3; taken from Appendix F of Reference 14. Similar assumptions were made for the TRU cases. This mass and geometry were selected in order to be consistent with the earlier studies investigating the impact of reprocessing and handling neptunium-containing MOX fuels, providing a basis for comparison. This mass corresponds to a radius of 2.93 cm, which was used for all the MCNP gamma dose calculations. A WG-Pu metal sphere was also calculated, as WG-Pu would most likely be found in the metallic, rather than oxide form.

In addition to bare sphere calculations performed for 1.0 (pure), 0.5 and 0.1 Pu/(U + Pu) or TRU/(U + TRU) ratios, a layer of steel to represent a container was included in some calculations, with thicknesses ranging from 0 to 1.0 cm. The Cr11Ni16Mo steel with a density of 7.971 g/cm^3 was assumed for the container. The ANSI/ANS-6.1.1-1977 photon flux-to-dose rate conversion factors were employed for converting the contact photon flux to contact dose rate [12]. No post-fuel processing decay time was assumed: therefore, no americium is present in the Pu vector. Results for these cases are summarized in Table 15. They are also displayed graphically in Figures 9 to 11, where the values have been normalized to those of WG-Pu metal.

Table 15. Contact Gamma Dose Rates for COEX-Pu and TRU Spheres.

Material (Oxides)	TRU Fraction	Steel Thickness (cm)	Dose Rate (Rem/hr)
Pu/(Pu+U)	0.1	0.0	2.74 ± 3.00 %
Pu/(Pu+U)	0.5	0.0	13.00 ± 3.11 %

Pu/(Pu+U)	1.0	0.0	24.38 ± 3.22%
Pu/(Pu+U)	0.1	0.5	0.0107 ± 13.37 %
Pu/(Pu+U)	0.5	0.5	0.0512 ± 13.65 %
Pu/(Pu+U)	1.0	0.5	0.0959 ± 14.03 %
Pu/(Pu+U)	0.1	1.0	0.0059 ± 15.65 %
Pu/(Pu+U)	0.5	1.0	0.0277 ± 16.17 %
Pu/(Pu+U)	1.0	1.0	0.0505 ± 16.83 %
TRU/(TRU+U)	0.1	0.0	10.72 ± 4.04 %
TRU/(TRU+U)	0.5	0.0	51.05 ± 1.95 %
TRU/(TRU+U)	1.0	0.0	99.82 ± 1.93 %
TRU/(TRU+U)	0.1	0.5	0.53 ± 5.62 %
TRU/(TRU+U)	0.5	0.5	2.72 ± 2.97 %
TRU/(TRU+U)	1.0	0.5	5.48 ± 2.94 %
TRU/(TRU+U)	0.1	1.0	0.27 ± 7.92 %
TRU/(TRU+U)	0.5	1.0	1.33 ± 3.72 %
TRU/(TRU+U)	1.0	1.0	2.66 ± 3.65 %
		0.0	1.76 ± 3.87 %
100% WG-Pu Metal	1.0	0.5	0.068 ± 4.69 %
		1.0	0.036 ± 6.03 %

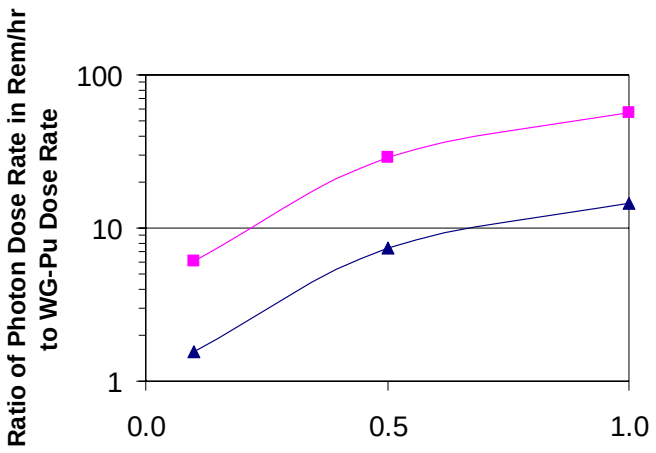

Figure 9. Gamma Dose Rate for Bare Fuel Spheres.

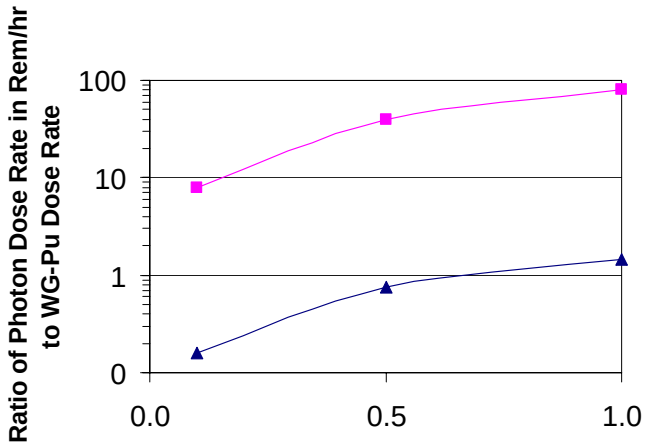

Figure 10. Gamma Dose Rate for Fuel Spheres with 0.5 cm Steel Casing.

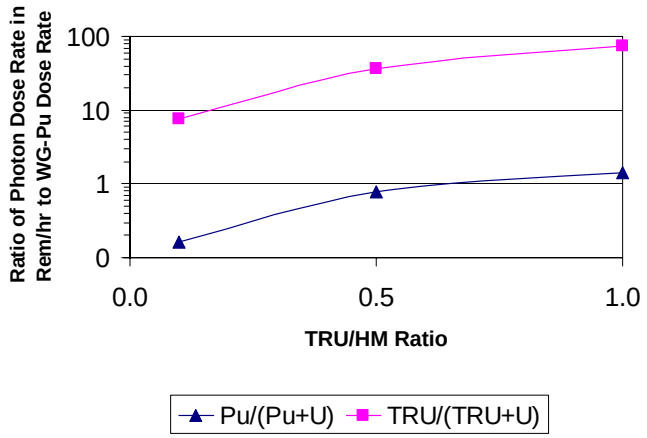

Figure 11. Gamma Dose Rate for Fuel Spheres with 1.0 cm Steel Casing.

It is noted that 20 million particle histories were used for the MCNP calculations, and the relative tally error (standard deviation divided by the mean) exceeded 15% for some of the lowest dose situations, but overall provided reasonably good results. The WG-Pu calculations have improved accuracy over the reactor-grade calculations for a given number of particle histories, as far more particles (60% more) make it into the steel reflector region in the WG-Pu case. This is because the average mean-free-path of the photons in WG-Pu is 0.0431 cm versus 0.0147 cm in 100% reactor-grade Pu, and 1.01 versus 0.91 cm in their steel encasing, respectively. This leads to more particles being measured at the outer surface by MCNP and thus better statistical accuracy.

The contact doses are lower than the spent fuel standard (100 Rem/hr at 1 m). Regarding the capabilities of these fuel types to provide an intrinsic barrier to proliferation, the TRU cases provide the most photon (gamma) dose in both the bare and steel-enclosed spheres. For a 50/50 mix of plutonium and uranium in a bare-sphere oxide material, the contact dose rate was found to be a factor of ~7 higher than that of a bare-sphere WG-Pu; with a 50/50 mix of transuranics and uranium, a factor of ~29 higher is obtained. *For a 10/90 mix of plutonium and uranium, as in MOX fuel derived from COEX separation product, the contact dose rate is slightly higher (by factor of 1.6) than for WG-Pu.* The steel-encased spheres have dose values orders of magnitude lower for both compositions [(Pu,U) and (TRU,U) oxides], but are many times higher for the TRU fuel than for the Pu-only fuels, largely due to the presence of Am-241 and Cm-244.

It is also observed that steel casing shields the lower-energy photons from reactor Pu more effectively than TRU or WG-Pu photons. As example, w*ith a steel casing of 0.5 cm thickness, the gamma dose for the 10/90 Pu/U fuel material is a factor of 6 lower than that of WG-Pu*. Furthermore, the gamma dose rate ratio for the TRU case compared to Pu case is much higher when materials are in steel casing; the TRU dose is ~50 times higher than for Pu with steel casing, and ~4 times higher than for Pu bare. This is because there are higher energy photons in TRU and shielding by steel is relatively less effective for these photons than for Pu photons. The much larger dose rate from the TRU fuel is a result of including the higher actinides in the fuel, particularly the Am and Cm. This provides some inherent proliferation resistance by requiring special procedures and/or equipment to handle quantities of the material.

5.0 CONCLUSIONS

Evaluations of the use of the COEX^{TM} separation process in the PWR and fast reactor fuel cycle have been performed. The study focused on the impact of utilizing reactor spent uranium and plutonium on the core performance of both systems and also on indices (physics quantities) that could provide indication of the relative intrinsic proliferation barrier of the separated materials. The study also took as given the notion that U-Pu co-extraction is potentially proliferation resistant, neglecting the ease of separation of the plutonium (or TRU) from the uranium. Thus, it is a detailed study of physics issues, rather than an in-depth study of potential consequences of the co-extraction process.

The use of spent uranium instead of natural uranium in a PWR core was found to require higher plutonium content in the loaded MOX fuel in order to meet the reactivity requirement for a given fuel burnup level. This higher plutonium content and the presence of U-236 in the spent uranium fuel result in over 13% higher TRU in the discharged fuel per initial heavy-metal. Additionally, it was observed that significantly more Np-237 (factor of 5 more) is produced in the spent fuel of a core using spent uranium based fuel than that using natural uranium based fuel, primarily because of the presence of U-236. Furthermore, less Pu-238 is consumed and more Am-241 is produced due to the use of spent uranium in the LWR MOX fuel cycle. These higher quantities of Np-237, Pu-238 and Am-241 (and of Pu and TRU) in the waste would decrease the effective repository utilization, relative to the use of natural uranium in the PWR MOX core; the Pu-238 and Am-241 masses at discharge are 20% and 14% higher, respectively, for the PWR MOX core using spent uranium. Regarding the overall fuel cycle, since the minor actinides would however continue to accumulate, a solution must therefore be found for removing them from the waste stream if significant gains are to be made for effective repository capacity. Consumption in fast reactors and the use of minor actinides containing targets in advanced reactors (thermal and fast) are potential approaches for minimizing the minor actinides passing to the repository.

Evaluation of the use of LWR SNF plutonium in the fuel for a 1,000 MWt ABR startup core instead of the weapons-grade plutonium currently proposed indicated an increase in the required TRU content by ~4%. This is due to the higher fissile content of the fuel using

weapons-grade plutonium. The implication is that the core conversion ratio (with reactor-grade plutonium) is lower and the TRU consumption rate is higher by 19%. No appreciable differences are found between the core performance parameters of recycle cores for the cases. The use of depleted uranium versus reactor spent uranium was found to result in a TRU content difference less than ~0.4%.

Critical masses, decay heat, neutron and gamma source rates and gamma doses were calculated using a spherical configuration of the materials (1 kg amount) to provide indications of the intrinsic proliferation barrier of the co-extraction products. Values for (Pu,U) and (TRU,U) and materials were compared to those for weapons-grade plutonium. It was found that the critical masses of (TRU,U) and (Pu,U) products are higher than that of weapons-grade plutonium. For the 10/90 mix of Pu/U, the critical mass was found to be a factor of 2.4 larger than that of low enriched uranium (20% U-235). Furthermore, the critical mass for (TRU,U) fuel is higher compared to that for (Pu,U) fuel with the same TRU content, because the Np, Am, and Cm, present in the TRU fuel are generally fertile or less fissile. The difference between the critical masses of the (TRU,U) and (Pu,U) fuels was found to decrease as the TRU (or Pu) content of the fuel increases; differences of 37% and 8% at TRU (or Pu) contents of 0.1 and 1.0, respectively. This trend is due to the differences in the fissile contents of the two fuels, the impact of which is exaggerated in the lower TRU content range.

Generally, heating and radiation sources for (Pu,U) oxide and (TRU,U) oxide are much higher than for the WG-Pu metal. In WG-Pu material, Pu-239 (mostly), Pu-241 (radioactivity) and Pu-240 (for neutrons) are the major sources. For the Pu and TRU cases, the primary sources are Pu-238 (in Pu) and Am-241 and Cm-244 (additionally in TRU); Pu-240 and Pu-242 are also important for the neutron source and Pu-241 for radioactivity in the materials.

Contact gamma dose rates for bare and steel-encased spheres of plutonium- and transuranics-containing oxide materials (1 kg) were also evaluated. They were found to be less than that of the spent fuel standard (100 Rem/hr at 1 m). For a 50/50 mix of plutonium and uranium in a bare-sphere oxide fuel, the contact dose rate was found to be a factor of ~7 higher than that of a bare-sphere weapons-grade plutonium; with a 50/50 mix of Transuranics and uranium, a factor of ~29 higher is obtained. The use of a steel container reduces by many orders

of magnitude the photon (gamma) dose and shields the low energy photons from reactor Pu more effectively than TRU or WG-Pu photons. *With a steel casing of 0.5 cm thickness, the gamma dose for the 10/90 Pu/U fuel material is a factor of 6 lower than that of shielded WG-Pu.* Furthermore, the dose rate ratio for the TRU case compared to Pu case is much higher when materials are in steel casing; the TRU dose is ~50 times higher than for Pu with steel casing, and ~4 times higher than for Pu bare. This is because there are higher *energy* photons in TRU and shielding by steel is relatively less effective for these photons than for Pu photons.

These critical mass, radiation source and dose, and decay heat indicators suggest that the transuranics-uranium mixtures (products) derived from the co-extraction process would require special/additional handling in the fuel cycle over that for weapons-grade plutonium.

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