AN ASSESSMENT OF THE PROLIFERATION RESISTANCE OF MATERIALS IN ADVANCED NUCLEAR FUEL CYCLES

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ABSTRACT

This paper summarizes a study, performed at the request of the United States Department of Energy (DOE), of materials mixtures containing special nuclear materials (SNM) produced in a number of advanced nuclear fuel cycles (NFCs). The primary focus of this study was the SNM-containing final product or mixtures produced during intermediate steps of reprocessing. The list of reprocessing schemes considered in the present study includes: PUREX, UREX+, and COEX.

For the SNM in these schemes, we have calculated "attractiveness levels," couched in terms chosen for consistency with those normally used for nuclear materials in DOE nuclear facilities.[1] Key findings include co-extracting Np with Pu does not alter the Attractiveness Level of the recycled material; transuranics become more attractive with age; dilution of SNM with spent or depleted uranium can lower the Attractiveness Level of SNM by a full letter grade; and dilution of Pu with significant quantities of ^{238}Pu can also lower the Attractiveness Level of Pu by a full letter grade.

Key Words: Reprocessing, Attractiveness Level, Proliferation Resistance

INTRODUCTION

The United States Department of Energy (DOE) requested a comparison of the Attractiveness Levels[1] of the special nuclear materials (SNM) associated with the COEX and UREX+1A reprocessing schemes. The Attractiveness Levels along with brief descriptions are reproduced in Table 1. The DOE requires more stringent material control and accountability for higher Attractiveness Levels. Only Attractiveness Levels B through E are applicable to SNM associated with reprocessing schemes.

	Attractiveness Level	FOM
WEAPONS Assembled weapons and test devices	A	
PURE PRODUCTS Pits, major components, button ingots, recastable metal, directly convertible materials	B	> 2
HIGH-GRADE MATERIALS Carbides, oxides, nitrates, solutions (≥25 g/L) etc.; fuel elements and assemblies; alloys and mixtures; UF4 or UF6 (≥ 50% enriched)	C	1-2
LOW-GRADE MATERIALS Solutions (1 to 25 g/L), process residues requiring extensive reprocessing; moderately irradiated material; Pu-238 (except waste); UF4 or UF6 (≥20%<50% enriched)	D	0-1
ALL OTHER MATERIALS Highly irradiated forms, solutions (<1 g/L), uranium containing <20% U-235 or <10% U-233 (any form, any quantity)	E	< 0

Table 1: Mapping of FOM into Attractiveness Levels[1] of DOE Nuclear Materials (*i.e.*, Depleted, Enriched, and Normal Uranium; ^{233}U; ^{238}Pu; ^{239}Pu; ^{240}Pu; ^{241}Pu; ^{242}Pu; ^{241}Am; ^{243}Am; Bk; ^{252}Cf; Cm; ^{2}H; Enriched Lithium; ^{237}Np; Th; ^{3}H; and Uranium in Cascades). The FOM column was added for the purposes of this study, and is not part of the regulatory process. Additionally, even if the FOM is calculated to be less than zero, the material cannot be assigned an Attractiveness Level of E unless the material is composed of a single element.

APPROACH

The dominant source of material for reprocessing is spent uranium oxide from pressurized water reactors (PWRs) and boiling water reactors (BWRs). Such spent fuel is typically characterized by its burnup, expressed in MW·d/kg of initial heavy metal. The average burnup of spent fuel in the USA historically has ranged from ~15 MW·d/kg for BWRs and from ~25 MW·d/kg for PWRs[2] to present day values of 45 – 50 MW·d/kg. The isotopic composition of spent fuel was generated with ORIGEN2.2[3] for burnups ranging from 7.5 to 90 MW·d/kg for the purposes of this analysis. Also varied was the spent fuel age at the time of reprocessing relative to discharge. The decay of any reprocessed material after reprocessing was not considered.

The Attractiveness Levels are calculated by first evaluating the following figure of merit (FOM):

$$FOM = 1 - \log_{10}(x),$$

where x is given by:

$$x = M\left[\frac{1}{800} + \frac{h}{4500}\right] + \left[\frac{D}{500}\right]^{\frac{1}{\log_{10} 2}},$$

and M is the bare critical mass of the metal in kg, h is the heat content in W/kg, and D is the dose rate of $0.2 \cdot M$ evaluated at 1 m from the surface in rem/h. The calculation is completed by mapping the FOM value into an Attractiveness Level according to Table 1. Note that the FOM is only applicable to metals.

The primary end product of COEX is Pu that has been diluted with uranium. The UREX+ options include the extraction of U, Pu, Am, Cm, and TRU (*i.e.*, the transuranics) as well as the co-extraction of U, Pu, and Np; of Am and Cm; and of U and TRU. This study considered only Pu, Pu + Np, and TRU end products diluted with varying concentration of U and Pu end product diluted with varying concentration of ^{238}Pu.

RESULTS

The ratio of the Pu + Np FOM to the Pu FOM is shown in Figure 1 for metals with a range of burnups, fuel ages, and uranium concentrations (The uranium diluent used herein is the spent uranium, but using depleted uranium yields the same results). Over a wide range of parameters, the ratio varies by less than 7 %. If one further restricts oneself to present day burnups and possible ages, then the variation in this ratio is less than 3 %. Note that UREX+2, UREX+3, and UREX+4 schemes co-extract Pu and Np.

The ratio of the FOM at an arbitrary age to the FOM at an age of 0.1 years is shown in Figure 2 for TRU metal. This ratio monotonically increases for all burnups for ages ranging from 0.1 to 3 years (Most reprocessing facilities will not take fuel that is younger than 2 or 3 years to reduce the radiation from the fission products in the feedstock spent fuel). For burnups ≥ 45 MW·d/kg, this ratio is even higher at 100 years. For all burnups, this ratio is greater than unity for all ages > 0.1 years. For the historical range of burnups and ages (15 MW·d/kg and 35+ years to 50 MW·d/kg and 2 years), this ratio is ~1.5.

Developers of reprocessing schemes desire their intermediate and end products be classified at the lowest possible Attractiveness Level. Developers have limited design options for reprocessing their spent fuel: element(s) (co-)extracted, diluent (*e.g.*, U or fission products), and amount of diluent. To aid developers achieve their goal, the design space for reprocessing Pu as a metal is displayed in Figure 3. The age of the Pu determines which of the line types (solid lines or long, medium or short dashed lines) are applicable in Figure 3. The Attractiveness Level of the Pu is determined by the relationship of its age and, if co-extracted with U, the U content to an Attractiveness Level boundary. For example, pure Pu metal obtained from spent fuel burned for ≤ 58 MW·d/kg has an Attractiveness Level of B. Diluting Pu metal with 60-70 % U degrades the Attractiveness Level to C. Further dilution of ≥ 84 % degrades the Attractiveness Level to D. Note the U content of spent fuel at burnups of 30, 60, and 90 MW·d/kg is 96, 93, and 89 %, respectively.

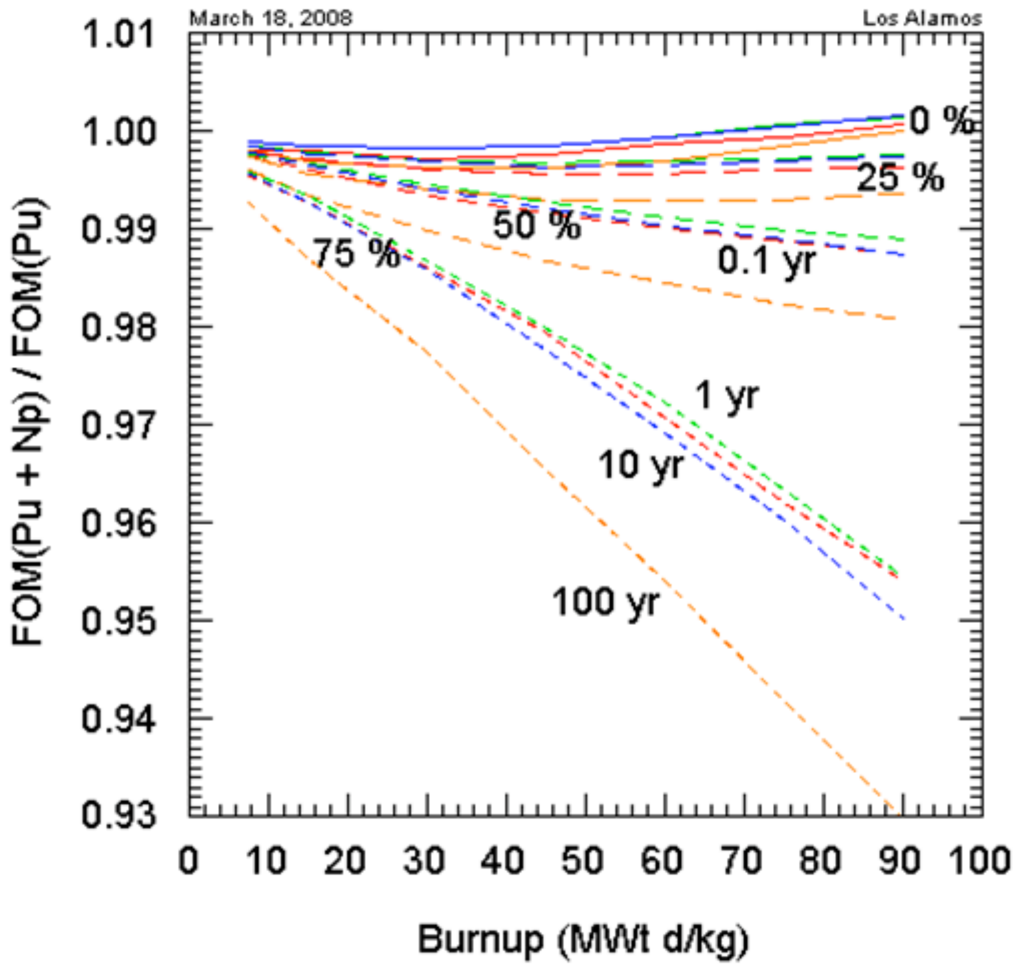

Figure 1: The ratio of the FOM of Pu + Np to the FOM of Pu plotted as a function of burnup. The red, green, blue, and orange lines denote a material age of 0.1, 1, 10, and 100 years, respectively. The solid line and the long, medium, and short dashed lines indicate a uranium content of 0, 25, 50, and 75 %.

The U content of the COEX end product is also displayed in Figure 3. For a burnup of 45 MW·d/kg, the COEX end product would be classified at an Attractiveness Level C. Note that Figure 3 is also applicable to the PUREX reprocessing scheme and the UREX+2, UREX+3, and UREX+4 schemes.

The design space for TRU is shown in Figure 3. The most obvious result from Figure 3 is that TRU has a significantly lower Attractiveness Level than Pu. Additionally, the Attractiveness Level of TRU is sensitive to its age, as was previously shown in Figure 2, whereas the Attractiveness Level of Pu is not significantly sensitive to material age.

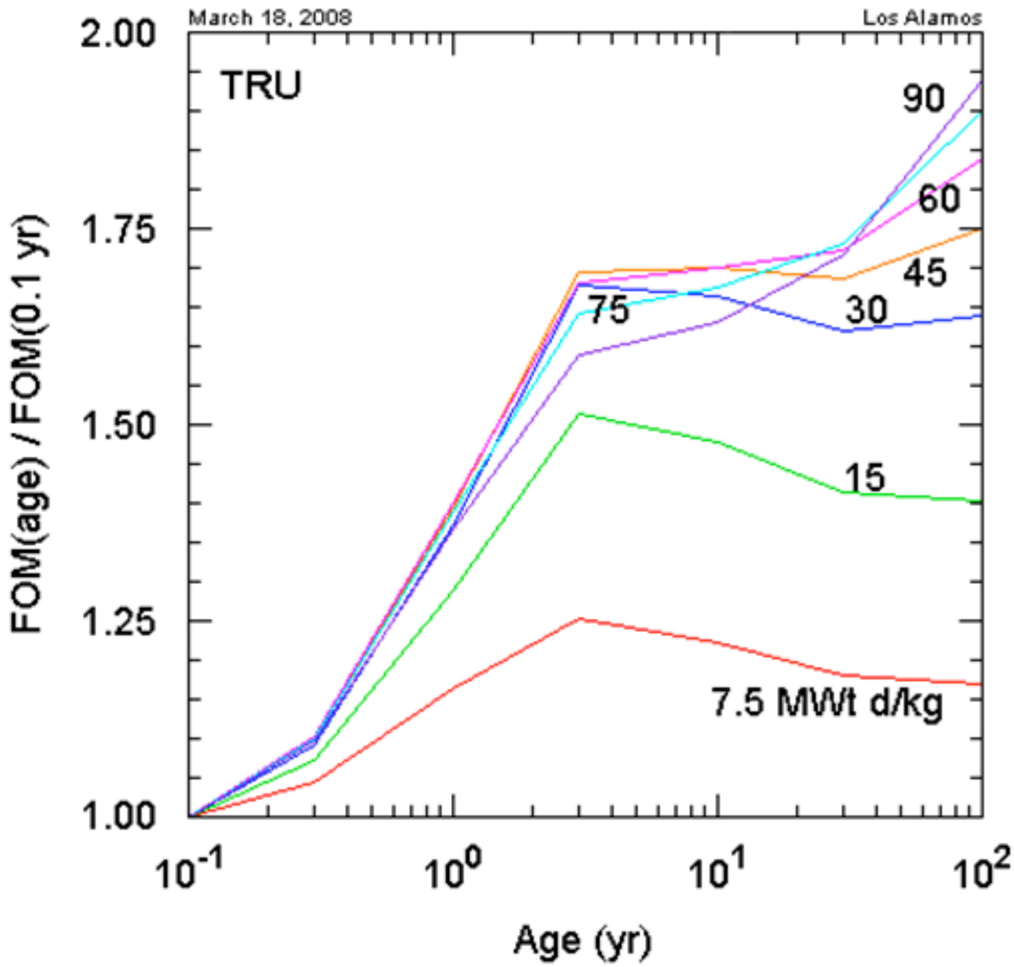

Figure 2: The ratio of the FOM at an arbitrary age to the FOM with an age of 0.1 years plotted as a function of the arbitrary age. The red, green, blue, orange, magenta, cyan, and purple lines denote burnups of 7.5, 15, 30, 45, 60, 75, and 90 MW·d/kg, respectively.

Diluting Pu with ^{238}Pu has been proposed.[4] The design space for Pu using ^{238}Pu dilution is shown in Figure 4. For burnups ≤ 55 MW·d/kg, reprocessed Pu from spent fuel has an Attractiveness Level of B. To achieve an Attractiveness Level of C requires either a burnup of > 55 MW·d/kg for fuel ≤ 10 years old or dilution at 4-6 % ^{238}Pu content, depending on the burnup. An Attractiveness Level of D requires 70-80 % ^{238}Pu content.

CONCLUSIONS

Plutonium and neptunium have the same Attractiveness Level. Co-extracting neptunium with plutonium will not reduce the Attractiveness Level of the reprocessed end product. Extracting just the plutonium puts highly attractive neptunium into the reprocessing waste stream. The Attractiveness Levels of Pu and Pu + Np are sensitive to the burnup of the spent fuel, but are not sensitive to the age of the spent fuel for ages < 30 years. For ages ≥ 30 years, the Attractiveness Level of Pu and Pu + Np increases slightly with age. For a burnup of 45 MW·d/kg, a dilution of Pu or Pu + Np metal with ≥ 20 % spent or depleted uranium is required for an Attractiveness Level of C, and a dilution of > 82 % uranium is required for an Attractiveness Level of D. These conclusions regarding plutonium are applicable to the following reprocessing schemes: COEX, PUREX, UREX+2, UREX+3, and UREX+4.

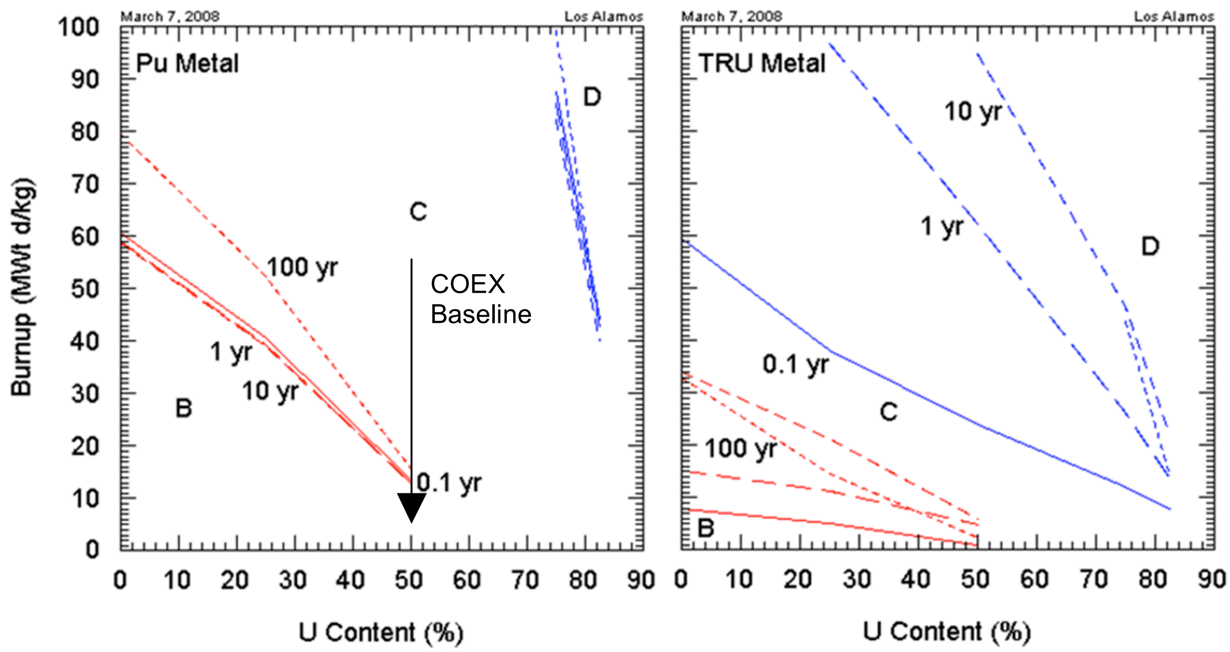

Figure 3: A comparison of the contour lines in burnup – U content space that indicate the boundaries between Attractiveness Levels B and C (red) and between C and D (blue) for Pu (left) and TRU (right) metals. The solid lines and long, medium, and short dashed lines denote material ages of 0.1, 1, 10, and 100 years, respectively.

The Attractiveness Level of TRU, which is a UREX+1A end product, is sensitive to the burnup and to the age of the spent fuel. Furthermore, the Attractiveness Level of TRU increases with age for burnups ≥ 45 MW·d/kg. For burnups < 30 MW·d/kg, TRU reaches its highest Attractiveness Level at an age of 3 years. Consequently, TRU should be reprocessed within 10 years of its generation or sooner if practical. For a burnup of 45 MW·d/kg, a dilution of > 75 % spent or depleted uranium is required to attain an Attractiveness Level of D for TRU metal. The Attractiveness Level of Pu can also be reduced from B to C by either increasing the burnup beyond 55 MW·d/kg or by dilution of ≥ 4-6 % ^{238}Pu content. To attain an Attractiveness Level of D for Pu metal requires a dilution of ≥ 70-80 % ^{238}Pu content, depending on the burnup.

This study is still in progress. The PYROX and THOREX reprocessing schemes are currently being analyzed. Other reprocessing schemes being considered include: DIAMEX-SANEX, GANEX, FLOREX, and DUPIC. Results of the analysis of these reprocessing schemes will be reported in the future.

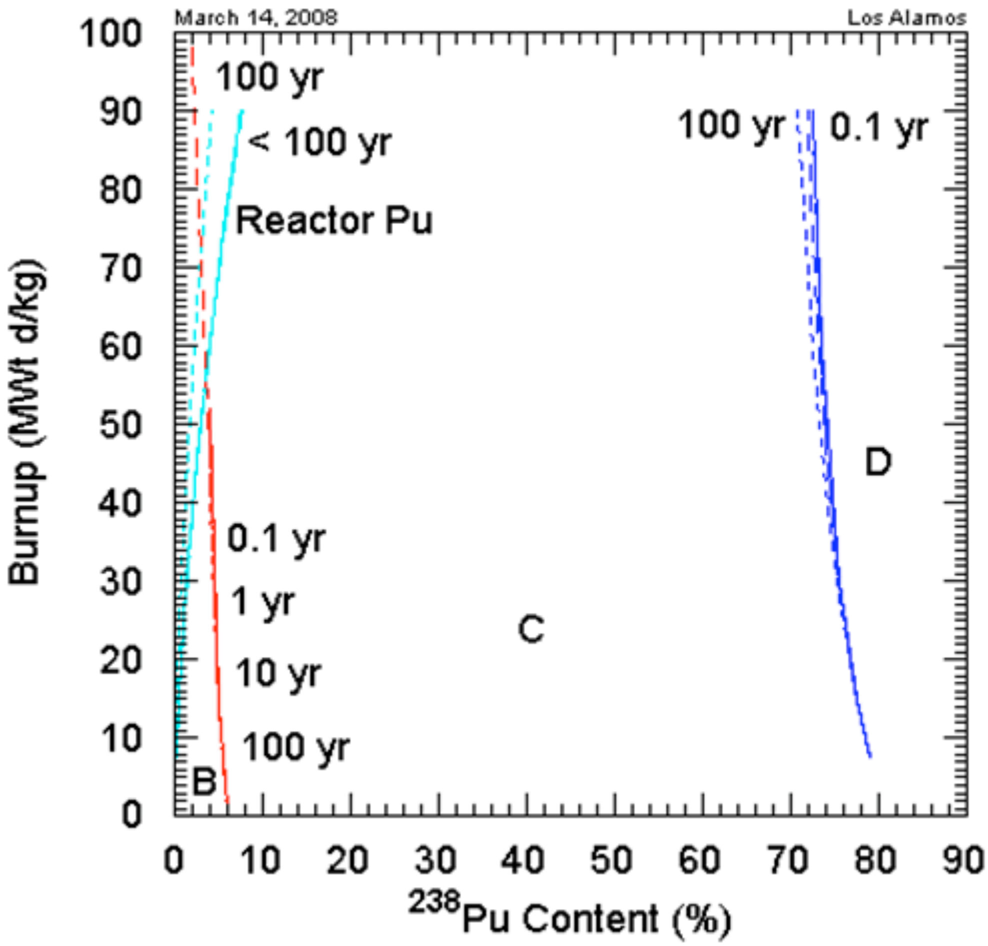

Figure 4: Contour lines in burnup – ^{238}Pu content space that indicate the boundaries between Attractiveness Levels B and C (red) and between C and D (blue) for Pu metal. The solid lines and long, medium, and short dashed lines denote material ages of 0.1, 1, 10, and 100 years, respectively. Also shown in cyan is the ^{238}Pu content of Pu obtained from spent fuel as a function of burnup and age.

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