

LA-UR-09- 02464

Approved for public release;
distribution is unlimited.

Title: The Attractiveness of Materials in Advanced Nuclear Fuel Cycles for Various Proliferation and Theft Scenarios

Author(s): Charles G. Bathke[1], Bartley B.Ebbinghaus[2], Brad W. Sleaford[2], Richard K. Wallace[1], Brian A. Collins[3], Kevin R. Hase[1], Gordon D. Jarvinen[1], Keith S. Bradley[2], John R. Ireland[1], M. W. Johnson[1], Andrew W. Prichard[3], and Brian W. Smith[3]

[1]Los Alamos National Laboratory

Intended for: Global 2009
Paris, France
9/6-11/09



Los Alamos National Laboratory, an affirmative action/equal opportunity employer, is operated by the Los Alamos National Security, LLC for the National Nuclear Security Administration of the U.S. Department of Energy under contract DE-AC52-06NA25396. By acceptance of this article, the publisher recognizes that the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes. Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy. Los Alamos National Laboratory strongly supports academic freedom and a researcher's right to publish; as an institution, however, the Laboratory does not endorse the viewpoint of a publication or guarantee its technical correctness.

Additional Authors

Last	First	Middle	Affiliation
Hase	Kevin	R.	Los Alamos National Laboratory
Jarvinen	Gordon	D.	Los Alamos National Laboratory
Ebbinghaus	Bartley	B.	Lawrence Livermore National Laboratory
Sleaford	Brad	A.	Lawrence Livermore National Laboratory
Bradley	Keith	S.	Lawrence Livermore National Laboratory
Collins	Brian	W.	Pacific Northwest National Laboratory
Smith	Brian	W.	Pacific Northwest National Laboratory
Prichard	Andrew	W.	Pacific Northwest National Laboratory

The Attractiveness of Materials in Advanced Nuclear Fuel Cycles for Various Proliferation and Theft Scenarios

Charles G. Bathke,¹ Bartley B. Ebbinghaus,² Brad W. Sleaford,² Richard K. Wallace,¹ Brian A. Collins,³ Kevin R. Hase,¹ Gordon D. Jarvinen,¹ Keith S. Bradley,² John R. Ireland,¹ M. W. Johnson,¹ Andrew W. Prichard,³ and Brian W. Smith³

¹Los Alamos National Laboratory, P.O. Box 1663, Los Alamos, NM, USA; Email: bathke@lanl.gov

²Lawrence Livermore National Laboratory, P.O. Box L-168, Livermore, CA 94551, USA

³Pacific Northwest National Laboratory, P.O. Box 999, Richland, WA 99352, USA

Abstract – This paper is an extension to earlier studies^{1,2} that examined the attractiveness of materials mixtures containing special nuclear materials (SNM) and alternate nuclear materials (ANM) associated with the PUREX, UREX, COEX, THOREX, and PYROX reprocessing schemes. This study extends the figure of merit (FOM) for evaluating attractiveness to cover a broad range of proliferant state and sub-national group capabilities. The primary conclusion of this study is that all fissile material needs to be rigorously safeguarded to detect diversion by a state and provided the highest levels of physical protection to prevent theft by sub-national groups; no “silver bullet” has been found that will permit the relaxation of current international safeguards or national physical security protection levels. This series of studies has been performed at the request of the United States Department of Energy (DOE) and is based on the calculation of “attractiveness levels” that are expressed in terms consistent with, but normally reserved for nuclear materials in DOE nuclear facilities.³ The expanded methodology and updated findings are presented. Additionally, how these attractiveness levels relate to proliferation resistance and physical security are discussed.

I. INTRODUCTION

The United States Department of Energy (DOE) requested an assessment of the attractiveness, from an international safeguards and national physical protection perspective, of the special nuclear materials (SNM) (*i.e.*, Pu, ²³³U, and ²³⁵U), alternate nuclear materials (ANM) (*i.e.*, ²³⁷Np and Am), and other actinides that have a critical mass (*e.g.*, Cm) that are associated with reprocessing and are handled in forms largely decontaminated of fission products. Each potential malefactor is unique in the material to which he has access and in the degree of sophistication he could utilize in weaponizing the material. Collectively, proliferant states and sub-national groups could consider a broad spectrum of SNM and ANM to be attractive for use in an explosive nuclear device. This paper delineates a set of figures of merit (FOM) that are intended to explain the attractiveness or preferences for a range of nuclear materials across a span of credible nuclear adversaries.

A credible nuclear threat from a sub-national group is different than that from a proliferant state. On the one hand,

the perceived threat from a sub-national group is more dependent upon the fact that a device may produce any nuclear yield than it is upon the actual amount of yield. Even in a low technology, low quality device, any nuclear yield will, in most cases, vastly exceed the conventional explosive yield. Thus, any device capable of generating a nuclear yield in the hands of a sub-national group would meet their requirements. On the other hand, a proliferant state is likely to have a preference for materials that are more easily and efficiently fabricated into higher yield nuclear weapons than those materials of interest to a sub-national group. All SNM and ANM should be protected and safeguarded according to the highest level of attractiveness derived from both of these threats.

The point at which the nuclear explosive energy exceeds the conventional explosive energy is the point at which there is a potential nuclear threat. This point is also known as a threshold nuclear yield. The primary factors of material attractiveness are the bare critical mass, the internal heat generation, and the radiation dose rate.⁴ The spontaneous neutron generation rate, another aspect of material attractiveness, is relevant to cases where militarily

significant nuclear yields are desired,⁵ but the proliferant state is only capable of building less sophisticated devices.

The need to carefully and rigorously distinguish between the proliferation of nuclear weapons or explosive devices by a nation state, and the use of nuclear material by a sub-national terrorist group to produce a nuclear explosive device is explicitly acknowledged in the growing volume of literature on assessing proliferation and physical security risks. Two major international efforts to develop methodologies to assess proliferation resistance — the Generation IV International Forum Working Group on Proliferation Resistance⁶ and the IAEA's INPRO activity⁷ — distinguish proliferation resistance from physical protection, based on the identity of the actors:

- *Proliferation resistance* is that characteristic of a nuclear energy system (NES) that impedes the diversion or undeclared production of nuclear material or misuse of technology by the *Host State* seeking to acquire nuclear weapons or other nuclear explosive devices.
- *Physical protection (robustness)* is that characteristic of an NES that impedes the theft of materials suitable for nuclear explosives or radiation dispersal devices (RDDs) and the sabotage of facilities and transportation by *subnational entities and other non-Host State adversaries*.

This distinction, and limiting the use of the term *proliferation resistance* to describing nation-state proliferation impacts, has been nearly lost in today's lexicon. Statements that characterize the physical protection impacts of a particular NES are often incorrectly associated with the term *proliferation resistance*. This misuse of terms conveys a misleading message with respect to the potential proliferation risks or benefits associated with a particular NES. Material attractiveness is one of several considerations when evaluating either *proliferation resistance* or *robustness* of a nuclear energy system.

This paper will focus on answering four questions that are increasingly being posed of advanced fuel cycles: 1) Is reactor-grade (RG) plutonium attractive for use in a nuclear explosive device, and at what point does increasing the ratio of ²³⁸Pu to other plutonium isotopes make the plutonium unattractive for use in a nuclear weapon or a nuclear explosive device? 2) Do advanced reprocessing approaches [for light-water-reactor (LWR) spent fuel] that produce grouped products in which plutonium is separated with one or more minor actinides render the product unattractive for use in a nuclear weapon or nuclear explosive device without further chemical separation? 3) At what point might diluting plutonium or a transuranic mixture render the mixture as unattractive for use in a nuclear weapon or nuclear explosive device? and 4) Do other advanced fuel cycles (e.g., thorium based cycles) produce products that are potentially attractive for use in a

nuclear weapon or nuclear explosive device? Related and no less important, does the answer to any or all of these four questions depend upon whether the proliferator is a nation state or a sub-national group?

II. METHODOLOGY

The first metric presented [see Eq. (1)] is applicable for evaluating the attractiveness of SNM or ANM for a sub-national group, for most of the less advanced proliferant nations, or for a technically advanced proliferant state. The latter would be capable of building nuclear devices that assemble very rapidly to limit the impact of pre-initiation.⁵ For a sub-national group any nuclear yield is acceptable so pre-initiation is not a significant issue. Such cases are evaluated using the following formula:

$$FOM_1 = 1 - \log_{10} \left(\frac{M}{800} + \frac{Mh}{4500} + \frac{M}{50} \left[\frac{D}{500} \right]^{\frac{1}{\log_{10} 2}} \right) \quad (1)$$

where 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 rad/h. The dose term in Eq. (1) has been modified with the insertion of a multiplicative factor, $M/50$, relative to the form that appears in Refs. 1 and 2, because lower quality, larger bare-critical-mass material requires an increased penalty for high dose due to increased handling requirements.

For a very few relatively unadvanced proliferant nations that desire a reliably high yield, pre-initiation is an issue. The material attractiveness for such a nuclear device must necessarily be reduced for materials with a high spontaneous neutron generation rate. Then, the second variant of the FOM is given by:

$$FOM_2 = 1 - \log_{10} \left(\frac{M}{800} + \frac{Mh}{4500} + \frac{MS}{6.8(10)^6} + \frac{M}{50} \left[\frac{D}{500} \right]^{\frac{1}{\log_{10} 2}} \right) \quad (2)$$

where S is the spontaneous-fission neutron production rate in n/s/kg. In a more specific evaluation using Eqs. (1) and (2), the dose rate, D , is the dose of the starting item and bare critical mass, M ; the heat content, h ; and the spontaneous-fission neutron production rate, S , are of the product material (pure element or impure metal) that is intended for the nuclear device.

The basic concept of the FOM is to relate candidate nuclear material to accepted standards. The four well-established standards are: 1) the threshold for low enriched uranium (i.e., ²³⁵U enrichment less than 20%), 2) radioisotope thermoelectric generator plutonium (i.e., ²³⁸Pu

enrichment greater than 80%), 3) a self-protecting dose rate (*i.e.*, 500 rad/h at 1 m), and 4) only in the case of an unadvanced proliferant state, a spontaneous fission neutron rate of reactor-grade plutonium (*i.e.*, ^{240}Pu content $\geq 20\%$). Historically, the self-protecting dose rate was assumed to be 100 rem/h at 1 m.⁸ Upon recent technical review,^{9,10} it has been increased to 500 rad/h at 1 m. The exponent in the dose term [*i.e.*, $1/\log_{10}(2) = 3.322$] is the result of a requirement to reduce the FOM by 1.0 when the dose increases from 500 to 1,000 rad/h.

→ The FOMs were reviewed by nuclear weapons experts at both LANL and LLNL. While it was determined that there are a number of smaller factors that are not captured, it was agreed that the FOM equations presented herein capture the dominant factors as well as possible in an unclassified format.

Table I gives the meaning of the FOM. To make a material unattractive for use in a nuclear device, the FOM must be less than 1. Note that Table I by itself does not distinguish, even in a qualitative sense, between degrees of proliferation resistance (*e.g.*, high, medium, low) that might characterize a nuclear material or a grouped product. Table I reflects the fact that while a particular nuclear material might be preferable for use in a nuclear weapon or explosive device, the differences do not preclude the design and construction of effective nuclear weapons from any of the materials with a FOM of >1 . For example, plutonium from typical civil spent fuel could be used in a nuclear device.⁸ As this paper will show, plutonium with ^{239}Pu content ranging above 90% (often characterized as low proliferation resistance) or between 50% and 20% (often characterized as high proliferation resistance) both have a FOM > 1 . The fact that potential proliferant states or sub-national groups might "prefer" one material over another should not imply that either material in question is "proliferation-proof," or that any reduction in international safeguards and national physical protection requirements can be justified.

TABLE I

The Meaning of FOM When Applied to Metals or Alloys

FOM	Weapons Utility	Attractiveness	Attractiveness Level ³
> 2	Preferred	High	~B
1-2	Attractive	Medium	~C
0-1	Unattractive	Low	~D
< 0	Unattractive	Very Low	~E

The reprocessing schemes analyzed to date and reported herein are: UREX, COEX, ^{238}Pu spiking, PYROX, and THOREX. The dominant source of material for all of the reprocessing schemes analyzed herein, except THOREX, is spent uranium oxide from pressurized water reactors (PWR) and boiling water reactors (BWR). Such

spent fuel is typically characterized by its burn-up, expressed in MW·d/kg of initial heavy metal. The average burn-up of spent fuel in the USA historically has ranged from ~15 MW·d/kg for BWR and from ~25 MW·d/kg for PWRs¹¹ to present day values of 45 - 50 MW·d/kg. Likewise, within a typical spent fuel pool and even along the length of a typical fuel assembly significant variation of burn-up exists. The isotopic composition of spent fuel was generated with ORIGEN2.2¹² for burn-ups ranging from 7.5 to 90 MW·d/kg for the purposes of this analysis. The calculations of the required ^{235}U enrichment of the fuel charge and the spent fuel compositions are in good agreement with similar published results.¹³⁻¹⁹ Also varied was the spent fuel age at the time of reprocessing relative to the time of discharge.

A burn-up of 45 MW·d/kg was assumed to calculate the isotopic composition of the thorium spent fuel. The burn-up calculations were completed using the TRITON module of SCALE 5.1,²⁰ and the decay after burn-up was modeled using ORIGEN-ARP.²¹ The advantage of TRITON over ORIGEN is that non-standard fuels can be used.

III. RESULTS

Table II provides a list of the UREX products. Figure 1 provides results of FOM calculations using Eq. (1) for the non-uranium bearing products listed in Table II. Additionally, the FOM of weapons-grade plutonium (WG-Pu), high and low enriched uranium (HEU and LEU, respectively), ^{237}Np , and ^{233}U contaminated with 10 ppm ^{232}U are shown on the left side of each figure for reference. The UREX products with the highest FOM₁ (*i.e.*, most attractive) are Pu and Pu+Np, which have nearly the same FOM value. The FOM₁ of Pu and Pu+Np decreases significantly with increasing burn-up, because the concentrations of ^{239}Pu and ^{241}Pu (*i.e.*, the isotopes with relatively high fission cross sections) decrease and the concentration of ^{238}Pu , which is an intense heat source, increases with increasing burn-up. The age of the spent fuel at the time of reprocessing has only a minor effect on the FOM (*i.e.*, the FOM increases slightly with increasing age). Heat is the primary proliferation barrier for Pu and Pu+Np.

The next highest FOM₁ value belongs to TRU. The FOM of TRU decreases significantly with increasing burn-up, because the concentrations of ^{239}Pu and ^{241}Pu decrease with increasing burn-up. However, the FOM of TRU increases significantly with increasing spent-fuel age; because ^{242}Cm and ^{244}Cm , which are intense heating sources, quickly decay away (their half lives are 163 days and 18 years, respectively).

In contrast, the FOM of Am increases with increasing burn-up, because of the build up of ^{243}Am relative to ^{241}Am as the burn-up increases and because ^{243}Am produces less heat relative to ^{241}Am . The FOM of Am decreases with

increasing age, because of the build up in the spent fuel of ^{241}Am relative to ^{243}Am with increasing spent-fuel age due to the beta decay of ^{241}Pu to ^{241}Am .

TABLE II

List of UREX Products and Their Associated Process(es)

Product	Process(es)
Pu + Np	UREX+2, UREX+3, UREX+4
Pu + Np + U	UREX+2a, UREX+3a, UREX+4a
TRU	UREX+1a
TRU + U	UREX+1b
TRU + Ln	UREX+1
Am	UREX+4, UREX+4a
Cm	UREX+4, UREX+4a
Am + Cm	UREX+3, UREX+3a
Am + Cm + Ln	UREX+2, UREX+2a

Although FOM_1 of Cm is too low to appear in Fig. 1, Cm's FOM_1 also increases with increasing burn-up, because of the build up of ^{244}Cm relative to ^{242}Cm as the burn-up increases and because ^{244}Cm produces less heat relative to ^{242}Cm . As with TRU, the FOM_1 of Cm increases significantly with increasing spent-fuel age. Although Cm has a significant neutron dose, the FOM_1 is dominated by its heating. Interestingly, when not chemically separated the Am+Cm mixture has a maximum attractiveness of "low," independent of burn-up and age.

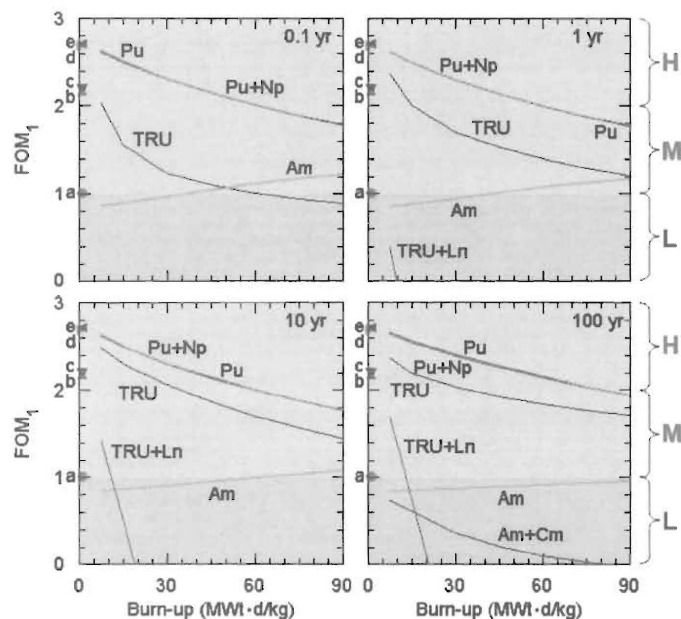


Fig. 1. The FOM_1 of Non-Uranium Bearing UREX Products Versus Burn-up for Various Spent-Fuel Ages at Time of Reprocessing. The letters H, M, and L denote high, medium, and low attractiveness, respectively (see Table I). Included for reference are the following data points: a – LEU (20%), b – HEU (93%), c – ^{237}Np , d – ^{233}U (10 ppm ^{232}U), and e – WG-Pu.

The retention of the lanthanides (Ln) with TRU or Am+Cm greatly reduces the FOM_1 (the FOM_1 of Am+Cm+Ln is too low to appear in Fig. 1), because the lanthanides provide an intense photon dose and, more importantly, an intense heat source. The FOM_1 of TRU+Ln decreases with increasing burn-up, because of the relative build-up of lanthanides with increasing burn-up. The FOM_1 of TRU+Ln increases with increasing age, because the lanthanide half lives are of the order of 100 years.

Figure 2 shows the effect of using different FOM formulae to calculate the attractiveness of the non-uranium bearing UREX products. The magnitude of the penalty incurred in Eq. (2) for spontaneous-fission neutron production relative to Eq. (1) is dramatic for WG-Pu and the UREX products containing plutonium (*i.e.*, Pu, Pu+Np, TRU, and TRU+Ln), and is responsible for the misconception by some that reactor-grade plutonium is not attractive for use in weapons. However, LEU, HEU, ^{233}U , ^{237}Np , and Am do not incur any penalty, and have the same attractiveness independent of the equation used. International safeguards and national physical protection programs for the UREX products should use Eq. (1) to evaluate material attractiveness.

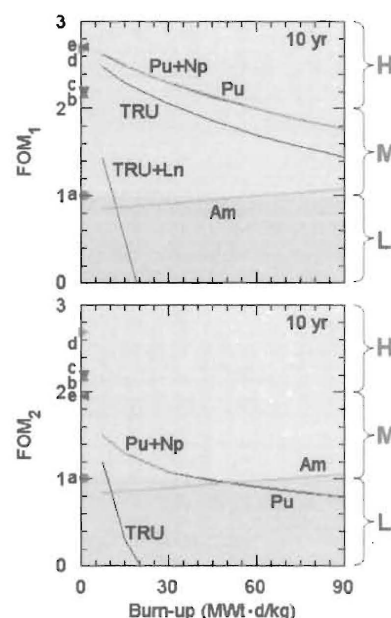


Fig. 2. The FOM_1 of Non-Uranium Bearing UREX Products Processed 10 Years after Discharge Versus Burn-up. Included for reference are the following data points: a – LEU (20%), b – HEU (93%), c – ^{237}Np , d – ^{233}U (10 ppm ^{232}U), and e – WG-Pu.

Figure 3 shows the effect of diluting TRU with incremental fractions of the lanthanides in spent fuel. The fraction of the lanthanides that must be retained with the TRU to render the mixture low attractiveness for use in nuclear weapons depends on the burn-up of the spent fuel.

For example, only 20% of the lanthanides must be retained with the TRU obtained from 10-year old spent fuel burned to 50 MW·d/kg to achieve low attractiveness.

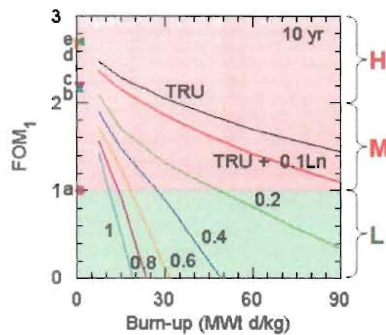


Fig. 3. The FOM₁ of TRU Plus Various Fractions of the Lanthanides in Spent Fuel Versus Burn-up. Included for reference are the following data points: a – LEU (20%), b – HEU (93%), c – ²³⁷Np, d – ²³³U (10 ppm ²³²U), and e – WG-Pu.

Figure 4 shows the effect of diluting Pu and TRU with uranium from the same spent fuel (*i.e.*, < 1% ²³⁵U). The FOM₁ is reduced with the addition of this uranium. However, significant quantities of uranium are required to attain a low attractiveness for use in nuclear weapons. For example, > 80% U is required for Pu, and ~ 75% U is required for TRU, both obtained from 10-year old spent fuel burned to 45 MW·d/kg. It should be noted that this study focused only on the attractiveness of these uranium mixtures and did not consider any subsequent reprocessing or purification by an adversary.

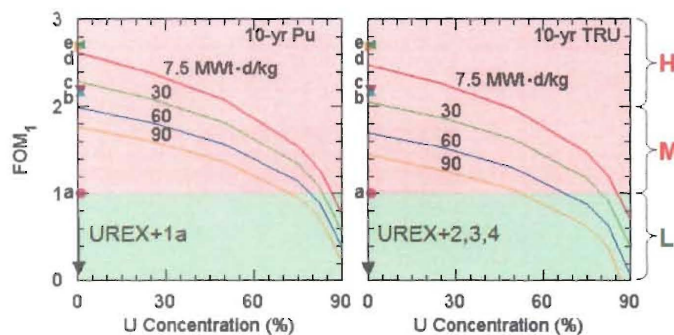


Fig. 4. The FOM₁ of Pu (Left) and TRU (Right) Versus Spent-Uranium Concentration for Various Burn-ups. Included for reference are the following data points: a – LEU (20%), b – HEU (93%), c – ²³⁷Np, d – ²³³U (10 ppm ²³²U), and e – WG-Pu.

Several proposals for "denaturing" the plutonium isotopic vector have been made.^{21,22} Figure 5 shows the results from Eqs. (1) and (2) for "denaturing" the plutonium isotopic vector. Depending on the burn-up, the plutonium in spent fuel has up to 8% ²³⁸Pu. Adding additional ²³⁸Pu further reduces the attractiveness of the plutonium for use

in nuclear weapons. On the one hand, 80% ²³⁸Pu is required to reduce the plutonium to "low" attractiveness using Eq. (1). A sustainable source for that much ²³⁸Pu has not yet been identified. On the other hand, very little additional ²³⁸Pu is required using Eq. (2). Only 8% ²³⁸Pu is enough to drop to "low" attractiveness for the few cases of an unadvanced proliferant state that requires reliably high yield nuclear devices. The analysis of international safeguards and national physical protection issues for all plutonium spiking proposals should use Eq. (1) to evaluate material attractiveness.

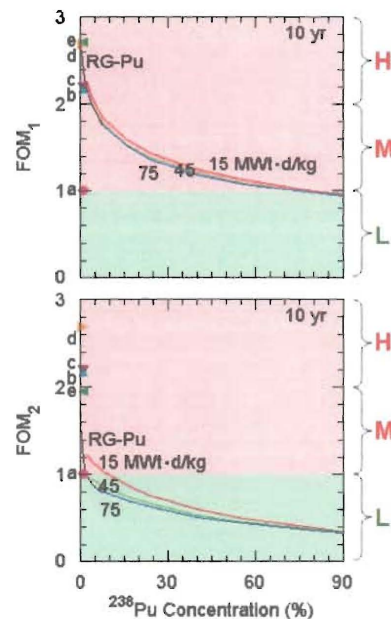


Fig. 5. The FOMs of Pu Versus ²³⁸Pu Concentration (%) for Various Burn-ups. The black line depicts reactor-grade plutonium. Included for reference are the following data points: a – LEU (20%), b – HEU (93%), c – ²³⁷Np, d – ²³³U (10 ppm ²³²U), and e – WG-Pu.

The PYROX²³ products are U with trace (~100 ppm) amounts of TRU and a U+TRU mixture that is 1/3 U with small amounts (12,500 ppm) of the rare earth fission products (REFP). However, there are locations in the flowsheet where the material is more attractive. For example, there is a location where there is no U and a ~5% mixture of active metal and rare earth fission products (AMFP+REFP). For this study, TRU with various concentrations of U and various concentrations of REFP or AMFP+REFP was examined. Figure 6 shows FOM₁ for a range of mixtures of U, TRU, and fission. The addition of increasing quantities of either U or fission products reduces the FOM. For a nominal reactor discharge that has been burned to 45 MW·d/kg and then cooled for 10 years before reprocessing, the TRU from that discharge would require ≥ 75% U for M_{FP}/M_{TRU} = 0, where M_{FP} is the mass of the fission products and M_{TRU} is the mass of the TRU.

Alternatively, the same TRU with 0% U would require $M_{FP}/M_{TRU} > 0.15$ when the fission products used are AMFP+REFP, but $M_{FP}/M_{TRU} > 0.38$ when the fission products used are REFP. Similarly, various combinations of U and M_{FP}/M_{TRU} can be used to achieve low attractiveness. The value of M_{FP}/M_{TRU} in spent fuel ranges from ~ 1.0 at 15-MW·d/kg spent fuel to 3.0 at 90-MW·d/kg spent fuel when the fission products used are

AMFP+REFP; and that same ratio ranges from $\sim 2/3$ at 15-MW·d/kg spent fuel to 2.0 at 90-MW·d/kg spent fuel when the fission products used are REFP.

Figure 6 also displays several trends. Increasing the burn-up reduces the amount of U required for low attractiveness. Increasing the age of the spent fuel at the time of reprocessing requires larger values of M_{FP}/M_{TRU} to achieve low attractiveness.

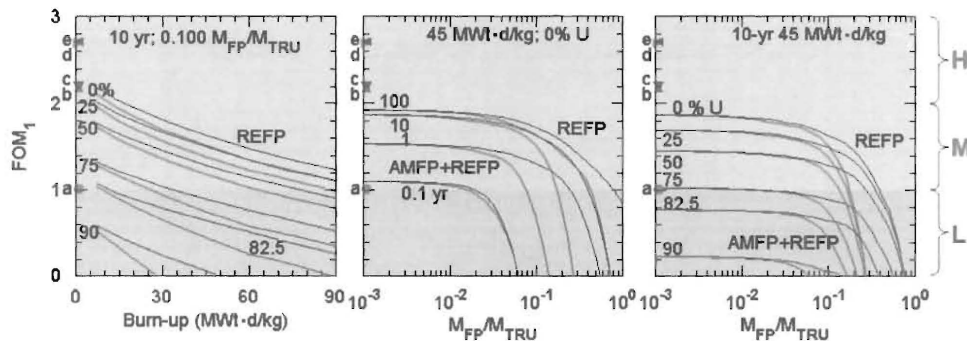


Fig. 6. The FOM₁ of PYROX Material Versus Burn-up and Ratio of Fission-Product Mass to TRU Mass for Various Uranium Concentrations (%) and Spent Fuel Ages. The red curves represent an intermediate product with active metal and rare earth fission products (AMFP+REFP), and the blue curves represent a final product with rare earth fission products (REFP). Included for reference are the following data points: a - LEU (20%), b - HEU (93%), c - ²³⁷Np, d - ²³³U (10 ppm ²³²U), and e - WG-Pu.

Table III provides the compositions at charge of the three thorium fuel mixtures that were analyzed. The percentages of the mixtures constituents were determined by achieving the same average k_{eff} as a LWR (4.08% enriched) at a burn-up of 45 MW·d/kg.

TABLE III

Compositions at Charge of Thorium Cases

Material	Pu (%)	²³⁹ Pu/Pu (%)	U (%)	²³⁵ U/U (%)	Th (%)
1	6.25	94	5.0	0.7	88.75
2	0.00	—	30.5	19.9	69.50
3	10.00	53	5.0	0.7	85.00

As shown in Fig. 7, the thorium fuel cycle produces two isotopes that are of concern from a safeguards perspective: ²³³U and ²³⁹Pu. The ²³³U is bred from thorium, and the ²³⁹Pu is bred from any low enriched, natural, or depleted uranium that is introduced to dilute the ²³³U that is bred from the thorium. For the three cases analyzed herein, the ²³⁹Pu is of greater concern from a safeguards perspective. However, burning thorium fuel produces smaller quantities of ²³⁹Pu than burning uranium fuel. Diluting the ²³⁹Pu with spent thorium requires the mixture to be $> \sim 2/3$ Th to achieve low attractiveness using Eq. (1). Diluting the ²³³U with spent thorium requires the mixture to be $> \sim 1/3$ Th to achieve "low" attractiveness, depending on the initial quantity and quality of the Pu in the thorium charge. If LEU is mixed with the thorium fuel at charge, then there is sufficient ²³⁸U to mask the build up of ²³³U

during its burn and to render the uranium unattractive at discharge.

Figure 7 also provides a gauge of the degradation in material attractiveness that results from materials being "burned" in a thorium-fueled reactor. The attractiveness of WG-Pu at charge is displayed as symbol "e" in Fig.7; whereas the attractiveness of that same plutonium at discharge and whatever plutonium is bred during burn corresponds to the intersection of the line for Mat 1 with the y-axis. Hence, burning WG-Pu in a thorium-fueled reactor degrades the attractiveness by ~ 0.4 . Similarly, the attractiveness values for LEU and reactor-grade plutonium decrease by ~ 0.5 and ~ 0.2 , respectively, by burning to 45 MW·d/kg in a thorium-fueled reactor.

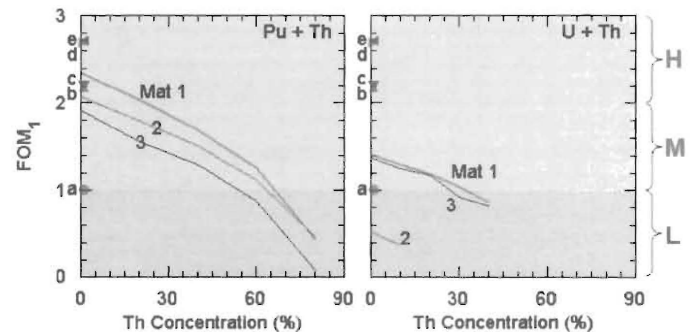


Fig. 7. The FOM₁ of the Pu + Th (Left) and U + Th (Right) THOREX Products at Discharge of the Fuels Listed in Table III Versus Thorium Concentration. Included for reference are the following data points: a - LEU (20%), b - HEU (93%), c - ²³⁷Np, d - ²³³U (10 ppm ²³²U), and e - WG-Pu.

IV. DISCUSSION

The most attractive UREX product is Pu+Np. The FOM of TRU, which is a UREX+1a product, is dependent upon spent-fuel age and burn-up. Because the FOM of TRU increases significantly with spent-fuel age, if spent fuel is going to be reprocessed, then processing should be done as soon as is practical. Furthermore, reprocessed TRU should be burned as soon as is practical, because the FOM of TRU increases significantly with increasing post-reprocessing time.² For 10-yr, 45-MW·d/kg UREX+1a material (i.e., TRU), a U concentration > 75% is required to reduce the FOM₁ to "low" attractiveness.

The Pu+Np product has the same FOM as Pu product; co-extracting Np with Pu does not reduce its attractiveness for use in nuclear weapons. Conversely, extracting just Pu leaves Np in the waste stream. For 10-yr, 45-MW·d/kg COEX material (i.e., Pu + U mixture), a U concentration of ~ 80% is required to reduce the FOM to low attractiveness. The FOM of Pu (and Pu + Np) is not significantly affected by changing the post-irradiation time or by changing the post-reprocessing time.

The FOM₁ of reactor-grade plutonium denatured with ²³⁸Pu concentration < 80% is still at least "medium" attractiveness. "Virtually any combination of plutonium isotopes can be used to make a nuclear explosive device (except > 80% ²³⁸Pu)."²⁴ A device that uses reactor-grade plutonium could have a significant nuclear yield, regardless of the concentration of troublesome isotopes (i.e., ²³⁸Pu and ²⁴⁰Pu).²⁵ Relative to weapons-grade plutonium, reactor-grade plutonium does present some challenges, but these are not considered prohibitive. Radiation levels require more shielding and greater precautions to protect personnel when building and handling nuclear devices made from reactor-grade plutonium than nuclear devices made from weapons-grade plutonium.²⁵ While the heat generated by ²³⁸Pu and the spontaneous-fission neutrons generated from ²³⁸Pu and ²⁴⁰Pu require careful management in a nuclear device, there are well developed means for addressing these problems; they are not a significant hurdle to the production of nuclear explosives, even for developing states or subnational groups.²⁵ At the lowest level of sophistication, a potential proliferant state or sub-national group using designs and technologies no more sophisticated than those used in first-generation nuclear weapons could build a nuclear explosive from reactor-grade plutonium that could have a significant nuclear yield.²⁵ Theft of separated reactor-grade plutonium therefore poses a significant security risk.⁵

The TRU-bearing electro-chemical reprocessing product displays the same characteristics as UREX TRU and TRU + Ln. For a nominal reactor discharge that has been burned to 45 MW·d/kg and then cooled for 10 years before reprocessing, the TRU from that discharge would require ≥ 75% U for M_{FP}/M_{TRU} = 0. Alternatively, the same

TRU with 0% U would require M_{FP}/M_{TRU} > 0.15 when the fission products used are AMFP+REFP, but M_{FP}/M_{TRU} > 0.38 when the fission products used are REFP.

The thorium fuel cycle produces two potentially attractive materials: ²³⁹Pu and ²³³U. The Pu is of greater concern from a safeguards perspective. The Pu product can be rendered unattractive by making a Pu-Th mixture that is > 2/3 Th during/after reprocessing. The ²³³U product can be rendered unattractive by adding natural or depleted U to the fuel before irradiation, but may exacerbate the ²³⁹Pu problem in the product. Additionally, the ²³³U product can be rendered unattractive by making a U-Th mixture that is > 1/3 Th during/after reprocessing.

The addition of a new figure of merit has provided significant insight into material attractiveness. The addition of a penalty term associated with spontaneous-fission neutron production in Eq. (2) reveals that only LEU, HEU, ²³⁷Np, and ²³³U are impervious to its effects. Furthermore, the application of the effects of spontaneous-fission neutrons to all potential nuclear weapons designs may be the source of the misconception by some that reactor-grade plutonium is not attractive for weapons use. It should be noted that any material with a critical mass requires some level of safeguards and security protection consistent with international guidelines regardless of its FOM.

V. CONCLUSIONS

In the introduction, four questions were raised that are increasingly being posed of advanced fuel cycles. Those four questions and their corresponding answers are given below:

1. Is reactor-grade plutonium attractive for use in a nuclear explosive device, and at what point does increasing the ratio of ²³⁸Pu to other Pu isotopes make the plutonium essentially unattractive for use in a nuclear weapon or a nuclear explosive device? *Yes, reactor-grade plutonium is attractive for use in a nuclear explosive device. This conclusion is consistent with Ref. 9 and 24. The ²³⁸Pu concentration must be > 80% to render plutonium unattractive for use in a nuclear weapon or a nuclear explosive device.*
2. Do advanced reprocessing approaches (for LWR spent fuel) that produce grouped products in which plutonium is separated together with one or more minor actinides render the product unattractive for use in a nuclear weapon or nuclear explosive device without further chemical separation? *No, the Pu+Np product has the same FOM as Pu product; co-extracting Np with Pu does not reduce its attractiveness for use in nuclear weapons. Conversely, extracting just Pu leaves Np in the waste stream. Nor does co-extracting Np, Am, and Cm with Pu (i.e., TRU) render the product unattractive for use*

in a nuclear weapon or nuclear explosive device. However, co-extracting Cm with Am produces a product that is unattractive for use in a nuclear weapon or nuclear explosive device; whereas a pure Am product obtained from recently (≤ 10 years) discharged high (> 45 - 60 MW·d/kg) burn-up spent fuel is attractive for use in a nuclear weapon or nuclear explosive device.

3. At what point might diluting plutonium or a transuranic mixture render the mixture as unattractive for use in a nuclear weapon or nuclear explosive device? For 10-yr, 45-MW·d/kg COEX material (i.e., Pu), a ^{238}U content of $\sim 80\%$ is required to be unattractive for use in a nuclear weapon or nuclear explosive device. The TRU from that same discharge would require $\geq 75\%$ ^{238}U or $> 20\%$ Ln to be unattractive for use in a nuclear weapon or nuclear explosive device. However, the addition of the lanthanides to TRU is of limited value, because they must be removed before fabrication of a recycled fuel. The TRU-bearing electro-chemical reprocessing product displays the same characteristics as UREX TRU and TRU + Ln, which are potentially attractive for use in a nuclear weapon or nuclear explosive device.
4. Do other advanced fuel cycles (e.g., thorium based cycles) produce products that are potentially attractive for use in a nuclear weapon or nuclear explosive device? Yes, the thorium fuel cycle produces two potentially attractive materials: ^{239}Pu and ^{233}U .

In general, dilution with ^{238}U , ^{232}Th , or even another inert material increases the bare critical mass and thus reduces the attractiveness of the material. With greater than 80% ^{238}U or 70% ^{232}Th (perhaps less with other materials) the material is "low" attractiveness. Except for dilution of ^{233}U and ^{235}U with ^{238}U , the material can still be made attractive by purification, but this takes time and some degree of technical capability.

The presence of about 8% ^{238}Pu reduces the attractiveness to "medium" except for an unadvanced proliferant state where it reduces the attractiveness to "low." For an advanced proliferant state or a sub-national group, 80% ^{238}Pu is required to reduce the attractiveness to "low." Spontaneous-fission neutron production rate is only significantly relevant to an unadvanced proliferant state. In this case, reactor-grade plutonium is "low" attractiveness. For an advanced proliferant state or a sub-national group, reactor-grade plutonium is still "high" attractiveness for low burn-ups and "medium" attractiveness for high burnups. The analysis of safeguards and physical protection issues should use Eq. (1) to evaluate material attractiveness for all materials. For the most part, dose rate is inconsequential in these analyses. Dose rate will be more

important in future analyses that look at dose rates from specific spent fuel assemblies.

There are safeguards and security benefits to dilution with inert materials and the addition of high heat content or in some cases high spontaneous-fission neutron rate materials. However, we have not identified a "silver bullet" technology that would eliminate safeguards and security issues. None of the proposed flowsheets examined to date justify reducing international safeguards or physical security protection levels. All of the reprocessing or recycling technologies evaluated to date still need rigorous safeguards and high levels of physical protection.

ACKNOWLEDGMENTS

The authors would like to thank Diana Hollis, Pete Jaegers, Matt Kirkland, Jim Koster, Mike Miller, and Stuart Vessard of Los Alamos National Laboratory, Dave Brown of Lawrence Livermore National Laboratory, and Per Peterson of the University of California at Berkeley for their many valuable discussions.

REFERENCES

1. Charles G. Bathke, *et. al.*, "An Assessment of the Proliferation Resistance of Materials in Advanced Nuclear Fuel Cycles," *Proc. of the 8th International Conference on Facility Operations-Safeguards Interface*, Portland Oregon, (March 30 - April 4, 2008).
2. Charles G. Bathke, *et. al.*, "Further Assessments of the Proliferation Resistance of Materials in Advanced Nuclear Fuel Cycles," *Proc. of the 10th Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation*, Mito, Japan, (October 6 - 10, 2008).
3. "Nuclear Material Control and Accountability," U. S. Department of Energy manual DOE M 470.4-6 Chg 1 (August 14, 2006).
4. D. E. Beller and R. A. Krakowski, "Burn-up Dependence of Proliferation Attributes of Plutonium from Spent LWR Fuel," Los Alamos National Laboratory document LA-UR-99-751 (1999).
5. The Department of Energy, "Nonproliferation and Arms Control Assessment of Weapons-Usable Fissile Material Storage and Excess Plutonium Disposition Alternatives," DOE/NN-0007, p.39, Washington DC (1997).
6. Generation IV International Forum, *Evaluation Methodology for Proliferation Resistance and*