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April 30, 2010

European Nuclear Conference  
Barcelona, Spain  
May 30, 2010 through June 2, 2010

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# NUCLEAR MATERIAL ATTRACTIVENESS: AN ASSESSMENT OF MATERIAL FROM PHWR'S IN A CLOSED THORIUM FUEL CYCLE

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## ABSTRACT

This paper examines the attractiveness of material mixtures containing special nuclear materials (SNM) associated with reprocessing and the thorium-based LWR fuel cycle. This paper expands upon the results from earlier studies [1,2] that examined the attractiveness of SNM associated with the reprocessing of spent light water reactor (LWR) fuel by various reprocessing schemes and the recycle of plutonium as a mixed oxide (MOX) fuel in LWR. This study shows that <sup>233</sup>U that is produced in thorium-based fuel cycles is very attractive for weapons use. Consistent with other studies, these results also show that all fuel cycles examined to date need to be rigorously safeguarded and provided moderate to high levels of physical protection. These studies were performed at the request of the United States Department of Energy (DOE), and are based on the calculation of "attractiveness levels" that has been couched in terms chosen for consistency with those normally used for nuclear materials in DOE nuclear facilities [3]. The methodology and key findings will be presented.

## 1. Introduction

The FOM is an empirically derived formula that has been reported in previous publications [1, 2] as a metric to describe the weapons utility of nuclear material. In the context of safeguards and security requirements, the FOM can be equated to the concept of nuclear material attractiveness. The bounding case is referred to as FOM<sub>1</sub> which is applied here to Th-based fuel cycles that are fuelled by <sup>233</sup>U and/or Pu. FOM<sub>1</sub> uses two physical parameters associated with the product material that is to be weaponized (e.g. bare critical mass and heat content) and one physical parameter that is normally associated with the source material from which the weapons usable nuclear material is derived (e.g. dose rate). There is a fourth physical parameter that is relevant to the desirability or preferrability of the material for use in weapons (e.g. intrinsic neutron rate), but it is generally not relevant to preventing the nuclear material from being used to make an effective nuclear weapon.

The metric used herein is given in Eq. (1) and is the bounding case for evaluating the weapons utility of special nuclear material (SNM) or alternate nuclear material (ANM) to various potential adversary groups.

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

In this equation,  $M$  is the bare critical mass 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.

In the context of safeguards, the bare critical mass and the heat content are of the purified element after it has been removed from the used fuel. In the context of security, the bare

critical mass and heat content of an impure alloy that is derived from the spent fuel but not chemically purified would normally be the reference case.

In this study, the dose rate is calculated after the material has been processed for potential weapons use; it is not of the starting item. This is a very conservative approach in accounting for the effect of the dose rate. This basically means that the adversary has access to shielded hot cells or equivalent handling facilities. If the adversary does not have access to these capabilities, then credit can be taken for the size and mass of the fuel assembly. In this case, the dose rate would be taken from the used fuel assembly and the M/50 term would be replaced with an N/10 term, where N is the net weight of the fuel assembly in kg.

The figure of merit formula is derived by comparing the properties of the material in question to accepted standards. The established standards are: 1) the threshold for low enriched uranium (*i.e.*, <sup>235</sup>U enrichment less than 20%), 2) radioisotope thermoelectric generator plutonium (*i.e.*, <sup>238</sup>Pu enrichment greater than 80%), and 3) a self-protecting dose rate (*i.e.*, 500 rad/h at 1 m). Historically, the self-protecting dose rate was taken to be 100 rem/h at 1 m [4]. Upon recent technical review [5,6], an increase to 500 rad/h at 1 m is proposed.

**Table 1** shows the relationship between FOM<sub>1</sub>, weapons utility, and materials attractiveness in a safeguards and security context. Materials that have a FOM<sub>1</sub> greater than 1 are attractive for weapons use and materials that have a FOM<sub>1</sub> less than 1 are not attractive for weapons use. Beyond this simple binary distinction, it should be noted that the lower the FOM<sub>1</sub> the better. Even though a material may still need to be safeguarded and secured, a process that produces a material with a FOM<sub>1</sub> of 1.1 should be encouraged over a process that produces a material with a FOM<sub>1</sub> of 2.5.

**Table 1. The relationship between FOM<sub>1</sub>, weapons utility, and materials attractiveness**

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

\*The DOE attractiveness levels are in reasonably good alignment with FOM<sub>1</sub> for metals and alloys.

FOM<sub>1</sub> was 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 FOM<sub>1</sub> captures the dominant factors quite nicely in an unclassified format.

The FOM represents a small but important part of the overall proliferation and security risks that are posed by various materials and processes in the nuclear fuel cycle. To contextualize the FOM, it overlaps strongly with one of the six proliferation resistance measures (Fissile Material Type) that is identified in the PR&PP methodology [7], and it overlaps strongly with the material attractiveness criteria which is a key part of the DOE graded safeguards table [3]. So in the case of proliferation resistance there are five other factors that need to be considered, *e.g.* proliferation technical difficulty, proliferation cost, proliferation time, detection probability, and detection resource efficiency. In the case of physical protection,

there are two other factors that need to be considered, e.g. material quantity and security category.

## 2. Background and Approach

For many years India has promoted the long term goal of a sustainable fuel cycle based on  $^{233}\text{U}$  and Th. With the discovery of its own large thorium reserves, the United States has taken a renewed interest in this fuel cycle. The current worldwide fleet of light and heavy water reactors breed reactor-grade plutonium which is weapons usable largely because the bare critical mass is small. The impurities in the plutonium,  $^{238}\text{Pu}$  &  $^{240}\text{Pu}$ , increase the heat content and the intrinsic neutron rate, respectively, thus making the material slightly less attractive. A new generation of light or heavy water reactors based on Th will produce  $^{233}\text{U}$ , which is also weapons usable because of its small critical mass. The primary impurity in the uranium,  $^{232}\text{U}$ , substantially increases the dose rate of the material. The high dose rate arises from an intense high energy gamma-ray decay from  $^{208}\text{Tl}$ , a daughter product of  $^{232}\text{U}$ . This study evaluates the attractiveness of the  $^{233}\text{U}$  that is produced in Th-based fuel cycles and compares it to other nuclear materials of interest.

For the purpose of this study, the reactor design is assumed to be that of the advanced heavy water reactor (AHWR) that has been proposed by India [8]. The AHWR is a vertical pressure tube type reactor cooled by boiling light water and moderated by heavy water. The reactor is designed for a sustainable Th-based fuel cycle. The used fuel is reprocessed to extract and recycle the  $^{233}\text{U}$ . The reactor uses  $^{233}\text{U}$ -Th rods in the inner fuel blanket and Pu-U rods in the outer fuel blanket.

In this analysis, three different material cases have been considered: 1. Pu-Th Fuel Rods, the 20 Rod Outer Fuel Blanket, 2.  $^{233}\text{U}$ -Th Rods, the 16 Rod Inner Fuel Blanket, and 3. Total  $^{233}\text{U}$ -Pu-Th in the assembly. The three cases are used to determine the attractiveness of the individual Pu-Th rods, the  $^{233}\text{U}$ -Th rods, and the complete assembly. Although a specific reactor design is considered, it is clear from the results that follow that the design and type of fuel cycle chosen will not have much effect on the overall attractiveness of these materials. The isotopic compositions of the used fuel were determined using SCALE [9]. The physical properties of the materials for the FOM calculations were determined using MCNP-X [10].

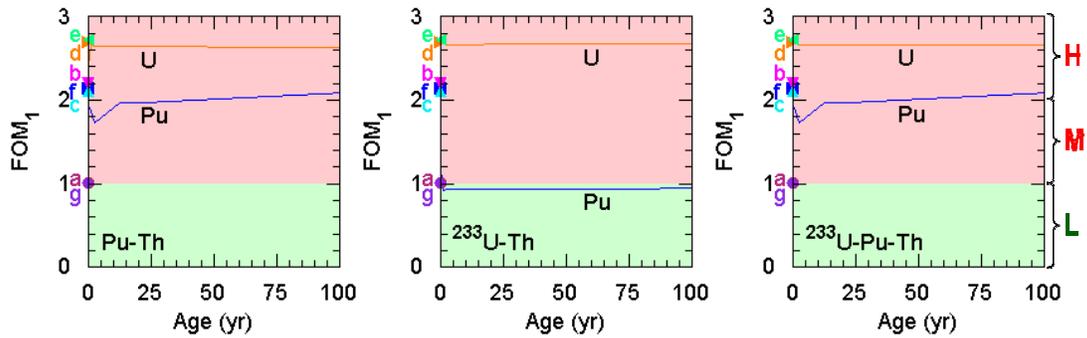
## 3. Results and Discussion

For each of the three cases, the  $\text{FOM}_1$  is calculated as a function of age, measured from insertion into the reactor. Then the material is in the reactor for the first two and a half years. This corresponds to a burn-up of 20,000 MWD/tHM. The remaining age is cooling time out of the reactor. The  $\text{FOM}_1$  is calculated for  $^{233}\text{U}$  and Pu when separated from the Th fuel matrix. In all cases, the  $\text{FOM}_1$  of the unseparated fuel has a value of  $-\infty$ . The Th-based fuel at charge even in metal form does not have a critical mass unless it is heavily moderated.

For the outer Pu-Th rods, the inventory of Pu per the outer 20 rods (~50 kg of heavy metal at start-up) is 2.58 kg and the inventory of  $^{233}\text{U}$  and Pu at discharge is 0.35 and 1.82 kg, respectively. For the inner  $^{233}\text{U}$ -Th rods, the inventory per the inner 16 rods (~30 kg of heavy metal) of  $^{233}\text{U}$  at start-up is 2.20 kg and the inventory of  $^{233}\text{U}$  and Pu at discharge is 2.04 kg and 0.3 mg, respectively. For the combined  $^{233}\text{U}$ -Pu-Th rods, the inventory of  $^{233}\text{U}$  and Pu (~80 kg of heavy metal) per the entire assembly at start-up is 2.20 and 2.58 kg, respectively. The inventory of  $^{233}\text{U}$  and Pu at discharge is 2.38 and 1.82 kg, respectively. Total mass of a typical assembly is 181 kg. The equilibrium concentration of  $^{232}\text{U}$  in the  $^{233}\text{U}$  in a closed, sustainable fuel cycle will be between 700 and 1,000 ppm.

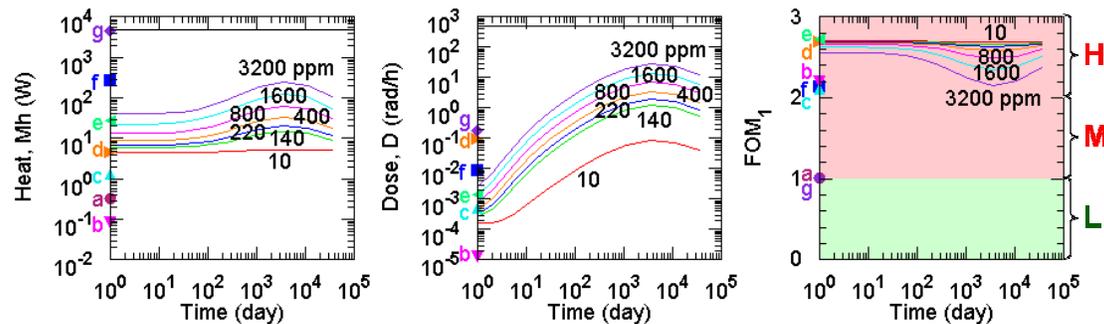
**Figure 1** shows the  $FOM_1$  for the SNM component in the used nuclear fuel. All of these materials are very attractive for nuclear weapons use, except for the small amount of Pu in the  $^{233}\text{U}$ -Th rods which is over 90%  $^{238}\text{Pu}$ . Only the Pu in the Pu-Th rods displays any significant change for the time period shown. For both cases, the Pu  $FOM_1$  drops while in the reactor because  $^{239}\text{Pu}$  is consumed. After discharge, the Pu  $FOM_1$  increases because initially  $^{241}\text{Pu}$  is decaying away and then  $^{238}\text{Pu}$ .

The intact used fuel before reprocessing is not substantially different in attractiveness than ordinary used LWR or HWR fuel. In other words, the Th-based spent fuel is not attractive as long as the dose rate is on the order of 500 rad/h at one meter or higher. A more detailed analysis of the attractiveness of the intact used fuel assemblies as a function of age is still needed to show how long the intact used fuel will remain self-protecting.



**Figure 1.** Figure of merit as a function of age. The first 2.5 years are in the reactor. The remaining years are out of the reactor. Reference points are labeled as follows: a-LEU (20%  $^{235}\text{U}$ ), b-HEU (93%  $^{235}\text{U}$ ), c- $^{237}\text{Np}$ , d- $^{233}\text{U}$  (10 ppm  $^{232}\text{U}$ ), e-WG-Pu (94%  $^{239}\text{Pu}$ ), f-RG-Pu (24%  $^{240}\text{Pu}$ ), and g- $^{238}\text{Pu}/^{239}\text{Pu}$  (80:20)

One can also examine the attractiveness of  $^{233}\text{U}$  as a function of  $^{232}\text{U}$  content and age. In **Figure 2**, the heat term (bare critical mass times heat content) and the dose term are plotted as a function of age. The bare critical mass is for practical purposes constant as a function of age, *i.e.* 15.5 kg. The  $FOM_1$  is also plotted as a function of age. In summary  $^{233}\text{U}$  is very attractive at any practical concentration of  $^{232}\text{U}$  and age. The material is the least attractive at about 10 years of aging. There is little difference in attractiveness for freshly purified materials and for very old materials. The curve with 800 ppm of  $^{232}\text{U}$  most closely matches what is expected in these Th-based fuelled reactors.



**Figure 2.** Heat, Dose, and  $FOM_1$ , as a function of decay time for various initial concentrations of  $^{232}\text{U}$  in  $^{233}\text{U}$ . Reference points are labeled as follows: a-LEU (20%  $^{235}\text{U}$ ), b-HEU (93%  $^{235}\text{U}$ ), c- $^{237}\text{Np}$ , d- $^{233}\text{U}$  (10 ppm  $^{232}\text{U}$ ), e-WG-Pu (94%  $^{239}\text{Pu}$ ), f-RG-Pu (24%  $^{240}\text{Pu}$ ), and g- $^{238}\text{Pu}/^{239}\text{Pu}$  (80:20)

## 4. Conclusions

Thorium-based reactors produce very attractive materials. The  $^{233}\text{U}$  that is produced has a substantial amount of  $^{232}\text{U}$ . The presence of  $^{232}\text{U}$  increases the dose of the material particularly at ages of about 10 years after irradiation. This is due to the in growth of  $^{208}\text{Tl}$  which has an intense high energy gamma-ray emission. In terms of weapons utility or material attractiveness this dose rate is only a nuisance to the adversary. It is not anywhere near sufficient to incapacitate a dedicated adversary. So if long term health and safety is not a concern to the adversary,  $^{233}\text{U}$  is one of the most attractive of all nuclear materials.

Even though  $^{233}\text{U}$  is very attractive, like reactor-grade Pu, it is not normally attractive when it is contained within used nuclear fuel. The high dose rate of the used fuel in combination with the large mass of the used fuel assembly and the low concentration of SNM makes the material self-protecting for many years. Like used LWR and HWR fuels, however, the material eventually becomes attractive as the dose rate decays with age.

Consistent with other studies of fuel cycles, the Th-based materials and processes need high levels of safeguards and moderate to high levels of security. Full safeguards would be needed on all facilities handling greater than 8 kg of  $^{233}\text{U}$  and Pu. However, security can be reduced for the used fuel while the dose rate is high enough for it to be self-protecting (e.g. Cat III), but security needs to be high in the recycling and fuel fabrication facilities (e.g. Cat I) and moderate to high in any fresh fuel handling facilities (e.g. Cat II or I).

## 5. References

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<sup>1</sup> Charles G. Bathke, *et al.*, "An Assessment of the Attractiveness of Material Associated with a MOX Fuel Cycle from a Safeguards Perspective," *Proc. of INMM 50<sup>th</sup> Annual Meeting*, July 12 – 16, 2009, Tucson, Arizona.

<sup>2</sup> Charles G. Bathke, *et al.*, "The Attractiveness of Materials in Advanced Nuclear Fuel Cycles for Various Proliferation and Theft Scenarios," *Proc. of Global 2009*, September 6-11, 2009, Paris, France.

<sup>3</sup> "Nuclear Material Control and Accountability," U. S. Department of Energy manual DOE M 470.4-6 Chg 1 (August 14, 2006).

<sup>4</sup> The Spent-Fuel Standard for Disposition of Excess Weapon Plutonium: Application to Current DOE Options Panel to Review the Spent-Fuel Standard for Disposition of Excess Weapon Plutonium, Committee on International Security and Arms Control, National Academy of Sciences, 28 (2000).

<sup>5</sup> C. W. Coates, B. L. Broadhead, A. M. Krichinsky, R. W. Leggett, M. B. Emmett, J. B. Hines, "Radiation Effects on Personnel Performance Capability and a Summary of Dose Levels for Spent Research Reactor Fuels," Oak Ridge National Laboratory document ORNL/TM-2005/261 (2005).

<sup>6</sup> The Department of Energy, "Technical Review of the Department of Energy Graded Safeguards Table," DOE/HSS, Washington DC (2007).

<sup>7</sup> "Evaluation Methodology for Proliferation Resistance and Physical Protection of Generation IV Nuclear Energy Systems, Revision 5," November 30, 2006. ([www.gen-4.org/Technology/horizontal/PRPPEM.pdf](http://www.gen-4.org/Technology/horizontal/PRPPEM.pdf))

<sup>8</sup> A. Kumar, U. Kannan, Y. Padala, G. M. Behera, R. Srivenkatesan, and K. Balakrishnan, "Physics design of advanced heavy water reactors utilizing thorium," Proceedings of three meetings held in Vienna in 1997, 1998 and 1999, IAEA-TECDOC-1319, pp 165-1785 (1999) ([www.iaea.org/inisnkm/nkm/aws/fnss/fulltext/te\\_1319\\_16.pdf](http://www.iaea.org/inisnkm/nkm/aws/fnss/fulltext/te_1319_16.pdf))

<sup>9</sup> SCALE: A Modular Code System for Performing Standardized Computer Analyses for Licensing Evaluation - Version 6. Oak Ridge National Laboratory. ORNL/TM-2005/39. January 2009.

<sup>10</sup> Denise B. Pelowitz, ed., "MCNPX User's Manual, Version 2.6.0," Los Alamos National Laboratory report, LA-CP-07-1473 (April 2008).