Analysis of Proliferation Resistance of Nuclear Fuels for Microreactors

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INTRODUCTION

With the growing interest in microreactors for remote power applications, climate change mitigation, and energy security, the fundamental challenge of nuclear proliferation must be addressed. As current events in Ukraine have demonstrated, nuclear facilities and materials may be seized by unfriendly state or non-state actors. With the development and envisioned future use of microreactors, the possibility of either covert or forcible overt expropriation of fuel materials should be considered.

Generally, most unirradiated nuclear reactor fuel used today is not very attractive for use in nuclear weapons. Once irradiated, however, some fertile isotopes present in the fuel will have been converted to fissile isotopes. The presence of these fissile materials, if readily separable, can render the irradiated fuel more attractive as a candidate for the production of nuclear explosive devices (NEDs).

To this end, research has been conducted to determine a methodology by which prospective nuclear core concepts—fuel type, core design, and operating cycle—can be evaluated for material attractiveness for nuclear weapons applications. By evaluating core concepts for proliferation resistance, a choice deemed unattractive for weapons use but advantageous for use in a microreactor may be identified. This project seeks to explore the current methods of assessing proliferation resistance and material attractiveness of nuclear fuels and examines areas of improvement for quantitative assessment methods.

CURRENT METRICS

Previous research has been done to explore the methodology of assessing nuclear fuel combinations for material attractiveness to non-state actors for nuclear proliferation. One such paper explores the potential use of irradiated thorium fuel as the principle material in a nuclear weapon [1]. This paper, written by Lisowski, expands upon a previous work done to develop a set of factors characterizing the attractiveness of nuclear materials associated with a uranium-plutonium fuel cycle. The paper undertakes a quantitative analysis of the core burnup and then analyzes the associated requirements for a nuclear explosive device through a mixed qualitative and quantitative analysis of six factors. These factors, taken from a paper by Aoki, Sagara, and Han, are the net weight of total material, acquisition time of the feed material, radiation dose rate during processing, processing time and complexity, bare critical mass, and heat content [2].

The studies by Lisowski and Aoki both assume that the non-state actors possess relatively low technical capabilities, particularly in the area of isotopic separation. This assumption is important and the non-state qualifier is also noted here as in a larger state sponsored nuclear weapons program, it could be assumed that, due to a greater access to financial and technological resources, the processing of any nuclear material of interest could be carried out with fewer technical barriers [2].

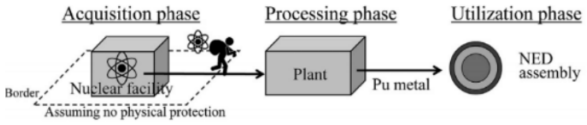

Fig. 1. Phases in Nuclear Explosive Device development and assembly.[2]

Figure 1 outlines the stages of development of a nuclear explosive device. The first phase is an acquisition phase, where the actor would need to acquire the materials necessary to create a NED. Of the six factors, "net weight, acquisition time, and radiation dose are indicators of the acquisition phase." [3] Net weight and radiation dose rate can be quantitatively assessed. The net weight is derived from the bare critical mass (BCM) of the weapon material and is generally defined to be total weight of irradiated fuel material necessary to obtain a Category I quantity of the fissile material; where Category I is taken to be one eighth of the BCM of the material in a metallic form [2]. The acquisition time is a qualitative measure as it is not based on calculation; rather, it is a function of the nuclear facility's security and ease of access to the non-state group.

The final factors associated with NED development are the required bare critical mass (BCM) and heat content (HC) [3]. These are associated with the utilization phase of NED development and focus on the quality and quantity of material that is available after the processing steps. The BCM is the mass of material after processing that when placed in a bare, unreflected metallic sphere would reach criticality.

METHODS

This analysis was conducted in the Lisowski paper using OpenMC and its criticality search feature based on the isotopic composition of the burnup calculation results. This method yielded good quantitative results. Since the publication of Lisowski's work, there have been additional features developed in OpenMC that may be useful in furthering the computational analysis undertaken by her project.

Identifying the core concepts of interest is the first step in analyzing the material attractiveness of a concept. For this project, we first chose to analyze a light water cooled, low enriched uranium (LEU) fuel concept similar to pressurized water reactors (PWRs) operated around the world. This concept was used to develop the workflow for our analysis and establish a baseline against which alternative core concepts could be compared.

In order to conduct the analysis using OpenMC, a model

must be developed and constructed in order to simulate the material composition and geometry of a core concept. For this project, we created a pincell model comprising a fuel pin and surrounding water. The fuel pin diameter, clad thickness, and pitch are based on the AP1000 design [4]. Relevant model parameters and dimensions are given in Table I. This pincell model had reflective boundaries, approximating an infinite lattice and thus is representative of a typical fuel pin within an interior fuel assembly. The pincell geometry is displayed in Figure 2.

TABLE I. Pincell Model Parameters.

Parameter	Value
Fuel Radius	0.42 cm
Clad Outer Radius	0.45 cm
Pitch	1.176 cm
Pin Height	300 cm
Fuel Temp	900 K
Moderator Temp	600 K
Moderator Density	0.712 g/cm^3

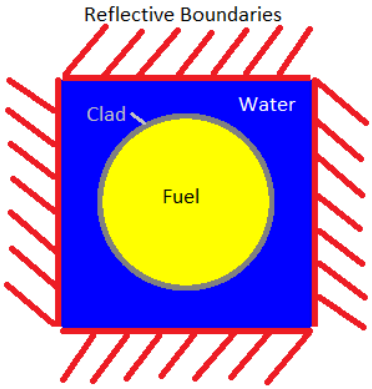

Fig. 2. Model of core pincell showing reflective boundaries and materials used.

Modeling of the core concept of interest includes the fuel operating cycle. For this project we approximated the operating pattern of typical PWR fuel by assuming 54 months (three 18-month cycles) at full power followed by two years of cooling down in a spent fuel pool. We chose a specific power of 30.5 kW/g as representative for a PWR at full power. Burnup calculations were carried out with OpenMC using the ENDF/B-VII.1 depletion chain for PWR spectrum and the CE/CM integrator. Each depletion timestep was simulated using 250 batches with 20,000 particles in each batch. The first 50 batches in each timestep were inactive.

A second core concept based on the Project Pele reactor was also modeled using a similar pincell geometry. The reactor design is a gas cooled reactor (GCR) with High Assay Low Enriched Uranium (HALEU) TRISO fuel in a prismatic compact [5]. Instead of modelling each individual TRISO fuel particle, a geometry consisting of a larger prismatic compact was assumed and the associated volume fraction of the fuel based upon a packing fraction of 0.3 was used. The geometry of the HALEU GCR pincell is included in Figure 3 and the relevant parameters used for this model are included in Table II.

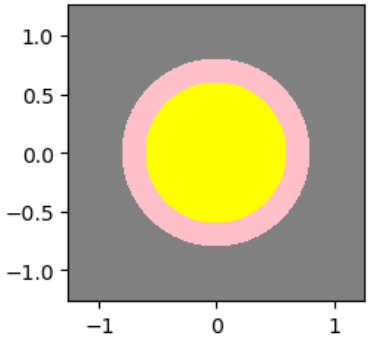

Fig. 3. Model of GCR pincell geometry.

TABLE II. TRISO Volume-Weighted Atom Densities.

Element or Isotope	Atom Density ($\frac{atom}{barn \cdot cm}$)
Carbon	0.077953
Silicone	0.0025875
Oxygen	0.0021279
U-238	0.00085221
U-235	0.00021172
Total	0.08373233

The output of the burnup analysis is isotopic composition of the irradiated fuel. As the core concept under consideration used LEU, plutonium was the target material for nuclear proliferation. For work reported here, the irradiated fuel composition was used to estimate the bare critical mass of chemically separated plutonium as well as the specific activity by isotope in Bq/g.

In order to conduct the BCM calculation, a pure metallic form of the isotope of interest is established. The isotopic composition of the plutonium in the irradiated fuel was parsed from the burnup output. We assumed no enrichment would be possible for the extracted plutonium, thus all isotopes of plutonium were included. We assumed the plutonium could be converted to a metal form with density of 19.84 g/cm^3 [6].

The BCM calculation was carried out using OpenMC. We assumed the metal plutonium would be formed into a sphere with vacuum boundary conditions. A representation of the model is shown in Figure 4. The criticality search feature of OpenMC was used with a tolerance of 150 pcm.

Once the critical radius is determined, the mass of the sphere can be determined using the assumed density and known spherical geometry. This yields the BCM, or the mass required to reach criticality in an unreflected spherical geometry. BCM calculations were also conducted using varying grades of plutonium available in the PNNL material compendium such as Department of Energy weapons grade plutonium as well as fuel and power grade plutonium compositions and used for comparison.

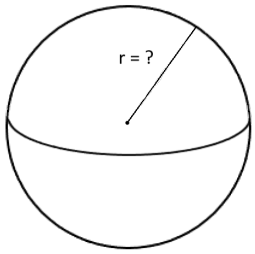

Fig. 4. Representation of bare sphere with variable radius.

RESULTS AND DISCUSSION

The results of the specific activity calculation are shown below in Table III and can be used to determine the dose rate as well as the heat content of the spent fuel. Both of those calculations will be pursued in order to create a more developed picture of the irradiated fuel properties. The resulting distribution of specific activity of select isotopes in the irradiated fuel is displayed in Figure 5.

TABLE III. Total Specific Activity.

Core Concept	Specific Activity (Ci/g)
LEU PWR	12.378
HALEU GCR	0.295

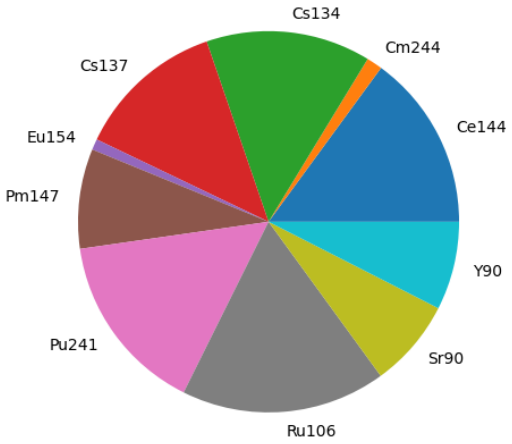

Fig. 5. Pie chart with relative activity of major isotopes present in the spent fuel (Bq/g).

The method to extract a single element from the spent fuel was successful and was able to be used in the BCM calculation in the code. The resulting BCMs for various plutonium compositions are shown below in Table IV. While the BCM is one good measure of the suitability of the material for creating a NED, other factors are important too. In particular, the presence of isotopes like Pu-240 that undergo spontaneous fission, detracts from that suitability in ways that may not be captured in the BCM alone.

TABLE IV. Plutonium Bare Critical Mass Results.

Pu Material	% Pu-239	BCM (kg)
Weapons Grade	93.52%	10.470
Fuel Grade	86.15%	11.126
Power Grade	62.51%	12.841
Spent PWR Fuel	48.56%	14.380
Spent GCR Fuel	83.03%	11.228

CONCLUSIONS AND FUTURE WORK

Current methods of analyzing fuels for proliferation resistance and material attractiveness are thorough and cover multiple aspects of the material utilization portion of nuclear weapons development. So far in this study we have quantitatively estimated the bare critical mass and specific activity of irradiated LEU PWR fuel and irradiated HALEU TRISO GCR fuel. However, further strides may be taken in the quantification of some of the factors of material attractiveness for this work using OpenMC. The quantification of the radiation dose rate and heat content may be able to be further refined and analyzed using OpenMC. These factors may also be incorporated into a methodology where different core concepts can be analyzed and compared through a common process. The standardization of the analysis and subsequent comparison of different fissile target isotopes, would also be an improvement in this research area.

This analysis assumed that the non-state actors captured the irradiated fuel only at the end of a complete fuel cycle. It is conceivable that prospective proliforators might obtain the materials at some mid-cycle point where the composition of the irradiated fuel may be more or less attractive for creating a NED. Consequently, even for the PWR-fuel reference case, it may be valuable to evaluate instances where the fuel was not irradiated to the end of its normal cycle.

Some of the original goals of this project include the quantitative analysis of the dose rate of the spent fuel to be processed as well as the specific heat content of the fuel. Present work is ongoing to complete these milestones before the ANS Student Conference.

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