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ARTICLE

Lightbridge nuclear fuel recycling to strengthen nuclear nonproliferation

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Recycling the uranium (U) in used nuclear fuel after subjecting it to reprocessing can reduce the mass of high-level radioactive waste by about 95% while providing valuable feed material for new nuclear fuel. Recycled U differs from that of mined U (natural U) due to the increased concentration of ^{236}U and left over ^{235}U, which is higher in concentration than in natural U. ^{236}U is a neutron absorber and is hence considered as an unwanted isotope in the nuclear fuel. However, research shown in this paper highlights how ^{236}U can be used to strengthen nuclear nonproliferation by reducing the material attractiveness for nuclear weapons purposes of the plutonium (Pu) produced in the used nuclear fuel. ^{236}U is created when a ^{235}U atom absorbs a neutron and does not undergo fission. This means that nuclear fuel that has higher concentration of ^{235}U and is irradiated with neutrons longer (higher burnup) has a larger concentration of ^{236}U in the used fuel. Lightbridge Corporation is developing an innovative metallic nuclear fuel for current pressurized water reactors (PWR), as well as other thermal nuclear reactor designs, that has a delta-phase U-Zr composition. The PWR fuel can have a ^{235}U enrichment of 19.7 wt.% and a very high burnup of 21 at.% (about 190 GWd/MTU). The combination of higher ^{235}U enrichment and the high burnup results in a high isotopic concentration of ^{236}U (4 wt.%) in the used nuclear fuel. Recycling and re-enriching of this U in the used nuclear fuel results in a U isotopic composition of 0.2% ^{234}U, 5.0% ^{235}U, 8.4% ^{236}U, and 86.4% ^{238}U for traditional UO_2 nuclear fuel and 0.8% ^{234}U, 19.7% ^{235}U, 27.2% ^{236}U, and 52.3% ^{238}U for the metallic nuclear fuel. Fuel burnup simulations using Monte Carlo based radiation transport show that both these recycled and re-enriched nuclear fuels have sufficient fissile content to fuel a nuclear reactor for the standard 3-year timeframe and that the Pu from their used fuel is practically unusable for nuclear weapons purposes because of the higher buildup of ^{238}Pu resulting from the higher concentration of ^{236}U present in the fuel. The higher buildup of ^{238}Pu is through a neutron capture by ^{236}U followed by a beta decay to ^{237}Np, which then captures another neutron and undergo a beta decay. The high decay heat and spontaneous fission neutron emission due to the higher concentration of ^{238}Pu in Pu create significant barriers for use in a nuclear weapon. Fuel burnup simulations show that the traditional UO_2 used fuel containing the recycled and re-enriched U has a ^{238}Pu concentration of 14.2 wt.% while that of the metallic fuel is even higher at 38.4 wt.%. Nuclear material attractiveness assessments for weapons usability indicate that the Pu from both the used nuclear fuels are unattractive and are impractical for use in a nuclear weapon.

Keywords: recycling used nuclear fuel; nonproliferation; proliferation resistance; material attractiveness; metallic fuel

1. Introduction

It is becoming increasingly clear to the scientific community that limiting greenhouse gas emissions is essential to mitigating the most significant effects of climate change. The United Nations estimates that "on the current path of carbon dioxide emissions, temperature could increase by as much as 4.4°C by the end of the century" [1]. There are multiple sources that contribute to greenhouse gas emissions as shown in **Figure 1** with electricity and heat production accounting for 25% of all energy use [2]. Electricity production is speculated to occupy a larger part of energy consumption in the future as developing countries increase their electricity use and developed countries transition their transportation sector from fossil fuels to electricity. Increasing the amount of electricity produced using fossil fuel power plants does not address the impending climate change due to increased greenhouse gasses in the atmosphere. Non-fossil-based power plants primarily consist of hydropower, nuclear, wind, solar, and biofuels as shown in **Figure 2** [3]. Each of these low-carbon sources of electricity has their own challenges for widespread implementation into an

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electric grid, however, those concerning used nuclear fuel will be the focus of this paper.

One of the primary perceived challenges with electricity production using nuclear fuel is the concern with the management of used nuclear fuel. The vast majority of this used nuclear fuel is U, with the remaining mass consisting of stable fission products, Pu, and less than 1% of other nuclides, as shown in **Figure 3** [4]. While the radioactive fission products are of greatest concern after the used fuel has been removed from the reactor due to the intense radiation and heat they create, their relatively short half-lives mean that most of the danger (radiotoxicity) of these fission products are gone after approximately 300 years, as seen in **Figure 4** [5]. What remains the primary danger after approximately 300 years are the transuranic elements, Pu and americium being the primary. The U and Pu in the used nuclear fuel can be recycled after reprocessing and used as fissile material in fresh fuel. This has been done by nations such as France and Japan. This recycling of used nuclear fuel not only reduces the long-term danger posed by used nuclear fuel but also reduces the mass of material that must be disposed of as radioactive waste by greater than 95%. The two primary concerns with recycling nuclear fuel are proliferation and economics, with proliferation being the focus of this paper.

Nuclear proliferation can be attempted during used fuel recycling by diverting separated Pu for weapons purposes. However, there are two main methods to ensure that

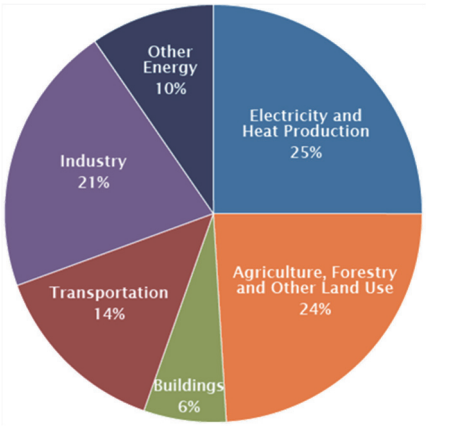

Figure 1. Global greenhouse gas emissions by economic sector in 2010 [2].

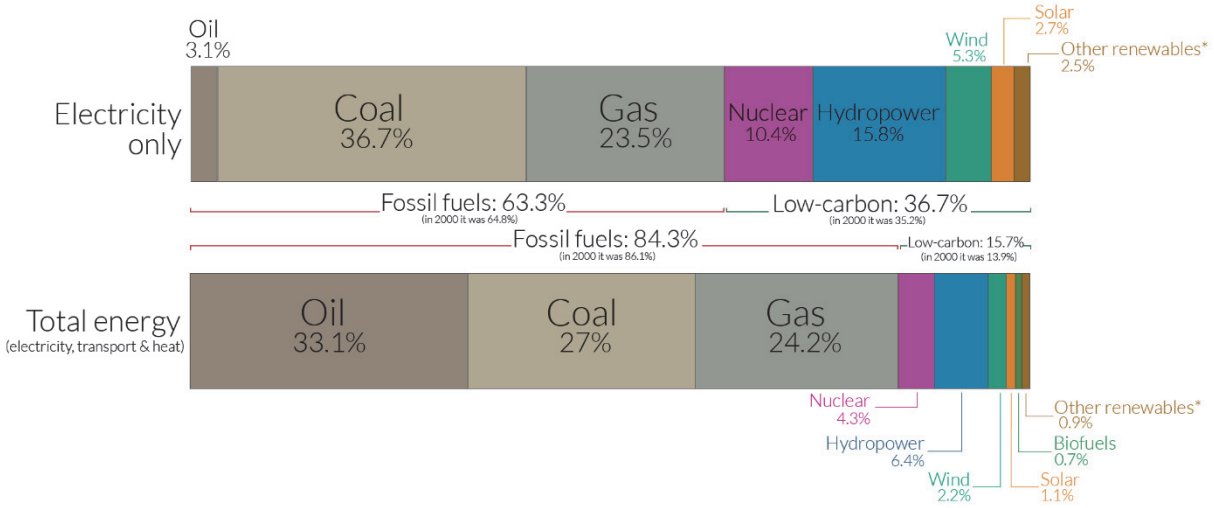

Figure 2. Sources of global energy and electricity production in 2019, including low-carbon sources [3].

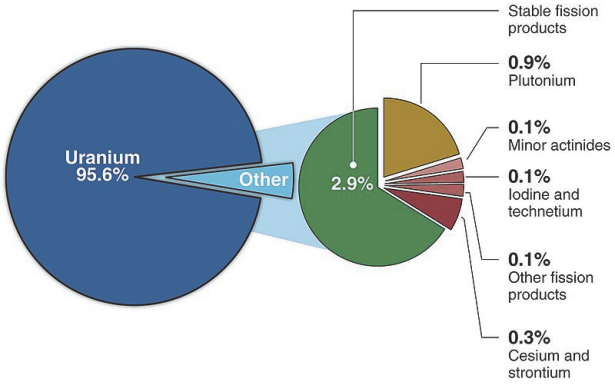

Figure 3. Composition of typical commercial used nuclear fuel [4].

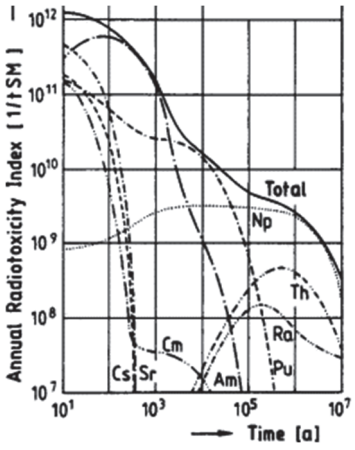

Figure 4. Radiotoxicity of typical commercial used nuclear fuel after discharge from the reactor [5].

nuclear materials are not used for weapons purposes. The first method is full accountancy through international safeguards, implemented by the International Atomic Energy Agency (IAEA) [6]. An alternative approach is to reduce the weapons usability of these materials by mixing them with impurities [7]. However, chemical and physical modifications to nuclear materials have limited potential, since any modification of this type can be undone without significant difficulty [8]. The most robust approach is to denature these materials by adding Pu isotopes that hampers the usability of these materials in weapons. It is known throughout the international community that the large heat generation and spontaneous fission rate of ^{238}Pu are key features in denaturing Pu and thus, making the material effectively not weapons useable at high concentrations [8-11]. This is best seen in the IAEA's assessment that Pu with a ^{238}Pu content of 80% or greater is excluded from international safeguards [6]. While Pu with an isotopic fraction of 80% ^{238}Pu or greater may be impractical to create in used commercial nuclear fuel, recent analysis has shown that this cut-off value for weapons usability is likely much lower. An open source model of a hypothetical nuclear explosive device (HNED) has been shown to be nonfunctional if the heat generation rate of the Pu reaches 0.6 kW, which corresponds to a ^{238}Pu fraction of 9% [12,13]. Other more conservative estimates show this value to be higher at approximately 17% ^{238}Pu [14-16]. Regardless of what estimate is correct, the ^{238}Pu value is likely significantly below 80%.

High burnup fuels naturally produce Pu isotopic compositions that are proliferation resistant. This increased ^{238}Pu content is accomplished through two mechanisms. First, high burnup fuel is either irradiated longer or experiences higher neutron flux. This increased neutron fluence allows more opportunities for the ^{235}U atoms to have multiple neutron absorptions and transmute to ^{238}Pu, as shown in Eq. (1) with corresponding thermal neutron absorption cross section values. Second, high burnup fuels often contain higher U enrichments (greater than 5% ^{235}U) that provides more ^{235}U atoms resulting in higher concentration of ^{238}Pu.

$$^{235}_{92}U \xrightarrow[98.71\ b]{(n,\gamma)} {}^{236}_{92}U \xrightarrow[5.123\ b]{(n,\gamma)} {}^{237}_{92}U \xrightarrow{\beta} {}^{237}_{93}Np \xrightarrow[178.1\ b]{(n,\gamma)} {}^{238}_{94}Np \xrightarrow{\beta} {}^{238}_{94}Pu \xrightarrow[412.8\ b]{(n,\gamma)} {}^{239}_{94}Pu \quad (1)$$

In addition to increased ^{238}Pu content in the used fuel, high burnup will increase concentrations of higher Pu isotopes, such as ^{240}Pu and ^{242}Pu, while suppressing the concentration of the more fissile ^{239}Pu [12]. The critical mass of these higher Pu isotopes are over three times larger than that of ^{239}Pu, thus requiring more Pu in a HNED [17]. The mass of Pu in a HNED is directly proportional to its heat generation. High burnup fuel is also proliferation resistant because the higher neutron fluence allows for more of the Pu that was created in the irradiated fuel to undergo fission, which results in less total Pu mass in the used fuel per MW of electricity generated compared to low and medium burnup fuels. For burnup values of 190 GWd/MTU, the Pu content per MW of electricity generated is half that of current commercial nuclear reactors at 44 GWd/MTU [18]. This reduction in Pu mass per MW of electricity generated will decrease the amount of Pu that will need to be recycled.

Historically recycling U in used fuel has not been especially financially attractive due to the high costs of recycling and the fact that the U isotopic composition in used fuel only contains ~1 wt.% ^{235}U. In addition to this low concentration of ^{235}U, the concentration of ^{236}U is elevated above that of natural U, which has a ^{236}U concentration of zero. ^{236}U is undesirable in nuclear fuel from a reactor physics perspective because of its relatively large thermal radiative capture cross section of 5.123 b and small thermal fission cross section of 0.2594 mb. As seen in Eq. 1, ^{236}U requires three neutron captures before it becomes fissile ^{239}Pu. For these reasons, many consider recycled U from commercial nuclear power plants that use ~4.5% ^{235}U enriched fuel to be of no more value than natural U. This cost benefit calculation may not be true for high assay low enriched uranium (HALEU) fuels that often use enrichments slightly less than 20% ^{235}U. U from used HALEU fuel contains a larger fraction of ^{235}U at ~2 wt.%. In addition to this, HALEU fuel is considered less proliferation resistant than traditional 4.5% enriched fuel among some due to its composition being closer to high enriched uranium (HEU). Highlighting additional proliferation resistance features for reactors that use HALEU fuel may be advantageous to address the concerns of those worried about potential proliferation risk of HALEU fuel.

There are multiple methods to evaluate the attractiveness of Pu from used fuel, but the methodology developed by Bathke et al. is often used due to its simplicity [8]. This method does not consider the total amount of nuclear material available, but instead focuses on the quality of the material for weapons purposes. Only four attributes are considered when determining the attractiveness of material using the Bathke methodology:

1. Bare critical mass (M) in units of kg
2. Heat generation (h) in units of W/kg [16]
3. Spontaneous fission neutron rate (S) in units of neutrons/(s·kg) [19]
4. Radiation dose rate (D) at 1m in units of rad/h

Each of these attributes are normalized by a value that is considered unattractive for weapons purposes. The critical mass attribute is normalized to U at a ^{235}U enrichment of 20%. The heat generation is normalized to Pu with a concentration of 80% ^{238}Pu. The spontaneous fission neutron rate is normalized to reactor grade Pu with a ^{240}Pu concentration of 20%. The radiation dose rate is normalized to a widely considered self-protecting value of 500 rad/hour at 1 m. These four factors are combined together to form a figure of merit (FOM), as shown in Eq. (2) and Eq. (3). FOM_2 includes all four of these factors in the calculation and is considered to be valid for less technically advanced states Eq. (2). FOM_1 excludes the spontaneous fission neutron rate from the calculations because it was deemed by Bathke et al. that technologically advanced states would

have capabilities to mitigate challenges caused by this attribute Eq. (3). To help policy and decision makers more easily categorize the attractiveness of different materials, the FOM values are grouped into four categories, as shown in **Table 1**.

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

$$FOM_1 = 1 - \log_{10}\left(\frac{M}{800} + \frac{Mh}{4500} + \frac{M}{50}\left[\frac{D}{500}\right]^{\frac{1}{\log_{10} 2}}\right) \quad (3)$$

Table 1. FOM categorization ranges for the Bathke et al. material attractiveness analyses method.

FOM	Weapons Utility	Attractiveness
> 2	Preferred	High
1-2	Attractive	Medium
0-1	Unattractive	Low
< 0	Unattractive	Very Low

The Bathke et al. material attractiveness analyses method has been applied to many different types of materials, including fresh, used, doped, high burnup, and reprocessed fuel mixtures [8,18,20,21]. While this methodology is quite flexible due to its simplicity it does have limitations. It cannot account for material composition changes due to enrichment or chemical purification technology. It also cannot consider weapon or facility specific characteristics. Other material attractiveness evaluation methods exist, but they are often more complex or require weapon or facility specific information and thus will be outside the scope of this paper [12,16,22-26].

The HALEU fuel that is being analyzed in this paper is that of Lightbridge Corporation's 19.7% ^{235}U enriched metallic fuel. Lightbridge's U-based fuel is a metallic δ-phase Zr-U alloy consisting of approximately 50 wt.% of each element [27]. Lightbridge Corporation has multiple fuel rod designs in various stages of development. This research utilized the geometry of Lightbridge's four-lobed U-Zr fuel rod design developed for 17x17 pressurized water reactors.

In order to assess the composition of U from the Lightbridge used fuel and to determine the Pu composition from used fuel that utilizes recycled U, computer simulations were performed using the Monte Carlo N-Particle (MCNP) radiation transport code version 6.2 [28] with its embedded CINDER90 fuel burnup and isotopic generation and depletion module. The proprietary geometry and composition of a four-lobe Lightbridge fuel rod with 19.7% ^{235}U enrichment was modeled in MCNP. To simplify the geometry, the helical axial twist was not modeled because internal Lightbridge studies have shown that not including this axial twist has minimal impact on neutronics simulations of the fuel, especially in the single-rod analyses performed in this work. The central displacer was also not modeled as the burnable poison composition of the displacer depends on the needs of the reactor, assembly, and fuel-cycle which the rod is going into. Fuel swelling was also not modeled in the MCNP simulations since this phenomenon is outside the capabilities of MCNP, which requires a fixed geometry. A unit fuel rod cell was modeled that has the standard fuel rod pitch of 1.26 cm and an active fuel height of 366 cm [29-31]. This cell includes the fuel, cladding, and coolant around the fuel rod. The coolant consists of pure water ($^1H_2{}^{16}O$) without any boron or other dissolvable additives. To represent the neutron flux distribution of a fuel rod not on the periphery of a reactor core, the sides of the fuel rod cell were made reflective, while the surfaces of the top and bottom were not reflective. The k-code results of this geometry represent k_∞ values, as opposed to k_{eff}. The fuel was modeled at a uniform temperature of 900K, with the cladding and coolant modeled at 600K. Test simulations showed that slight changes in temperature had statistically no effect on k_∞ or actinide composition in the used fuel. The ENDF/B-VII.1 cross-sections were used for all materials in the MCNP simulations [32]. The light water moderation treatment (S-α,β) was applied to the coolant. The MCNP k-code simulations were performed with 5000 particles per cycle for 550 cycles, with the first 50 cycles being

Figure 5. Diagram of Lightbridge's metallic helical cruciform geometry. (left) Segment of the rod showing the helical twist; (right) cross-section of the U-Zr fuel core, central displacer and cladding. This diagram is not to scale.

excluded from mean k_∞ calculation. These k-code specifications result in k_∞ values with a 1σ statistical uncertainty of approximately ±0.00045. A total of 22 burnup time steps over a duration of 1150 days with each burnup duration time step consisting of 0.1, 0.2, 0.4, 0.5, 0.8, 1, 2, 4, 8, 16, 25, 50, 75, 100, 100, 100, 100, 100, 100, 100, 100, and 167 days. The final burnup of the fuel was 190.9 GWd/MTHM, which corresponds to approximately 21 at.% burnup. The default recommendation of fission product tier 1 was used for all burnup simulations. Modeling of the metallic U-Zr fuel utilized a U isotopic composition of: 0.18% ^{234}U, 19.70% ^{235}U, 0.09% ^{236}U, and 80.03% ^{238}U.

The used U from the Lightbridge fuel was recycled and re-enriched using a separative work unit (SWU) calculation methodology developed by Texas A&M University [33]. This methodology uses a matched-abundance ratio cascade (MARC) model with the multicomponent (four uranium isotopes-^{234}U, ^{235}U, ^{236}U, and ^{238}U) enrichment capability.

2. Results

The U composition from the used Lightbridge fuel can be seen in **Table 2**. This fuel was then re-enriched to 5 wt.% ^{235}U and 19.7 wt.% ^{235}U, for use as traditional UO_2 and metallic fuel respectively. These two fuels were re-burned for 44 GWd/MTU and 146.6 GWd/MTU respectively. The Pu composition in the used fuel of these re-enriched and re-burned fuels can be seen in Table 3 along with typical weapons-grade and reactor-grade Pu compositions.

To determine the materials attractiveness of the Pu from the U in **Table 3**, Eqs. (2) and (3) were used. Values from these equations can be seen in **Table 4** [34]. The bare critical masses (M) of the Pu compositions were determined though k-code MCNP simulations. The heat generation (h) and spontaneous fission neutron emission rates (S) were determined by combining the Pu composition with known nuclear data [16,19]. The radiation dose rate (D) at 1 m was estimated to be negligible based on previous studies of Pu compositions [20].

Table 2. U content of the used Lightbridge fuel (feed material) after being enriched to 5 wt.% and 19.7 wt.% ^{235}U. The tails had 0.3 wt.% ^{235}U for both product calculations.

wt.%	Feed material	5 wt.% product	19.7 wt.% product
^{234}U	0.0685	0.1849	0.7578
^{235}U	1.9840	5.0000	19.7000
^{236}U	3.9757	8.4359	27.2162
^{238}U	93.9718	86.3792	52.3260

3. Discussion

From Table 2 it can be seen that there is approximately twice as much ^{236}U in the once burned used Lightbridge fuel as ^{235}U. This difference becomes less extreme as the ^{235}U content is re-enriched to either 5% or 19.7% due to the slightly greater mass of ^{236}U compared to ^{235}U. The Pu content of the re-burned U from Table 2 is shown in Table 3, along with the composition of weapons-grade and reactor-grade Pu. The Pu content in Table 3 shows that fuel that has had more burnup over its complete life, including recycling and re-enriching, has higher concentrations of ^{238}Pu and lower concentrations of ^{239}Pu. U compositions with higher concentration of ^{236}U will lead to increased fractions of ^{238}Pu in the used fuel. The lower concentration of ^{239}Pu is caused by fewer ^{238}U atoms existing in the U. As seen from Table 2, the re-enriched U composition has a ^{238}U fraction of approximately 50%, which is significantly less than the approximately 95% ^{238}U concentration found in most currently used commercial U fuel.

Intuitively, a lower fraction of ^{239}Pu in Pu might lead readers to believe that this is the dominant factor in material attractiveness for weapons purposes, but the fast fission cross-section for ^{238}Pu (1.968 b) is comparable to that of ^{239}Pu (1.781 b). This means the bare critical mass of the Pu from the used re-enriched metal fuel has a similar value to that of weapons-grade Pu, shown in Table 3, even though their ^{238}Pu and ^{239}Pu contents are very different. However, increased concentrations of ^{238}Pu does negatively affect the materials attractiveness due to an increased heat generation and spontaneous fission rate, as shown in Table 4. These two factors cause lower FOM values for Pu compositions with high fraction of ^{238}Pu,

Table 3. Pu composition of the re-enriched and re-burned fuels along with weapons and reactor-grade Pu.

wt.%	Re-burned used UO_2 fuel	Re-burned used metal fuel	Reactor-grade Pu	Weapons-grade Pu
^{238}Pu	14.19	38.42	2.38	0.00
^{239}Pu	49.95	33.08	53.62	93.60
^{240}Pu	19.05	11.71	23.56	5.90
^{241}Pu	12.40	11.49	14.35	0.40
^{242}Pu	4.40	5.30	6.09	0.10

Table 4. FOM values for four different Pu compositions of interest.

Pu composition	M (kg)	h (W/kg)	S (n/s/kg)	D (rad/h)	FOM_1	FOM_2
Weapons-grade Pu	16.30	2.18	6.20×10^4	~0	2.55	1.75
Reactor-grade Pu	21.21	16.40	4.07×10^5	~0	1.98	0.86
Re-burned UO_2	19.39	81.79	6.38×10^5	~0	1.42	0.66
Re-burned metal	17.44	215.98	1.21×10^6	~0	1.07	0.40

seen in Table 4. For FOM_2, which includes spontaneous fission, all non-weapons-grade Pu compositions have low attractiveness, although it should be noted that the FOM equations (Eq. 2 and 3) are logarithmic, making large differences in negative attributes appear small. For the FOM_1 values, all non-weapons-grade Pu compositions also have medium attractiveness, although the contrast between the different FOM_1 values is even greater with the value for reactor-grade Pu being borderline high attractiveness and the re-burned used metal Pu compositions being borderline low attractiveness. Material attractiveness assessments made by researchers other than Bathke et al. show that both re-burned Pu compositions have a ^{238}Pu concentrations significantly above the 9% threshold [12,13] and the re-burned metal value is significantly above the 17% threshold [14-16]. The material extractives values are not expected to change much with decay times on the order of several years due to the long half-lives of half-life of all Pu isotope of interest, with ^{238}Pu being the fastest decaying Pu isotope at a half-life of 87.7 years.

4. Conclusion

Due to the classified nature of nuclear weapons and the challenges with making definitive absolute statements that a material cannot be directly used as the fissile content in a nuclear weapon, it is impossible to say that any Pu composition is weapons-proof. However, it is clear that recycling HALEU creates fresh fuel with higher concentrations of ^{236}U that will result in larger fractions of ^{238}Pu in the used Pu composition. These larger fractions of ^{238}Pu make the Pu less attractive regardless of which assessment methodology is used. As HALEU fuel becomes increasingly likely for use in some future reactor designs, a discussion of recycling the used fuel should be had for the goals of resource utilization, waste management mitigation, economic improvements, and proliferation resistance. This paper demonstrates the benefits to proliferation resistance by recycling HALEU fuel.

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