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Innovative Fuel Design to Improve Proliferation Management

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Innovative Fuel Design to Improve Proliferation Management

A thesis submitted in partial fulfillment of the requirements for the degree of

Master of Science at Virginia Commonwealth University.

By

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December 2018

ACKNOWLEDGEMENTS

I would first like to thank my parents for their unconditional support. Without them, I doubt I would have attempted coming back to graduate school, much less at VCU. Having them close gave me a strong emotional crutch for whenever I faltered. I would also like to thank my sister and nephew for the countless amount of hours we have been able to enjoy to make up for my time away.

I want to thank Dr. Goddard for being the impetus for coming back to VCU. He showed me a unique perspective, which enticed me to work with him during my studies. He allowed me freedom in my research endeavors, which only encouraged me to strive further. Even though I may be leaving earlier than he desired, it was pleasant learning from him, not only during our research discussions, but from the multiple classes he taught.

I want to thank Dr. Phongikaroon for being the best professor to TA under. The amount of time I was able to participate with the students and teach during the recitations made me want to continuously develop my teaching abilities to better help them during their time as undergrads. I was able to develop many strong relations with the students that I hope to continue into the professional world. I also want to give him a huge appreciation for allowing me an opportunity that has led to my endeavors at North Anna.

I want to thank Dr. Manic for accompanying Dr. Goddard and Dr. Phongikaroon on my thesis committee. This allowed my peace of mind while preparing my defense and for my future outside of VCU.

I want to thank the countless friends I made during my time here as a graduate student. I especially want to thank Eric Depew and Sina Rahini for our long nights of Catan to help unwind. I also want to thank Dr. Brandon Dodd and Michael Cartwright for being the best lab mates I could ask for. You gave me insight into the many different discussions and arguments we had, and you all made the time in the lab much more entertaining.

TABLE OF CONTENTS

LIST OF FIGURES

LIST OF TABLES

ABSTRACT

This research uses an existing innovative fuel design (IFD) that has intrinsic safety features and enhanced economics over the current uranium dioxide (UO₂) light water fuel design and evaluates promising methods to improve the waste management and proliferation resistance of the IFD by doping the fresh fuel with select actinides. The most robust approach for proliferation resistance is to denature these materials by adding a uranium or plutonium isotope that hampers the usability of the materials in weapons. The proposed modifications to the IFD use this approach through elevated fractions of 238 Pu. 238 Pu generates large quantities of heat and neutrons through its radioactive decay and is estimated to make plutonium potentially "proliferation-proof." The IFD this work uses as a foundation is an advanced metallic fuel designed for use in current light water reactors. Due to the high fission density of metallic fuel and the proposed uranium enrichments, the plutonium produced by irradiating this fuel has promising isotopic content for proliferation resistance. This proliferation resistance will be further increased by adding ²³⁷Np and/or ²⁴¹Am to the initial fresh fuel composition that will result in increased ²³⁸Pu content. Adding these actinides into the fresh fuel at 0.2 wt.%, the amount of 238 Pu produced in the used fuel can be used for proliferation resistance. Increasing the actinide wt.% can potentially produce "proliferation-proof" used fuel. Also, by utilizing neptunium and americium in fresh fuel, many of the challenges with permanent geological disposal of used fuel can be mitigated.

CHAPTER 1: MOTIVATION AND OBJECTIVES

1.1 Motivation

Nuclear energy is an ever-growing market where small developments in technology can vastly improve the energy demands of a large and constantly consuming world. Whether these advancements are in the manufacturing process, the energy production, or the delivery of the energy, any of these steps can help bring power to the populace. While typically seen as beneficial to the world, nuclear energy also brings some negative consequences along with it. Radiation, environmental concerns, and nuclear weapons are only a small sample of some issues the nuclear community attempts to combat. This research will look specifically at attempting to produce a new fuel design that will not only benefit the energy side of the equation, but also combat the nuclear weapons issue that comes with fuel after it's been burned, as well as the waste management issues that arise with storing used fuel.

There are two main methods to ensure that nuclear materials are not used for weapons purposes. The first method is to monitor and assay these materials to ensure they are fully accounted for, which is being implemented under the international safeguards regime by the International Atomic Energy Agency (IAEA) pursuant to the Treaty on the Non-Proliferation of Nuclear Weapons.¹ An alternative approach is to reduce the weapons usability of these materials by mixing them with impurities, which could alleviate efforts required on the part of the IAEA.² However, any chemical and physical modifications to nuclear materials have limited potential, since any modification of this type can be undone without significant difficulty.³

An innovative fuel design (IFD) is proposed that follows a denaturing approach, adding elevated fractions of 238 Pu or 238 Pu producing isotopes. By adding these isotopes, the plutonium spent fuel vector can be altered to improve its proliferation resistance. 238 Pu generates large quantities of heat and neutrons through its radioactivity decay (half-life of 87.7 years) and is estimated to make plutonium "proliferation-proof" at concentrations as little as 9%-18%, depending on the hypothetical weapon model.^{4,5,6}

1.2 Goal

The primary goal of this thesis is to develop an IFD that can be implemented into commercial nuclear reactors and, after being burned inside the reactor, will be undesirable as a fuel for nuclear weapons. Specifically, this IFD needs to have a large enough ²³⁸Pu fraction that it generates enough heat to either melt or damage the high explosives in nuclear weapons. This IFD also needs to be functional as a fuel source. This requires it to not be detrimental to energy production in a nuclear reactor as well as maintaining the safety standards that are consistent within the nuclear community.

1.3 Approach

In order to create this IFD and test its viability as a fuel and its proliferation resistance the following objectives have been created to prove the capability of the fuel:

Create a radiation transport model of the IFD

Model the burnup of a singular IFD fuel rod with the chosen actinide poisons

- Create a Pressurized Water Reactor (PWR) assembly with IFD fuel rods
- Model the burnup of a singular IFD assembly with the chosen actinide poisons
- Create a PWR core utilizing the IFD assemblies
- Model the burnup of an entire core with IFD assemblies
- Analyze the core characteristics

With each step, the goal is to determine the effects that the actinides have on the fuel, mostly focusing on the plutonium vectors after the fuel has been burned. With the later stages, power density and other core parameters need to be monitored to ensure this fuel won't be harmful to energy production, while still maintaining the precedent set in the beginning objectives.

1.4 Organization of the Dissertation

This thesis is composed of 5 chapters. Chapter 2 will discuss the background of the thesis. This details the background of nuclear energy, nuclear fuel, radiation, and how these topics influence the design of a new fuel type. Lightbridge fuel is also discussed as their proposed fuel idea is an influence for the IFD. MCNP is also reviewed as it is the main program used for evaluating the IFD. Chapter 3 discusses in depth the details about Lightbridge fuel, as well as the step-by-step process in developing the IFD in MCNP. This chapter also details how ²³⁸Pu is created while the IFD is burned inside a reactor, as well as how this burn process works inside MCNP.

Chapter 4 shows the results of the burn code on the IFD created in Chapter 3. Multiple graphs of each simulation are shown and compared against each other to show how the plutonium vector changes with weight percent (wt. %) of the denaturing nuclide. The mass of total plutonium in the used fuel for each denaturing nuclide is compared against the total mass of plutonium for $UO₂$ used fuel. Chapter 4 also details the self-sustainability of the IFD and $UO₂$ fuel for reprocessing purposes. Chapter 5 summarizes the entire thesis, drawing conclusions from the data in the previous chapter. It also discusses potential future steps required to further this research. Additional data for the burn simulations, MCNP input, and UO₂ fuel data are all contained in the Appendix.

CHAPTER 2: BACKGROUND

2.1 Nuclear Energy

Nuclear energy makes up roughly 20% of the energy production in the United States⁷. In order to actually produce this, nuclear reactors across the country operate in 18 month (540 day) cycles. Nuclear reactors use a fissile isotope, generally ²³⁵U, in order to fission. This fission process generates heat inside the primary side of the plant. This heat is transferred into the water contained inside the primary, which travels through the steam generator, which transfers its energy into the water on the secondary side of the plant. This causes the water to boil and create steam. The steam travels through the secondary side into a turbine, where it transfers its energy into the turbine, causing it to generate electricity.

The main process behind the energy generation is the fissioning of the fuel. In most cases, the primary fuel type is ^{235}U ; however, ^{233}U and ^{239}Pu have been used as the fuel in nuclear reactors. All three nuclides are fissile, which means they can sustain a chain reaction on thermal neutrons, so long as you have adequate fissile material to maintain the reaction.⁸ This process gives off energy based on the binding energy curve as seen in Figure 1.

Figure 2.1: Binding Energy Curve⁹

Binding energy refers to the amount of energy required to break a nucleus into its protons and neutrons.¹⁰ This graph shows how heavier isotopes (²³⁵U) give off energy when it is fissioned. This amount of energy is small compared to fusing lighter elements together, but when large amounts of fissile material are split, large amounts of energy are also released. Fissile nuclides also generate two or more neutrons on average upon fission. This enables the chain reaction to be self-sustaining.

2.2 Nuclear Fuel

To actually generate heat in the primary side of the reactor, the fuel must be properly manufactured into a fuel rod, so that it fissions and generates heat, but will not contaminate the

coolant with severely hazardous radioactive nuclides. In a typical pressurized water reactor (PWR), uranium is used as an oxide fuel. The uranium is sintered and pressed into small ceramic pellets, which are then put inside a metal rod (cladding), and backfilled with helium gas. The uranium is not solely ²³⁵U. When it is originally mined, the uranium is 99.275% ²³⁸U and 0.720% 235U.¹¹ To make this material usable in light water reactors, the uranium must be enriched. In the United States, enrichment is limited to 5% for commercial operations.¹² The metal rod is a zirconium alloy, which has been fabricated to maximize its heat transfer capabilities, while reducing its neutron absorption cross-sections. It is also corrosion resistant and maintains good structural properties, making it widely used as cladding in PWR's. These fuel rods are bundled together into a fuel assembly. These assemblies will then be arranged into the full core. Depending on the type of reactor design, the size and shape of the fuel rods and assembly will differ. In a PWR, the fuel rods are circular rods which are arranged into a 17x17 square array, where 25 of the slots are filled with control rods, instead of fuel rods. These assemblies are arranged into a 15x15 grid, approximating a circular shape inside the core. The designs of the fuel rods, assemblies, core, and parameters are seen in the Figures 2-5.¹³

Figure 2.2: Fuel Pellets¹³

Figure 2.3: PWR Fuel Assembly¹³

Figure 2.4: Cross-Section of PWR Core¹³

Figure 2.5: Fuel Rod Parameters¹³

Fuel rods used in commercial reactors are made with ceramic fuel. There are also metallic fuel rods that can be used instead. These metallic fuel rods have the advantage of a greater heat conductivity than the ceramic fuel, which allows for a lower operating centerline temperature of the fuel. This makes the fuel a potentially safer option during normal operations. The downside of metallic fuels is that they cannot withstand as high a temperature due to their lower melting temperature. Metallic fuels also swell much more than ceramic fuels due to buildup of fission product gases inside the fuel. Metallic fuels don't need to be solely the isotope being used as fuel – they are typically alloyed to improve the heat conductivity, while maintaining the high fission density.

2.3 Radiation

The fuel is radioactive, which means it emits radiation. The main focus for this research is the radiation due to alpha particle emission. Alpha emitters, such as 238 Pu generate large quantities of heat. This will be important once the fuel is ejected from the core. Gamma emission is also important for safety concerns, as gamma rays are highly penetrating and cause tissue damage in people. Because the fuel is radioactive, it will decay into its constituent isotopes based on its decay chain, as well as constantly being fissioned while the reactor is in operation. The decay chain of an isotope is modeled based on its probability of decaying per any of the modes of decay. An example of the 238 U decay chain can be seen in Figure 6.

Figure 2.6: ²³⁸U Decay Chain¹⁴

Some isotopes have multiple ways of decaying. This is due to having cross sections that allow the isotope to absorb neutrons. Once it absorbs a neutron, it follows a different decay series. This typically only happens in a neutron-rich environment (nuclear reactor). For isotopes that have a high neutron capture cross-section, meaning that the probability of absorbing a neutron is higher, then the isotopes are more likely to follow a different decay series. Figure 7 shows how the capture cross-section of ²³⁸U is much greater than the fission cross-section at energies below 1 MeV. For commercial reactors, which operate at thermal energies (0.025 eV), the capture cross section for ²³⁸U dominates.

Figure 2.7: ²³⁸U Neutron Capture and Fission Cross-Section¹¹

Once the 238 U atom absorbs a neutron, shown in equation 1, it then follows a new decay series.¹⁵

$$
{}_{92}^{238}U + {}_{0}^{1}n \rightarrow {}_{92}^{239}U \stackrel{\beta-}{\rightarrow} {}_{93}^{239}Np \stackrel{\beta-}{\rightarrow} {}_{94}^{239}Pu
$$
 (1)

The main focus with this research will be the decay series of certain actinides that decay into ²³⁸Pu. As stated before, ²³⁸Pu is an alpha emitter, which in turn generates a large amount of heat. Once the fuel is removed from the reactor, the heat is still generated within the plutonium until the ²³⁸Pu decays away; however, its half-life is 87.7 years¹¹. The fuel needs to be constantly cooled to prevent decay heat from causing fires in the used fuel. Combined with the radiation that is being emitted from not only the plutonium but the other actinides and radioactive isotopes in the fuel, this makes handling of the used fuel dangerous.

2.4 Safeguards and Nonproliferation

A potential use of used fuel is for nuclear weapons. Nuclear weapons also use fissile material to take advantage of the large energy outputs from fissioning these materials. Because nuclear reactors need to have ²³⁵U, a fissile isotope, in order to operate, and produce ²³⁹Pu, another fissile isotope, the used fuel is seen as a potential source of nuclear material for nuclear weapons. This capability of nuclear weapons requires safeguards to be placed on used fuel. Safeguards are a set of technical measures applied by the IAEA on nuclear material and activities, through which the Agency seeks to independently verify that nuclear facilities are not misused and nuclear material not diverted from peaceful uses. States accept these measures through the

conclusion of safeguards agreements.¹ The IAEA is the international agency that regulates international safeguards around the world. Domestic agencies, such as the United States Nuclear Regulatory Commission regulate domestic safeguards. All nuclear reactors are placed under safeguards, as well as the used fuel; however, if it was possible to denature the used fuel – making it unsuitable for nuclear weapons, it might be possible to relax or remove safeguards from this material, saving money and improving global security. Through the use of safeguards, the risk of proliferation is greatly reduced, but there is still the possibility of nuclear material being used for non-peaceful purposes. This would allow for more resources to focus on safeguarding the front end of the fuel cycle.

2.5 Lightbridge

Currently, most commercial reactors use ceramic fuel for their fuel rods. Lightbridge, a nuclear energy company known for its proliferation resistant fuel technology, has proposed a new metallic fuel design that is intended to replace current ceramic fuel in PWRs while reducing proliferation concerns with used fuel, improving fuel economics, reactor safety, and waste management. In Figures 8 and 9, Lightbridge shows that the amount of plutonium produced during a normal fuel cycle is much less than a typical ceramic fuel assembly as well as the amount of ²³⁸Pu is greater while the ²³⁹Pu is lower.

Figure 2.8: Plutonium Concentration per Assembly over Time¹⁶

Figure 2.9: Plutonium Concentration per Fuel Rod at Discharge¹⁶

Lightbridge's metal fuel is a Zr-U alloy of U-50Zr. This alloy is used to decrease the swelling due to fission fragments inside the fuel. Other than the typical benefits of metallic fuel, Lightbridge's fuel also has a much larger surface area due to having its unique cruciform shape, (35-40% greater), allowing for greater heat transfer to the coolant. This not only minimizes the operating temperature, it also lowers the amount of heat that must be dissipated into the coolant during reactor shutdowns. This adds to the safety of the overall design. The fuel is metallurgically bonded to the Zircaloy-4 cladding, which helps retain radioactive material. Another major difference for this fuel design is the enrichment. Lightbridge states that the fuel is minimally enriched to 13% ²³⁵U, but can reach a maximum of 19.7% ²³⁵U.¹⁶ This will require additional testing and confirmation by regulations in order to be used in a commercial facility. Currently, Lightbridge is seeking testing of its fuel to ensure that it will operate and function as expected.

CHAPTER 3: METHODOLOGY

For this research, Monte Carlo N-Particle Radiation Transport Code (MCNP) was used as the main analysis tool. MCNP is a general-purpose Monte Carlo code that can be used for neutron, photon, electron, or coupled neutron/photon/electron transport.¹⁷ MCNP can be utilized to create a 3D space in which the transport equation will detail. This can extend to fuel and reactor design. MCNP has two built in codes that will be beneficial to this research: kcode, and burn code. kcode will generate a keff value, which is a measure of the criticality. This can be used to measure different designs against each other. Burn code uses time steps to determine how the material composition of a space will change over time due to transport events happening. MCNP also contains all of the specific cross-sectional data to correctly identify how each isotope within the space will fission and decay as it is burned inside the reactor. The versatility of MCNP makes it a crucial tool for performing this research.

The card below is an example of the kcode used in this research. Each line tells MCNP how to interpret the text, and what it should do to calculate the keff. The first line is commented out to give a heading to the K Card section. The second line, MODE, tells MCNP which mode to operate kcode. In this case, only neutrons (n) were used. The third line tells MCNP to run the kcode, how to run it, and for how long. KCODE is the input for running kcode, and the four numbers following tell it how long. The first number is how many particles are run in a cycle; the second number is a guess at the k_{eff} that it should achieve; the third number is how many cycles are discarded in the total summation of the k_{eff} ; and the last number is how many cycles are run

in total to accurately determine the keff. The larger the amount of particles and cycles will greatly increase computation time, but leads to a greater precision. The final line tells MCNP where to initiate the source for calculation. This is expressed in *x1, y1,* and *z¹* values after KSRC. Multiple source locations can be expressed.

C K Card MODE N KCODE 1000 1.36 10 400 \$# particles per cycle, guess, thrown, cycles KSRC -0.15 0 190 0.15 0 190 0 -0.15 190 0 0.15 190

An example of the burn card used is seen below. This determines exactly how the materials were burned. The first line is commented out to give a heading to the burn card. In the second line, the entire burn card is denoted with BURN. This tells CINDER, a depletion code within MCNP, that the following inputs will define how to burn the material. The first input is TIME, which describes at what time steps, in days, to burn the materials. In this case, the fuel was burned at 0.3 days, again 0.7 days later, 2 days later, and so on. Smaller time steps at the beginning of the burn cycle develop the total inventory of nuclides. This time step can be lengthened later in the cycle because the rate of change in nuclide concentration is smaller. The second input is MAT, or material. This denotes which materials undergo the burn. Theoretically all materials will undergo the burn, but only the materials needed for evaluation should be put here to lower computation time. Power is the next input. This is listed in megawatts (MW), and should be representative of the power of whatever is being burned. Even though the fuel will be placed inside a reactor, it's important to determine the power per a singular fuel power for this case instead of total power of the reactor. The next input is PFRAC, or power fraction. This is the fraction of total power per time step. In this case, it was assumed that the reactor operated at 100% power for each time step. This value extends from 0 to 1. AFMIN, or atomic fraction is the value below which an isotope will no longer be tracked. The second value after AFMIN denotes the convergence criteria. BOPT is the last input used in this burn cade. This input has three different uses in the burn code. The first value is a Q value multiplier, which was not used in this case. The second value determines the ordering and output of the data. If the value is positive, the data is printed at the end of each step, whereas it would only be printed at the end if it were positive. The first digit determines how many tiers of fission products are shown in the data. 0 corresponds to tier 1, 1 with tier 2, and 2 with tier 3. The second digit determines the ordering of the data. The 4 orders the data in correspondence with increasing atomic number and mass number. The last value tells CINDER whether to use cross-section models to calculate 1-group cross-sections or not. 1 allows CINDER to do so, where a 0 would not.

```
C Burn Card
BURN TIME = 0.3 0.7 2 7 20 40 80 100 100 100 100
             100 100 100 100 100 100 $1 full cycles
     MAT = 1 2 POWER = 0.1310667 $MW
       PFRAC = 1.0 1.0 1.0 1.0 1.0 1.0 1.0 1.0 1.0 1.0 1.0
              1.0 1.0 1.0 1.0 1.0 1.0
       AFMIN = 1e-10 1e-10
      BOPT = 1.0 -24 1
```
The IFD approach is founded on utilizing an advanced metallic fuel design for applications in commercial PWRs, which uses a unique cruciform shape. Each fuel rod consists of – a central displacer, which is a zirconium alloy that houses burnable poisons; a fuel, which is a 50 wt.% Zr-U alloy¹⁸; and cladding, which is Zircaloy-4, all metallurgically bonded to one another during fabrication. The IFD has a helical twist throughout the length of the fuel rod to promote cross flow of the coolant, as well as reduce thermal concentrations along the edges of the cladding.¹⁶ The fuel geometry which this research is based on is shown in Figure 3.1.

Figure 3.1: IFD a) Axial rod showing helical twist b) Cross section showing the displacer, fuel core, and cladding¹⁶

This IFD will be enriched to 19.7% ²³⁵U, and has a proposed burnup of 190 GWd/MTU.¹⁶ The high fission density of metallic fuel and the proposed uranium enrichments will produce plutonium with promising isotopic concentration such that the irradiated IFD, even after separation, will be proliferation resistant. This proliferation resistance can be further increased by adding ²³⁷Np and/or ²⁴¹Am to the initial fresh fuel composition that will result in increased ²³⁸Pu content. These fuels were chosen because of their decay series to create additional ²³⁸Pu.

 238 Pu is an ideal choice to choose as a denaturing actinide. Because not all of the fuel is burned while inside the reactor, having more ²³⁸Pu in the fresh fuel will produce more after being burned. ²³⁷Np and ²⁴¹Am aren't as straightforward. Both of these actinides have a decay chain that needs to be followed to produce ²³⁸Pu. ²³⁷Np can absorb a neutron and will beta decay into ²³⁸Pu, shown in equation 2.

$$
{}_{93}^{237}\text{Np} + {}_{0}^{1}\text{n} \rightarrow {}_{93}^{238}\text{Np} \stackrel{\beta-}{\rightarrow} {}_{94}^{238}\text{Pu}
$$
 (2)

 241 Am can absorb a neutron, which turns it into 242 Am. It can then alpha decay into 238 Pu, shown in equation 3.

$$
{}_{95}^{241}\text{Am} + {}_{0}^{1}\text{n} \rightarrow {}_{95}^{242}\text{Am} \stackrel{\beta-}{\rightarrow} {}_{96}^{242}\text{Cm} \stackrel{\alpha}{\rightarrow} {}_{94}^{238}\text{Pu} + {}_{2}^{4}\text{He}
$$
 (3)

Burnup is the measure of how much energy is extracted from the fuel. The burnup is required to accurately determine how much of the fuel is consumed during operations, and is typically written as energy multiplied by time per mass of the fissile material.¹⁹

$$
Burnup = \frac{Power(GW) * time (days) * Capacity Factor}{mass(MTU)}
$$
 (4)

The IFD is to replace typical ceramic fuel in a PWR. As such, the power should not change very much (~3400 MWt). For capacity factor, it is assumed to be at 100% to idealize what the maximum burnup could be. This may not always be true, but this will generate the maximum plutonium vectors possible. Using the calculated mass and the equation for burnup seen in equation 4, this burnup should match the proposed burnup.

To accurately determine the plutonium vector of a freshly burned fuel rod, a model was created using MCNP. The actual dimensions of the IFD individual fuel rods are proprietary information, however the burnup, fuel pitch, as well as the k_{eff} , were considered for the IFD based on current PWR fuel rod designs.¹³ Using this knowledge, a PWR fuel rod was modeled in MCNP. The process for creating the PWR rod required using typical PWR dimensions, which were shown in Figure 2.5; however, not everything was modeled about the PWR rod. For example, a typical PWR rod contains a spring that compresses the fuel pellets; however, this was not modeled. The fuel rods are also capped, but for ease of assumption, the models had fuel from the base of the rod to the top. PWR fuel rods also undergo swelling of the fuel pellets, but only fresh fuel was modeled. The dimensions of the PWR rod are listed in Table 3.1.

MCNP's kcode was used to determine the initial k_{eff} value of the fuel rod, and the MCNP burn code was used to determine the plutonium vector of the rod. An initial value of 1.3651 was

found for keff. After determining the initial constraints from the PWR fuel rod, the IFD model was created.

Because the dimensions were not known, matching the k_{eff} values took many kcode runs and changes of the geometry. The points of the IFD extended to the edge of the pitch, but the curvature of the fillets between the points had to be constantly changed, while keeping the thickness of the cladding constant throughout. The thickness at the points of the IFD are 1 in. thick while the fillets are 0.5 in. thick. The transition from 1 in. to 0.5 in. is an assumption. The displacer is centered in the IFD, but the size also changed with each evolution in determining keff. The same assumption that the fuel extended the entire length of the rod was used for the IFD. The dimensions of the IFD can be seen in Table 3.2. These dimensions were broken down into sections that corresponded with the surface created within MCNP. This allowed for easier calculations of the area and volume.

Table 3.2: Area and Volume for IFD Fuel Rod

The dimensions were modified until the keff value matched that of the PWR fuel rod. As can be seen in the MCNP cross sections in Figure 3.2, both fuel rods fit within the same pitch, and both cases assumed reflective boundary conditions along the edges.

Figure 3.2: MCNP cross-sectional view of the a) PWR fuel rod, b) IFD fuel rod

CHAPTER 4: RESULTS

The PWR-UO₂ plutonium vector, shown in Table 4.1, is the output of typical used fuel from a PWR. These vectors are not conducive for nonproliferation. The IFD vector is much closer to the potential "proliferation-proof" vector, making it proliferation resistant, but it is still below the 18.1% ²³⁸Pu limit, making it less than potentially "proliferation-proof".

Isotope	"Proliferation- proof" ⁶	$PWR - UO2$	IFD
238Pu	18.1%	2.3%	14.2%
239 Pu	35.7%	56.8%	35.5%
240 Pu	21.1%	21.7%	19.7%
241 Pu	13.5%	14.3%	14.6%
242 Pu	11.6%	4.9%	16.0%

Table 4.1: Plutonium Vector Comparison

In order to reach the 18.1% ²³⁸Pu threshold, the IFD can be denatured with different actinides, namely 237 Np, 238 Pu, and 241 Am. Each actinide was added to the fresh IFD fuel at a concentration of 1.0 wt.% total fuel mass. This is used as a proof of concept to determine if these nuclides would sufficiently enhance the proliferation resistance of the IFD. Each denatured IFD rod was burned for two 18-month fuel cycles at a total burnup of approximately 190 GWd/MTU.¹⁶ This burnup corresponds with the initial burnup proposed for this fuel design. Each case, shown

in Table 4.2, has a large increase in the wt.% of 238 Pu. This follows with the initial assumption that by denaturing the fresh fuel, the proliferation resistance could be improved upon.

For each doping actinide shown in Table 4.2, the addition of 1.0 wt.% more than doubled the amount of ²³⁸Pu than in the reference case, and exceeded the 18.1% ²³⁸Pu threshold as well. This shows that it is possible to denature the fuel by adding these actinides before burning. It's important to note that at 1.0 wt.% of the fuel, ~15.9 g of an actinide per fuel rod would be required to reach these values, making it difficult to obtain.

Isotope	Reference	$238p_{11}$	237 Np	241 Am
238p _U	14.2%	32.9%	35.3%	30.9%
239 Pu	35.5%	30.9%	30.2%	30.2%
240P _U	19.7%	14.8%	14.1%	14.5%
241 Pu	14.6%	12.4%	11.7%	12.0%
242P _U	16.0%	9.0%	8.6%	12.4%

Table 4.2: Plutonium vector of the IFD with added nuclide concentrations at 1.0 wt.%

Additional MCNP simulations were performed to reach the 18.1% ²³⁸Pu threshold values, thereby limiting the amount of each actinide needed to be doped. The full plutonium vectors of these simulations can be seen in the appendix. Figures 4.1-4.5 detail how the plutonium concentrations change based on wt.%, from 0 wt.% to 1.0 wt.% as well as how the values for 238Pu and ²³⁹Pu change per actinide.

Figure 4.1: Plutonium Vector of the Used Fuel with ²³⁸Pu Doping in the Fresh Fuel

Figure 4.2: Plutonium Vector of the Used Fuel with ²³⁷Np Doping in the Fresh Fuel

Figure 4.3: Plutonium Vector of the Used Fuel with ²⁴¹Am Doping in the Fresh Fuel

Figure 4.4: ²³⁸Pu Percentage of the Used Fuel per Doping Actinide in the Fresh Fuel

Figure 4.5: Amount of Plutonium in Used Fuel per Doping Actinide

It's important to realize that in Figures 4.1-4.5, the 238 Pu concentration increases as the 239 Pu concentration decreases with an increase in wt.% of each actinide. This shows that an increase with each actinide makes the used fuel increasingly proliferation resistant. This trend will continue to improve until it reaches a limiting value, but these values greatly exceed what is needed base on the 18.1% ²³⁸Pu threshold value. It's also important to note that ²⁴¹Pu concentration also decreases. While it isn't as important as ²³⁹Pu or ²³⁵U, it is still a fissile isotope, which could be useful in some weapons uses. Figure 4.4 shows that each actinide reaches the 18.1% ²³⁸Pu threshold value around 0.2 wt.%, meaning that it only takes around 3.1 g of a singular actinide per fuel rod to achieve proliferation resistance.

If each fresh rod contains 3.1 g additional actinide material at an initial loading of 0.2 wt.%, and a typical PWR core contains approximately 51,000 rods, this corresponds to 160 kg of doping material being loaded into a fuel core. This is a large amount of actinide required for fresh fuel, which is difficult to obtain. The only ways to obtain these actinides are through an isotope production reactor or through reprocessing. The question becomes, is the addition of these actinides self-sustaining – does the used fuel have 3.1 g of the actinide that was added into the fresh fuel? If so, it becomes possible to reprocess this used fuel to obtain the actinides needed to keep the fuel production ongoing. Table 4.3 shows the used fuel masses for each actinide for 0.2 wt.% doping, and Table 4.4 shows the used fuel masses of each actinide for the ceramic fuel case and the non-doped IFD case.

Actinide	Initial Mass (g)	Final Mass (g)
238P _U	3.162	2.769
237 Np	3.162	3.116
241 Am	3.160	0.040

Table 4.3: Initial Mass vs Used Fuel Mass for the Actinide Doped into the Fresh Fuel at 0.2 wt.%

Isotope	$UO2$ Mass (g)	IFD mass (g)
238P _U	0.514	1.908
237 Np	1.274	2.684
241 Am	0.095	0.026

Table 4.4: Used Fuel Mass for each Actinide for Ceramic and Non-Doped IFD

For both the ²³⁷Np and ²³⁸Pu cases, the mass of the actinide remaining in the used fuel is close to the initial mass doped into the fresh fuel - the neptunium case being the most valuable, with it being 0.046 g below the fresh loading. In both the $UO₂$ and Non-Doped IFD case, the amount of each actinide in the used fuel are over 1 g from the initial loading needed for the IFD, except for neptunium, but this is less than the ²³⁷Np doped IFD case.

CHAPTER 5: CONCLUSION

Summary

The results of the burnup calculations for a singular IFD fuel pin show that it's plausible for this design to be proliferation-resistant, if not potentially "proliferation-proof". The increased levels of 238 Pu (Table 4.1) in the used IFD fuel are capable of generating enough heat to classify the fuel as proliferation-resistant; however, the goal is to make it potentially "proliferationproof". The analyses of denaturing the fresh IFD fuel shows that it is possible to create enough 238 Pu within the used fuel that the plutonium vector exceeds the 18.1% 238 Pu threshold. The challenge lies in obtaining these denaturing nuclides to mix into the fresh fuel, as large amounts are not currently readily available and would require reprocessing to collect them. This will further increase resource utilization and create proliferation-resistant plutonium in the fuel at potentially any burnup level. Utilization of plutonium and ²⁴¹Am will also have a significant positive impact on permanent geological disposal waste management as these are the primary heat producing nuclides after 300 years, but more insight will be required for a full conclusion on waste management. Realizing a potentially "proliferation-proof" plutonium in used fuel will also have a positive impact on reprocessing. Proliferation concerns are one of the primary factors limiting the use of reprocessing to address high-level waste management. This analysis was a starting point to prove that this new fuel type was capable of proliferation-resistance, and with the positive results, further analyses should be conducted.

33

Future Work

The future work should involve developing an assembly MCNP PWR model based on the IFD fuel. The plutonium isotopic content in the used IFD fuel from simulation analyses should be compared to typical PWR used fuel, and the proliferation-resistance effectiveness evaluated using different analysis methods.^{3,20,21} In order to enhance the proliferation-resistance capability of the IFD fuel, the addition of ²³⁷Np and/or ²⁴¹Am in the fresh fuel should be assessed in depth with the focus on the plutonium vector in the used fuel. Various conventional reactor performance characteristics (such as flux peaking factors, core reactivity, fuel pin wised power distribution, fuel and moderator coefficients, etc.) should be monitored for each modified fuel composition. Simulations should be performed to evaluate the impact of introducing separated neptunium, plutonium, or americium into the fresh fuel as a mixed metallic fuel, e.g. U-Pu-Zr.

Acknowledgements and Notes

I also would like to thank the United States Nuclear Regulatory Commission for partially supporting this work (NRC-HQ-84-14-G-0051). It should be noted that this work is not sponsored by the Lightbridge Cooperation and that all designs and calculations are based on open source information and best estimate assumptions.

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APPENDIX

²³⁸Pu Actinide Data

²³⁷Np Actinide Data

²⁴¹Am Actinide Data

Non-Doped IFD Data

Ceramic Fuel Data

Ceramic Fuel MCNP Input Deck

```
C Fuel Rod
C = =C Cell Card
1 1 -10.97 -62 1 -2 VOL=183.6321 u=0 imp:n=1 $Pellet
2 2 -0.000164 -63 62 1 -2 VOL=7.4245 u=0 imp:n=1 $Gap
3 3 -6.4987 -64 63 1 -2 Vol=58.5413 u=0 imp:n=1 $Cladding
C
4 4 -0.717 1 -2 -47 48 -49 50 64
      VOL=353.6901 u=0 imp:n=1 $Water
C
5 0 (47:-48:49:-50:-1:2) u=0 imp:n=0 $Void
C = =C Surface Card
1 PZ 0 $Bottom
2 PZ 380 $Top
\mathsf C*47 PX 0.63 $RightBoundary
*48 PX -0.63 $LeftBoundary
*49 PY 0.63 $TopBoundary
*50 PY -0.63 $BotBoundary
\mathcal{C}61 CZ 50 $Void
```
62 CZ 0.3922 \$FuelPellet 63 CZ 0.40005 \$FuelGap 64 CZ 0.45725 \$FuelCladding C == C K Card MODE N KCODE 1000 1.36 10 400 \$# particles per cycle, guess, thrown, cycles KSRC -0.15 0 190 0.15 0 190 0 -0.15 190 0 0.15 190 C == C Burn Card BURN TIME = 0.3 0.7 2 7 20 40 80 100 100 100 100 100 100 100 100 100 100 \$1 full cycles $MAT = 1$ POWER= 0.0735709 \$MW PFRAC = 1.0 1.0 1.0 1.0 1.0 1.0 1.0 1.0 1.0 1.0 1.0 1.0 1.0 1.0 1.0 1.0 1.0 AFMIN = 1e-10 1e-10 $BOPT = 1.0 -24 1$ MPHYS ON C == C Material Card M1 92238 -.841833 92235 -.039668 90228 -1e-37 90229 -1e-37 90230 -1e-37 90232 -1e-37 91231 -1e-37 93235 -1e-37 93236 -1e-37 93237 -1e-37 93238 -1e-37 93239 -1e-37 94238 -1e-37 94239 -1e-37 94240 -1e-37 94241 -1e-37 94242 -1e-37 95241 -1e-37 95242 -1e-37 95243 -1e-37 98252 -1e-37 $nlib = .83c$ \$Fuel PU M2 02004.82c -1 \$Gap M3 40090.81c -.508326 40091.81c -.110854 40092.81c -.169442 40094.81c -.171714 40096.81c -.027664 50118.81c -.012000 \$Clad M4 01001.81c .67 08016.81c .33 \$Water MT4 lwtr.16t \$Light Water

 C

M5 02004.82c -1 \$ControlRod

IFD MCNP Input Deck

```
C Fuel Rod
C ======================================================================
C Cell Card
1 1 -6.5075 -3 -4 -5 -6 1 -2 VOL=11.9130 u=0 imp:n=1 $Displacer
\overline{C}2 2 -9.7012 -23 21 1 -2 VOL=11.9381 u=0 imp:n=1 $TopFuelCircle
3 2 -9.7012 -24 -22 1 -2 VOL=11.9381 u=0 imp:n=1 $BottomFuelCircle
4 2 -9.7012 -25 19 1 -2 VOL=11.9381 u=0 imp:n=1 $RightFuelCircle
5 2 -9.7012 -26 -20 1 -2 VOL=11.9381 u=0 imp:n=1 $LeftFuelCircle
C
6 2 -9.7012 15 16 17 18 -19 20 -21 22
             1 -2 #1 VOL=115.2535 u=0 imp:n=1 $FuelCenter
\overline{C}7 3 -6.4987 (-7 26 -43 1 -2):(-8 24 -45 1 -2):
           (-9 25 42 1 -2): (-10 23 44 1 -2) VOL=45.1028 u=0 imp:n=1 $CladEnds
8 3 -6.4987 (-18 14 -21 -19 1 -2):(-15 11 -21 20 1 -2):
            (-16 12 20 22 1 -2):(-17 13 22 -19 1 -2)
                    VOL=36.4110 u=0 imp:n=1 $CladFillets
9 3 -6.4987 (-48 49 19 -42 25 1 -2):(47 -46 -44 21 23 1 -2):
            (-48 49 43 -20 26 1 -2):(47 -46 -22 45 24 1 -2)
                    VOL=7.5423 u=0 imp:n=1 $CladGaps
C
10 4 -0.717 #1 #2 #3 #4 #5 #6 #7 #8 #9
         -27 28 -29 30 1 -2 VOL=339.3131 u=0 imp:n=1 $Water
C
11 0 (27:-28:29:-30:-1:2) u=0 imp:n=0 $Void
C ======================================================================
C Surface Card
1 PZ 0 $Bottom
2 PZ 380 $Top
C
3 P 0.0799 0.0799 0 0.01 $Inner Spacer
4 P 0.0799 -0.0799 0 0.01 $Inner Spacer
5 P -0.0799 -0.0799 0 0.01 $Inner Spacer
6 P -0.0799 0.0799 0 0.01 $Inner Spacer
C
7 C/Z -0.48 0 0.15 $Clad Left Circle
8 C/Z 0 -0.48 0.15 $Clad Bottom Circle
9 C/Z 0.48 0 0.15 $Clad Right Circle
10 C/Z 0 0.48 0.15 $Clad Top Circle
\mathcal{C}11 C/Z -0.43 0.43 0.28 $Outer TopLeft Fillet
12 C/Z -0.43 -0.43 0.28 $Outer BottomLeft Fillet
13 C/Z 0.43 -0.43 0.28 $Outer BottomRight Fillet<br>14 C/Z 0.43 0.43 0.28 $Outer TopRight Fillet
14 C/Z 0.43 0.43 0.28 $Outer TopRight Fillet
```
 C 15 C/Z -0.43 0.43 0.33 \$Inner TopLeft Fillet 16 C/Z -0.43 -0.43 0.33 \$Inner BottomLeft Fillet 17 C/Z 0.43 -0.43 0.33 \$Inner BottomRight Fillet 18 C/Z 0.43 0.43 0.33 \$Inner TopRight Fillet \cap 19 PX 0.43 \$Right Fuel Line 20 PX -0.43 \$Left Fuel Line 21 PY 0.43 \$Top Fuel Line 22 PY -0.43 \$Bottom Fuel Line C
 23 C/Z 0 0.43 0.10 \$Top Fuel Circle 24 C/Z 0 -0.43 0.10 \$Bottom Fuel Circle 25 C/Z 0.43 0 0.10 \$Right Fuel Circle 26 C/Z -0.43 0 0.10 \$Left Fuel Circle \overline{C} *27 PX 0.63 \$RightBoundary *28 PX -0.63 \$LeftBoundary *29 PY 0.63 \$TopBoundary *30 PY -0.63 \$BotBoundary \mathcal{C} 31 PX 10.71 **bigger and SRightAssemblyBoundary** 32 PX -10.71 \$LeftAssemblyBoundary 33 PY 10.71
34 PY -10.71
34 PY -10.71
34 SBotAssemblyBoundary \$BotAssemblyBoundary C^- 35 PX 10.81 \$RightAssemblyShroud 36 PX -10.81 \$LeftAssemblyShroud 37 PY 10.81 \$TopAssemblyShroud 38 PY -10.81 \$BotAssemblyShroud 39 PZ -0.1 \$BotZShroud 40 PZ 380.1 \$TopZShroud \cap 41 CZ 50 \$Void \mathcal{C} 42 PX 0.48 $\text{SExtra Plane}}$ 43 PX -0.48 $\text{SExtra Plane}}$ 44 PY 0.48 \$Extra Plane 45 PY -0.48 \$Extra Plane C 46 PX 0.15
47 PX −0.15 \$Extra Plane 47 PX -0.15 \$Extra Plane 48 PY 0.15 \$Extra Plane 49 PY -0.15 \$Extra Plane \mathcal{C} 50 CZ 0.475 SPuelRodGuide 51 CZ 0.6235 \$FuelRodClad C == C K Card MODE N KCODE 1000 1.36 10 400 \$# particles per cycle, guess, thrown, cycles KSRC -0.15 0 190 0.15 0 190 0 -0.15 190 0 0.15 190 C ==

```
C Burn Card
BURN TIME = 0.3 0.7 2 7 20 40 80 100 100 100 100
           100 100 100 100 100 100 $1 full cycles
     MAT = 1 2 POWER = 0.1310667 $MW
      PFRAC = 1.0 1.0 1.0 1.0 1.0 1.0 1.0 1.0 1.0 1.0 1.0
            1.0 1.0 1.0 1.0 1.0 1.0
      AFMIN = 1e-10 1e-10
    BOPT = 1.0 -24 1MPHYS ON
C ======================================================================
C Material Card
M1 41093.83c -.01 40090.81c -.508583 40091.81c -.110910
      40092.81c -.169528 40094.81c -.171801 40096.81c -.027678
      64157.83c -.0015 $Displacer
M2 40090 -.255449 40091 -.055707 40092 -.085150
      40094 -.086292 40096 -.013902 
      92238 -.388690 92235 -.097811 
     95241 - .007 90228 -1e-37
      90229 -1e-37
      90230 -1e-37
      90232 -1e-37
     91231 -1e-37
      93235 -1e-37
      93236 -1e-37
      93237 -1e-37
     98252 -1e-37
     nlib = .83c $Fuel PU
M3 40090.81c -.5145 40091.81c -.1122 40092.81c -.1715
     40094.81c -.1738 40096.81c -.0280 50118.81c -.012 $Cladding
M4 01001.81c .67 08016.81c .33 $Water
MT4 lwtr.16t butch water $Light Water
M5 02004.82c -1 $ControlRod
```
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