A QUALITATIVE ASSESSMENT OF THORIUM-BASED FUELS IN SUPERCRITICAL PRESSURE WATER COOLED REACTORS

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ABSTRACT

The requirements for the next generation of reactors include better economics and safety, waste minimization (particularly of the long-lived isotopes), and better proliferation resistance (both intrinsic and extrinsic). A supercritical pressure water cooled reactor has been chosen as one of the lead contenders as a Generation IV reactor due to the high thermal efficiency and compact/simplified plant design. In addition, interest in the use of thorium-based fuels for Generation IV reactors has increased based on the abundance of thorium, and the minimization of transuranics in a neutron flux; as plutonium (and thus the minor actinides) is not a by-product in the thorium chain. In order to better understand the possibility of the combination of these concepts to meet the Generation IV goals, the qualitative burnup potential and discharge isotopics of thorium and uranium fuel were studied using pin cell analyses in a supercritical pressure water cooled reactor environment. Each of these fertile materials were used in both nitride and metallic form, with light water reactor grade plutonium and minor actinides added. While the uranium-based fuels achieved burnups that were 1.3 to 2.7 times greater than their thorium-based counterparts, the thorium-based fuels destroyed 2 to 7 times more of the plutonium and minor actinides. The fission-to-capture ratio is much higher in this reactor as compared to PWR's and BWR's due to the harder neutron spectrum, thus allowing more efficient destruction of the transuranic elements. However, while the uranium-based fuels do achieve a net depletion of plutonium and minor actinides, the breeding of these isotopes limits this depletion; especially as compared to the thorium-based fuels.

1. INTRODUCTION

Generation IV nuclear reactor designs will need to feature improved economics and safety, minimized waste generation, and higher proliferation resistance than the current generation of reactors. The use of supercritical pressure light water as the coolant in a direct-cycle nuclear reactor offers potential for considerable plant simplification and consequent capital and O&M cost reduction compared with current light water reactor (LWR) designs, while building on the extensive and successful experience with LWR's. Several of the major advantages are given below:

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- It is possible to achieve high thermal efficiencies (up to 46%) given the thermodynamic conditions of the coolant at the core outlet (i.e. temperature and pressure beyond the critical point of water), which might result in significantly reduced fuel and capital costs. Also, for a given electric output, a higher thermal efficiency translates into a lower decay heat rate, which reduces the emergency core cooling system (ECCS) requirements.
- The reactor coolant does not undergo any change of phase. Therefore, the need for steam/water separation systems (e.g. steam separators and dryers) and BWR-like re-circulation pumps is eliminated. This makes the reactor vessel shorter and radially more compact. Also, the control rods can be inserted from the top of the core, which improves their reliability and accessibility.
- The core coolant directly drives the turbine and no steam generators or pressurizer are needed. The steam turbine throttle valve can control the pressure.
- Supercritical water at high temperature exhibits very high specific volumes, which can result in a core-average coolant density as low as 200kg/m3. This is a remarkable characteristic of these systems that enables design of a fast-spectrum core, hence making it possible to effectively burn actinides (from LWR spent fuel) or to maintain a near-unity breeding ratio to achieve long irradiation cycles without refueling.
- Because the whole coolant flow undergoes a rather large temperature rise (typical inlet and outlet temperatures are 300 and 450°C, respectively), the flow rate in the core is considerably lower than in LWR's. Therefore, the pressure drop and pumping power are also lower. Also, because the coolant can accommodate large specific enthalpy rises, a two-loop primary system can be employed even at large reactor powers (e.g. 3,500MWth). This results in a lower coolant inventory in the primary system and ultimately in a more compact containment.

The use of supercritical water as a coolant was studied in the US [1], and in Japan [2,3]. However, several of the fast reactor designs are fairly complicated as they consist of one outer and two inner radial blankets, an axial blanket, two seed regions and several layers of zirconium hydride to attain a negative void reactivity coefficient. The Canadians have studied both indirect and combined superheated/saturated direct cycle supercritical water pressure tube reactors [4,5]. However, it is felt that significant simplification of the aforementioned systems can be achieved, resulting in further cost reductions. In particular, advancements in core design and fuel cycles could make a large impact in this area. Therefore, an alternative approach is being studied that uses a blanket-free fertile fueled core and retains the hard spectrum for effective burning of plutonium and minor actinides from spent LWR fuel.

The work discussed here is part of a larger effort under a Nuclear Energy Research Initiative (NERI) grant to investigate the suitability of a supercritical pressure light water cooled reactor for producing low-cost electricity as well as for actinide burning. The purpose of the NERI project is to explore the possibility of utilizing water at supercritical pressure and temperature to design an economic and safe reactor with a simple and flexible core and plant design as was described above. This reactor should be able to support a broad variety of missions including actinide burning, pure electricity generation, heat and electricity co-generation, district heating, desalinization, and fuel cell recharging. While there are several key issues in the development of this reactor concept, the issue of simple, inexpensive core designs that attain high fuel utilization (i.e., uranium resource utilization) and minimize waste (i.e., minor actinide management) are what will be discussed in this paper; particularly a comparison of thorium and uranium fuels. On the basis of fuel utilization and proliferation resistance, the use of thorium-based fuel would tend to support both of these concerns,

i.e., it would conserve uranium and lessen the plutonium (and thus minor actinide) production within the fuel, while allowing a higher destruction rate of any initially charged plutonium and minor actinides.

2. FUEL PARAMETERS AND COMPUTATIONAL TOOLS

The fuels studied here include both uranium-based and thorium-based fuels. Previous work has shown that the use of thorium in fast spectrum systems can be advantageous for both safety characteristics (reactivity coefficients) and actinide destruction [6]. The fuel forms being considered are oxide, nitride, and metallic fuel, with this paper focusing on the two latter candidates.

A qualitative analysis has been performed to observe which fuel form would support the highest reactivity-limited burnup, and would have the most proliferation resistant isotopics at a particular burnup. The results presented here will define the trends of specific fuels that will be used in the full core analysis. For this study, a pin cell analysis was used, where the full length of a cylindrical fuel pin was incorporated to accommodate the change in coolant density along the pin. The parameters of the pin cell and BOL plutonium/minor actinide isotopic fractions within the fuel can be found in Table I, and Figure 1 shows a graphical representation of the unit cell used in the calculations.

Table I. Fuel parameters used in the analysis.

Parameter	Value
Fuel Radius (cm)	0.368
Gap Thickness (cm)	0.02
Clad Outer Radius (cm)	0.44
Active Fuel Length (cm)	120
Pin Pitch - triangular (cm)	1.01
Fuel Temperature - average (K)	900
Number of Coolant Nodes	22
Pu and MA BOL Fractions	
Pu-238	1.6%
Pu-239	46.3%
Pu-240	20.8%
Pu-241	8.0%
Pu-242	3.2%
Np-237	8.5%
Am-241	9.1%
Am-242	0.0%
Am-243	1.8%
Cm-244	0.6%

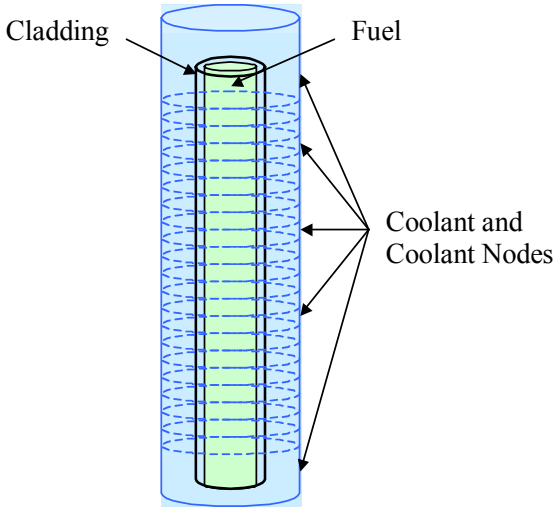

Figure 1. Representation of unit cell model used (not to scale).

The plutonium and minor actinides comprise 13-20wt% of the fuel, with the remainder of the fuel consisting of uranium or thorium as mono-nitrides or in a zirconium-metal matrix. More specifically, the thorium-based fuels have a constant plutonium and minor actinide content of 20%, while the uranium-based fuel plutonium and minor actinide content varies.

The MOCUP [7] code was used to analyze the reactivity-limited burnup and isotopic content of the fuel through each burnup step. MOCUP employs MCNP [8], a well-known Monte Carlo code capable of calculating fluxes, reaction rates, and eigenvalues in general, 3-D geometry using continuous cross-section data; and ORIGEN [9], a matrix exponential method code that calculates the generation and depletion of isotopes, or elements, in a given neutron flux. MOCUP takes specific output data (including cross-section data, fluxes, and reaction rates) from MCNP and passes it to ORIGEN, where new isotopic information is generated and passed back to MCNP for the next calculation. This gives time dependent information about the reactivity swing and isotopics for the specified problem. In this work, one-year time steps were used which has been validated for fast spectrum systems previously [6].

3. REACTIVITY LIMITED BURNUP

A long core life is highly desirable from the standpoint of proliferation and resource utilization. However, the material and safety issues associated with high neutron fluences and the fuel behavior at very high burnups may be a problem. Nonetheless, it is important to perform scoping studies that can produce results that lie beyond current technologies. The work presented here shows some fuel lifetimes in excess of 10 effective-full-power-years, although no calculations were performed for times longer than 10 years. The longer lifetimes were extrapolated based on the 10-year trend of each fuel.

Figures 2 and 3 show the reactivity versus the effective-full-power-years (EFPY) for both nitride and metallic fuels, respectively. Note that the pin was held at a constant power of 24 kW, or a linear power density of 200 W/cm.

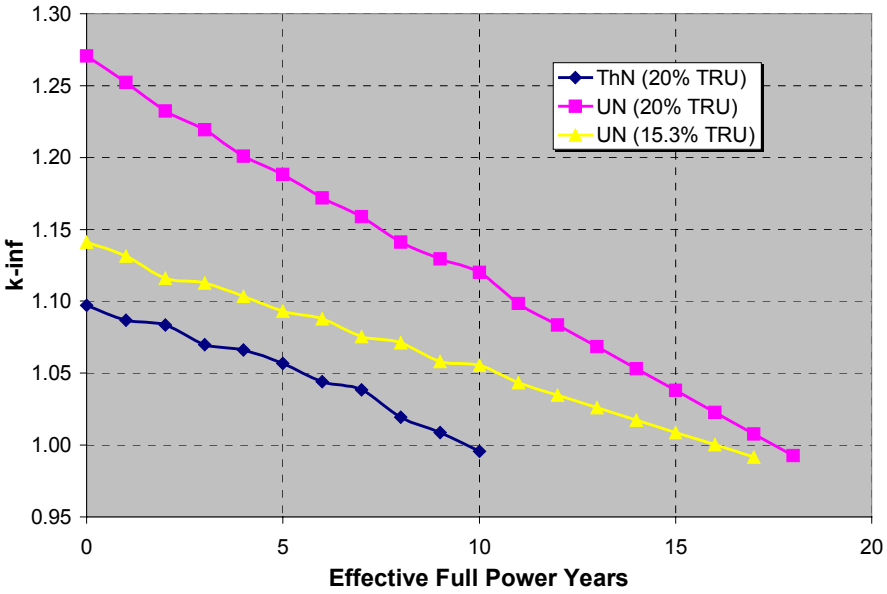

Figure 2. Reactivity versus effective-full-power-years for nitride fuels.

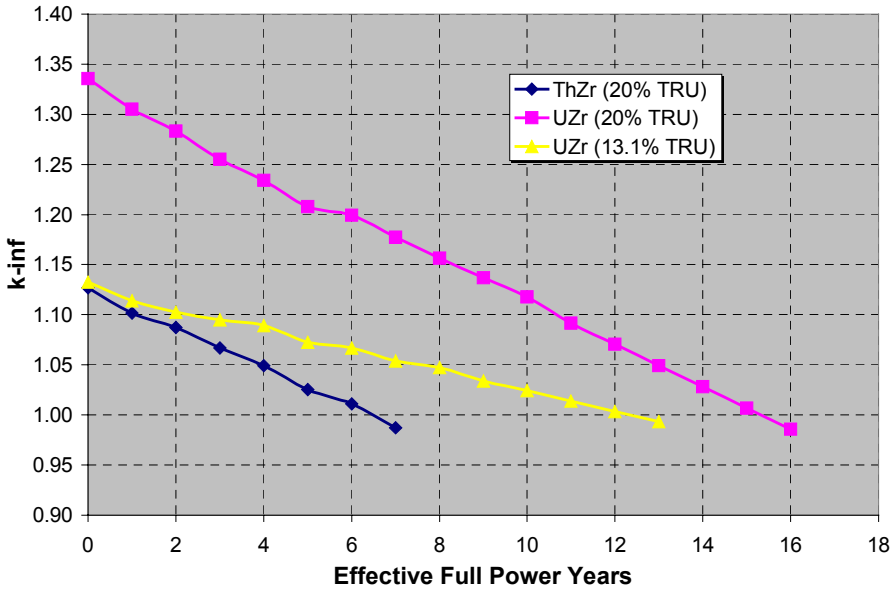

Figure 3. Reactivity versus effective-full-power-years for metallic fuels.

If the end-of-life (EOL) is chosen at $k_\infty = 1.0$, then all but two of the fuels will have lifetimes beyond 10 years; equal to approximately 200 – 240 MWd/kg burnup. The exceptions are the ThZr and ThN fuels, which become "subcritical" at approximately 6.5 years and 9.7 years, respectively. The achievable burnup is then 140 – 145 MWd/kg for these fuels. In addition, the lower enriched (i.e., lower weight percent of plutonium and minor actinides) uranium fuels have relatively flat reactivity curves, which are highly desirable from a safety and control aspect. Initially, these lower enriched cases were analyzed in an attempt to lower the BOL reactivity.

The use of pin cells in such an analysis will not give quantitatively correct results due to the high leakage in a full core. However, all of the cases presented here used the same parameters, with the exception of the fuel type. Thus, the qualitative information can be used to select a fuel, which in turn will be used in full core analyses.

4. ISOTOPIC ANALYSIS

The isotopics (plutonium and minor actinides) of the fuel are important; particularly at discharge. This can determine whether the fuel is a good candidate based on proliferation concerns and waste management. Specifically, a total net reduction in plutonium and minor actinides is desirable. Figures 4 and 5 show the average concentration of the transuranics throughout the burnup for both nitride and metallic fuels, respectively. Note that the negative values indicate, on average, a depletion of the isotope, while the positive values indicate isotopic generation.

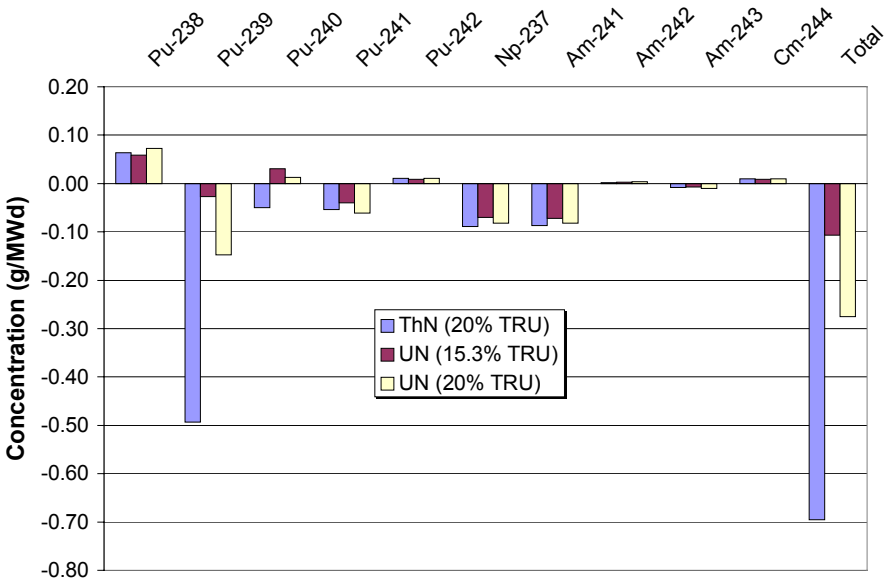

Figure 4. Generation and depletion of transuranic isotopes in nitride fuel.

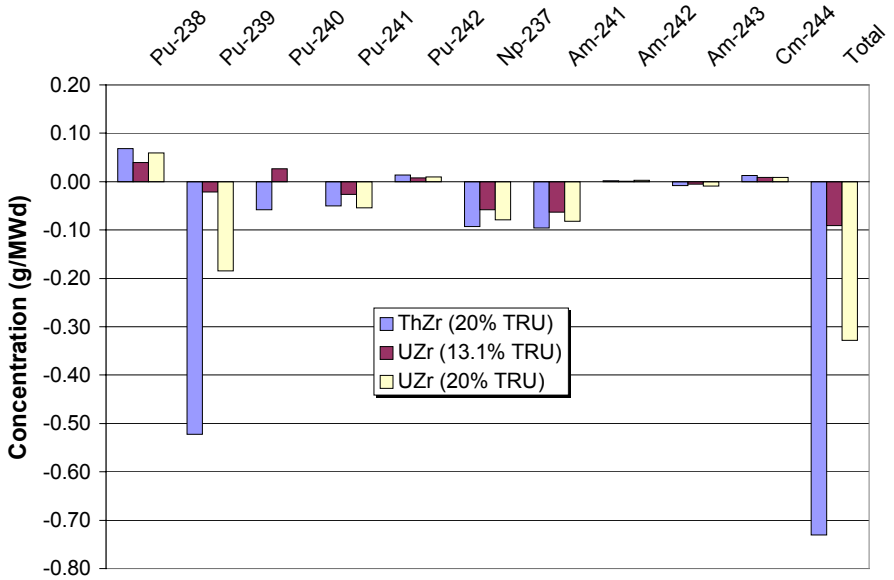

Figure 5. Generation and depletion of transuranic isotopes in metallic fuel.

The information from Figures 4 and 5 indicate a higher destruction rate in the thorium-based fuels. This is better seen in Figure 6, which shows the total plutonium and minor actinides that are "burned" for 7-10 effective full power years of operation.

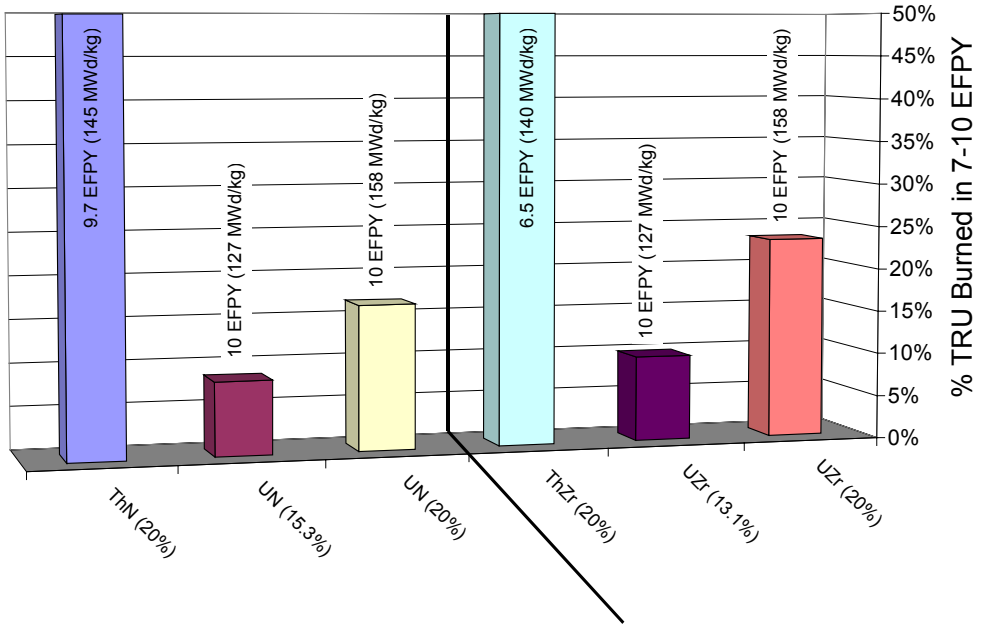

Figure 6. Comparison of plutonium and minor actinide destruction.

Figure 6 shows that the thorium-based fuels will consume about one-half of the plutonium and minor actinides in the fuel. Also of interest is the plutonium vector (plutonium fractions) at specific points in time, which can be seen in Table II.

Table II. Plutonium vector at specific times.

Fuel	EFPY	Initial Pu+MA	Plutonium Fractions				
			Pu-238	Pu-239	Pu-240	Pu-241	Pu-242
ThN	10	20%	14.4%	25.7%	40.7%	9.8%	9.4%
ThZr	7	20%	14.7%	25.1%	39.6%	10.8%	9.8%
UN	10	15.3%	7.6%	53.5%	28.2%	5.9%	4.8%
UN	10	20%	8.2%	51.0%	29.5%	5.9%	5.3%
UZr	10	13.1%	7.1%	53.1%	28.6%	6.3%	4.9%
UZr	10	20%	8.6%	48.8%	30.6%	6.2%	5.8%

Note the high Pu-238 fractions in the thorium-based fuel (at 14.4% and 14.7%). Pu-238 is of special interest because of its high decay heat rate and spontaneous neutron rate, which would make this fuel more proliferation resistant [10]. The high concentration of even-numbered plutonium isotopes for all fuels would make them fairly resistant to proliferation.

CONCLUSIONS

The reactivity and isotopics of nitride and metallic fuels containing uranium or thorium, and loaded with reactor grade plutonium and minor actinides, were studied and compared in a supercritical pressure, light water reactor environment. The large axial change in density, resulting in only a 200 kg/m^3 towards the top of the fuel, and the very tight pitch used in this study result in a fairly hard spectrum. This spectrum is ideal for destruction of the plutonium and minor actinides due to the high fission to capture ratio.

The reactivity limited burnup favors the uranium-based fuels with lifetimes that were 1.6 to 2.7 times greater than the thorium-based fuels for the same initially loaded weight percent of plutonium and minor actinides; and 1.3 to 2.5 times greater for lower initially loaded weight percent of plutonium and minor actinides. The lower weight percent of plutonium and minor actinides in the uranium-based fuels were used to achieve similar (i.e., lower) BOL reactivities. However, this also lowers the amount of plutonium and minor actinides that can be destroyed during the uranium fuel cycles. While the cycles can be longer than for thorium-based fuel, the trade-off is a much lower destruction rate.

The average net depletion rates per unit energy are fairly similar for both types of thorium fuel (nitride and metallic), at a rate of about -0.70 g/MWd. Although the uranium-based fuels also have net depletion rates, they are approximately 2-7 times less than that of the thorium-based fuels. This is due primarily to the additional breeding of plutonium and its associated minor actinides. It is important to note, however, that the thorium fuels will have a significant amount of U-233 present at end-of-life (EOL). Also, addition of uranium to the thorium fuel will be necessary to denature the U-233, which will affect the performance of this fuel. It is important to note, however, that the plutonium vector is highly degraded in all cases (i.e., there are high concentrations of the even numbered plutonium isotopes). Of particular interest is the high concentration of Pu-238, which has a high decay heat rate and spontaneous neutron rate.

While, qualitatively, the uranium-based fuels can achieve much higher burnups than the thorium-based fuels, clearly the use of a thorium-based fuel is superior in the management of plutonium and minor actinides in a fuel cycle.

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