Avaliação de combustível cerâmicos compostos baseados em urânio, tório e plutônio para reatores nucleares

(Evaluation of composed ceramic fuels based on uranium, thorium, and plutonium for nuclear reactor)

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Abstract

The energy generated using thorium as nuclear fuel is an attractive way to preserve the uranium reserves and reduce the radiotoxicity wastes. Today, global thorium reserves are around four times that of uranium reserves, and ThO_2 is cheaper than UO_2. The next generation of reactor shows fast reactor types using $(U\text{-}Pu)O_2$. It contains combinations of ceramic fuels based on UO_2, PuO_2, and ThO_2. Using a new version of the FRAPCON code, get the ability to predict thorium fuels. The FRAPCON fuel codes permit the addition of capacity to simulate thorium fuel and compare fuel performance with standard UO2. The proposed strategy uses ThO_2 75 wt% composed of UO_2 25 wt% with enrichment equal to 19.5 wt%. In comparison, the second strategy uses a balanced composition of ThO_2 at 93 wt.% combined with PuO_2 at 7.0 wt.%.

Keywords: Thorium dioxide, Plutonium dioxide, FRAPCON.

INTRODUCTION

Around the 1950 decade, analysts presumed that uranium reserves were a limited resource. Th-232 is about four times as abundant as U-238 in the earth's crust. The only naturally occurring isotope of thorium is Th-232 compared with uranium, which shows three isotopes, U-238, U-235, and U-234. During wartime in 1944, at Chicago University, the detection of plutonium isotopes resulted from successive neutron capture reactions, generating the Pu-239, with a half-life of 24,100 years [1]. There are three primary fissile materials U-235, U-233, and Pu-239. Comparatively, naturally fertile materials occur, such as Th-232 and U-238. The breeder reactor uses these fertile materials, which are transmuted by capturing neutrons and converting them into fissile isotopes such as Pu-239 and U-233 [2]. Once-through thorium fuel cycles proposed for nuclear units display heterogeneous and homogeneous design classes.

When uranium is still relatively rare, the thorium breeder reactor could be a viable solution for civilian nuclear use. The Th-232 conversion in the U233 breeder fuel cycle has advantages over the U238 transmuting to the Pu-239 breeder fuel cycle. However, the thorium conversion to fissile U-233 is less efficient than the conversion of U-238 to Pu-239 with high efficiency [3]. Thus, it would be crucial to design power systems based on breeder reactors using the recovered fissile isotopes isolated from irradiation. The nuclear generation of electric power mainly uses the pressurized water reactor (PWR), which uses water at a very high pressure of 15 MPa in a primary circuit and forms steam in a secondary circuit.

The Indian Point-1 plant was the pioneer commercial power plant with total thorium fuel. In 1976, a research program built the Shipping Port Light Water Breeder Reactor (LWBR), which used $(Th–U)O_2$ from 1977 to 1982 [4]. In the mid-1990 years, Alvin Radkowsky proposed a reactor based on thorium fuel, following the seed-blanket-unit (SBU) concept that was a one-for-one replacement for a standard PWR assembly. Using thorium fuel in a blanket produces less actinide than uranium fuel. It typically creates a significant part of the power in the subcritical regions known as the blanket regions in the SBU core design. The nuclear generation of electric power mainly uses the pressurized water reactor (PWR), which uses water at a very high pressure of 15 MPa in a primary circuit and forms steam in the secondary.

Nuclear units, under regular operation, can produce a sensible number of plutonium isotopes, Pu-239 and Pu-241, stored in spent fuel. The PWRs, having over 1.0 GWe, discharged about 180 Kg of Pu-239 and 220 Kg of U-235 in the spent fuel; nuclear waste-producing plutonium stockpiles achieve about 540 tons, with 316 tons resulting from civilian reactors, estimated with significant uncertainties. Then, reprocessing separates plutonium from uranium and fission products through a chemical route called Plutonium and Uranium Recovery by Extraction PUREX [5]. This process separates uranium and plutonium from nuclear fission products and spent nuclear fuel.

Worldwide has used uranium oxide fuels commercially in nuclear reactors. For the near future, atomic fuels must provide proliferation resistance, less atomic waste, and prevent the increase of plutonium stockpiles and minor actinides. An important option to reduce the excess plutonium is to use this plutonium in advanced options, such as thorium-plutonium-based fuel.

India has developed a thorium plan because it only has 2% of the World's uranium reserves. In contrast, India has 25% of the World's thorium reserves. Currently, uranium and plutonium $(U–Pu)O_2$ mixed oxide (MOX) represents around 2% of the fuel market. This study created an assessment for thorium-based fuel as $(Th–U)O_2$ and $(Th–Pu)O_2$ proposed to AP1000 without changing the design. The simulation used a fuel performance code FRAPCON adapted to support $(Th–U)O_2$ and ThMOX [6]. Table I reveals the design parameters of AP1000 Westinghouse and EPR Framatome.

Table I: Design parameters of AP1000 and EPR units.

Design parameters	**AP1000**	**EPR**
Thermal power output (MWth)	3415	4590
Number of coolant loops	2	4
Active length (m)	4.26	4.20
Fuel outside diameter (mm)	9.10	9.50
Average linear heat (KW/m)	18.76	16.67

The Nuclear Regulatory Commission (NRC) endorsed the FRAPCON of Pacific Northwest Laboratory (PNNL). The licensing process works with fuel performance codes to predict the steady-state thermal-mechanical behavior. Today, FAST is the current NRC thermal-mechanical fuel performance code, the next evolution of FRAPCON, containing transient capabilities from FRAPTRAN with a modern code architecture.

MATERIALS AND METHODS

Today, AP1000 technology innovation has achieved up to 1400 Mwe as CAP-1400. It uses adapted versions of the FRAPCON code for (Th-25% U)O_2 and (Th-7% Pu)O_2, available for the thermal response in a steady state. It uses Vegard's law permit to calculate the physical properties, such as the melting point, enthalpy, and specific heat, of pure ThO_2, UO_2, and PuO_2 and (Th-U)O_2 or (Th-Pu)O2 mixed oxides. Vegard's law permits the estimation of the physical properties, such as the melting temperatures, enthalpy, and specific heat, of pure ThO_2, UO_2, and PuO_2 and (Th-U)O_2 or (Th-Pu)O_2 mixed oxides.

Physical properties

The FRAPCON fuel code can foresee the fuel response of UO_2 and (U-Pu)O_2 pellets coated with zirconium alloys [7]. Thus, FRAPCON must need a deep revision to account for the (Th-U)O_2 and (Th-Pu)O material properties to examine steady-state thermal performance in a PWR 17×17. Composite fuels would show thermal properties as intermediates between the pure contents if the production route produced a homogeneous system [8]. Table II shows the most referenced ceramic fuels' physical properties, with lattice parameters and thermal properties such as conductivity, expansion, and heat capacity at 25 °C.

Table II: Physical properties of the ceramic fuels UO_2, ThO_2 PuO_2, and U_3Si_2.

Physical parameters	**UO_2**	**ThO_2**	**PuO_2**	**U_3Si_2**
Lattice parameter (10^{-10}m)	5.5974	5.4704	5.9920	7.3299
Melting point (°C)	2850	3340	2744	1665
Thermal conductivity (W/m-K)	8.68	10.5	7.5	8,0
Thermal expansion (10^{-6} m)	9.75	8.43	9.04	15.5
Heat capacity (J/kg K)	235	234	240	201

Vegard's law is an empirical approach resembling the Koop–Neumann rule (KNR) for compiling mixture properties. Melting temperatures using two-phase simulations of ThO_2, UO_2, and PuO_2, which lie in the range of (3662 ± 12) K, (3062 ± 18) K, and (2812 ±18) K, respectively. Equation (A) shows the melting point of the composite fuel $UO_{2(X)}ThO_{2(1-X)}$. Equation (B) represents the theoretical density of the same formulation $UO_{2(X)}ThO_{2(1-X)}$ given as a function of UO_2 content and temperature. Thus, using Vegard's law can calculate mixture melting temperatures.

$$T_{(MP)} = 0.008275 \times X^2 - 6.018 \times X + 3638 \qquad \text{(A)}$$

where X is the UO_2 fraction, and $T_{(MP)}$ represents the melting temperature of the composite in K.

$$D = 10.087 - 2.891 \times 10^{-4}T - 6.354 \times 10^{-7}(X)T + 9.279 \times 10^{3}(X) + 5.111 \times 10^{-6}(X)^2 \text{ (B)}$$

Where X is the UO_2 fraction and T is the temperature in K.

The thermal conductivity of UO_2 is 10% lower than that of pure ThO_2, coded in the FTHCON subroutine. FTHCON computes the thermal conductivity given as a function of temperature, burnup, porosity, and fission products [8],[9]. However, the impact of PuO_2 fractions inserting deviations is more substantial for $(Th\text{-}Pu)O_2$ than $(Th\text{-}U)O_2$ because of PuO_2 contents. Equations (C) and (D) represent the thermal conductivities of ThO_2 and UO_2 given as functions of temperature in K, valid to (298 K ≤T≤ 2800 K)

$$k_{ThO_2}(T) = 6.232 \times 10^{-12}.T^4 - 3.485 \times 10^{-8}.T^3 + 7.218 \times 10^{-5}T^2 - 0.06825.T + 29.06 \text{ (C)}$$

$$k_{UO_2}(T) = 2.469 \times 10^{-12}.T^4 - 1.394 \times 10^{-8}.T^3 + 3.022 \times 10^{-5}T^2 - 0.038156.T + 16.74 \text{(D)}$$

where K is thermal conductivity (W/m-K), and T is the temperature in K.

The thermal expansion ratio at room temperature of PuO_2 is 7.8 um/m-°C, of ThO_2 is 8.9 um/m-°C, and UO_2 is 9.76 um/m-°C. Currently, SPS route change slight all thermal coefficients of the ceramic fuels, also increase your densities. Equations (E), (F), and (G) express the linear thermal expansion of ceramic pellet fuels UO_2, ThO_2, and PuO_2 sintered.

$$\frac{\Delta L}{\Delta L_0} UO_2(T) = 0.00001 \times T - 0.003 + 0.04.\ e^{\left(\frac{6.9\times10^{-20}}{1.38\times10^{-23}.T}\right)} \qquad \text{(E)}$$

$$\frac{\Delta L}{\Delta L_0} ThO_2(T) = \frac{(-0.179+5.097\times10^{-4}.T+3.732\times10^{-7}.T^2-7.594\times10^{-11}.T^3}{100} \qquad \text{(F)}$$

$$\frac{\Delta L}{\Delta L_0} PuO_2(T) = \quad 9\times10^{-6}.T - 2.7\times10^{-3} + 0.007\times\ e^{(\frac{-7\times10^{-20}}{1.38\times10^{-23}.T})} \qquad \text{(G)}$$

where $\frac{\Delta l}{\Delta L_0}$ is a linear thermal expansion dimensionless, and T is the temperature in K, proposed in the range (200 K ≤ T ≤ 2000 K)

The subroutines of FRAPCON describe the specific heat capacity and enthalpy of FCP and FENTHL of fuel. Equations (H), (I), and (J) represent the fitting correlations used for the specific heat of the UO_2, ThO_2, and PuO_2. Equations (K) and (L) denote the enthalpy relative to the temperature de 298 K. The thorium has a reduced heat capacity and enthalpy than UO_2 and PuO_2 and no existing burnup dependence.

$$Cp_{UO_2}(T) = -1.381\times10^{-11}.T^4 + 9.990\times10^{-8}.T^3 - 0.0002608.T^2 - 0.3169.T + 173.5 \text{ (H)}$$

$$Cp_{ThO_2}(T) = -7.348\times10^{-12}.T^4 + 5.301\times10^{-8}.T^3 - 0.0001378.T^2 - 0.1891.T + 198.4 \text{ (I)}$$

$$Cp_{PuO_2}(T) = -1.451\times10^{-11}.T^4 + 1.049\times10^{-7}.T^3 - 0.0002736.T^2 - 0.3099.T + 200.7 \text{ (J)}$$

where Cp represents the heat capacity in (J/kg-K), and T is the temperature in K, suggested in the range (298 K ≤T≤ 2800 K)

The fuel enthalpy results from integrating the heat capacity from 298 K to the temperature analyzed. The specific enthalpy of UO_2, measured as h(T)-h(298.15 K), varies from zero at room temperature to ~ 11100 (kJ/kg) at 2800 °C.

$$\Delta H_{UO_2}(T) = 3.056\ \times10^{-8}T^3 - 9.186\ \times10^{-5}T^2 - 0.3382\ .T\ - 120.9 \text{ (K)}$$

$$\Delta H_{ThO_2}(T) = 1.115\ \times10^{-8}T^3 - 3.176\ \times10^{-5}T^2 - 0.3382\ .T\ - 107.8 \text{ (L)}$$

where $\Delta H(T)$ is enthalpy in J/kg, and T represents the temperature in K stated in the range (298 K $\leq$T$\leq$ 2800 K)

Mechanical models, including the elastic properties with numeric fitting, such as FELMOD, calculate Young's Modulus and FPOIR for the Poisson ratio. Table III shows the estimated values of the elastic properties for UO_2 and ThO_2 at room temperature.

Table III: Elastic properties of UO_2 and ThO_2 sintered fuels at 25 °C

Elastic properties	**UO_2**	**ThO_2**
Modulus of elasticity (GPa)	206.45	256
Shear modulus (GPa)	78.48	99.5
Poisson's ratio	0.3025	0.291

RESULTS AND DISCUSSION

The simulation of thorium fuels using the AP1000 is a PWR reactor that uses a robust fuel assembly 17x17 with 157 fuel assemblies, 264 fuel rods, and 24 control rod guide tubes. Figure 1 shows the average temperature of the fuel rod during a burn cycle of 1200 days. The fuel distribution shows three regions using enrichment levels of 2.35%, 3.40%, and 4.45%.

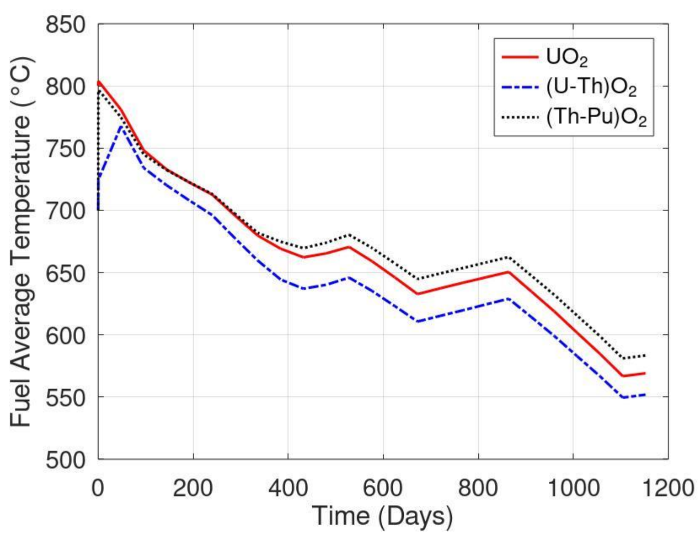

Figure 1. Fuel temperature in the burn cycle for UO_2, $(Th\text{-}U)O_2$, and $(Th\text{-}Pu)O_2$.

The dimensional changes of the active fuel length of 4.2 m are factors involving fuel densification and swelling aspects with solid dependence on gap closure. Fuel stack elongation is an effect of thermal expansion, while it gave densification at low burnup as a function of the irradiation and temperature. Figure 2 displays fuel stack elongation during a burn cycle of 1200 days.

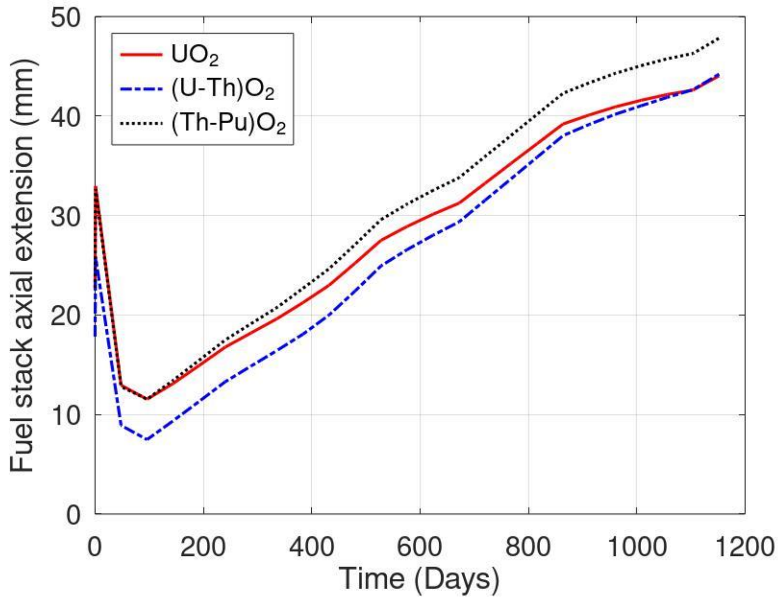

Figure 2. Fuel stack elongation under the burn cycle for UO_2, (Th-U)O_2, and (Th-Pu)O_2.

The expected fuel-cladding gap in operation shows dependence only on fuel and cladding thermal expansion. Figure 3 shows the gap closure.

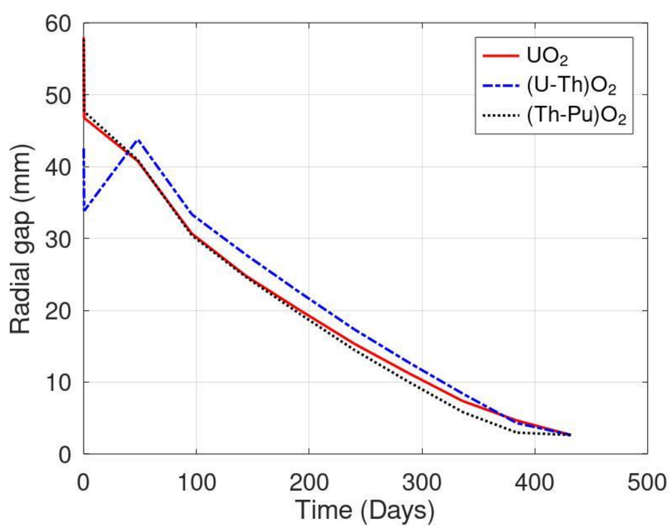

Figure 3. Radial gap space during the burn cycle for UO_2, (Th-U)O_2, and (Th-Pu)O_2.

Stored energy in the fuel pellets includes fuel densification and the effects of fission products that decrease the gap conductance increasing the stored energy. Developments such as pellet cracking speed up outer relocation of the pellet parts, promoting an additional gap closure. Thus, reducing the heat transfer capability, the stored energy increases. Figure 4 shows the stored energy during burn cycles for ceramic fuels under analysis.

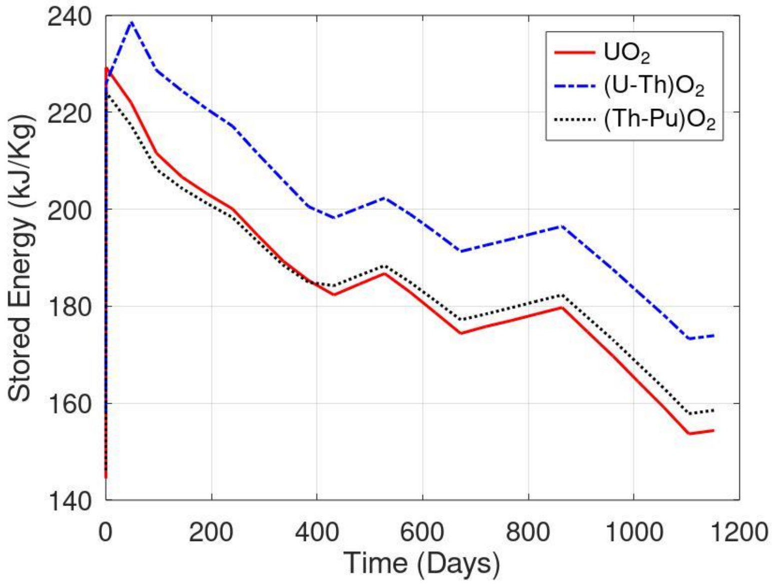

Figure 4. Stored energy during the burn cycle for UO_2, (Th-U)O_2, and (Th-Pu)O_2.

CONCLUSIONS

Plutonium and uranium stockpiles have become problems that need new solutions. The most critical features of thorium composite fuels are the higher thermal conductivity, melting point, and lower thermal expansion, which are essential for safety analysis. These beneficial attributes allow an extended burn cycle and operation at a higher power level, reducing the FGR compared with UO2. Reactors with a thermal neutron spectrum permit the use of the essential characteristics of the U-233/Th-232 cycles, showing conversion ratios near unit so-called breeding reactors. International regulations for the peaceful use of nuclear energy help prevent the proliferation of atomic weapons.

Using thorium because of lower and stable international prices and substantial natural reserves mixing with plutonium represents a decisive advantage in the near time. While (Th-Pu)O_2 fuel shows a reasonable reactivity control depending on the fission isotope dominating, plutonium isotopes Pu-239, and uranium isotopes U-235, U-233, which causes the dynamic reactivity control possible with little difficulty. ThO_2 spent fuel has the benefit of chemical inertness. PUREX can recover plutonium that is a fissile isotope engineered after a chain reaction, which minimizes the risk of making weapons with it. Comparative to waste, thorium fuels have the advantage of lower radiotoxicity but produce a reduced amount of U-232.

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