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# ANALYSIS OF A PRESSURIZED POWER REACTOR USING THORIUM MIXED FUEL UNDER REGULAR OPERATION

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## ABSTRACT

This work discusses a parametric study applied to nuclear power generation based on a mixed fuel formed by the composition of thorium-uranium oxide (Th-U)$O_2$. Also, approached in this study the physical neutrons models of a fuel system composed of $ThO_2$ 75 wt% and $UO_2$ 25 wt%, with 19.5% enrichment of U-235. The thermodynamic features of the thorium-uranium fuel system compared with the properties of uranium dioxide. Thorium-based fuel operating extended fuel cycles reach of over 80 GWd/MTU in a pressurized water reactor (PWR). Homogenous distribution of thorium-based fuel, used on the reactor core, could reduce Pu-239, once U-233 production capacity dependent on Th-232 replacing U-238 in the fuel matrix. The mixed oxide fuel has a lower buildup of Pu-239, causing the linear heat rate distribution slope to flatten and lowering fuel porosity. The release of gaseous fission products models for (Th-U)$O_2$ could have different diffusion coefficients when compared to uranium oxide models. Besides, resulting in lower thermal gradients than $UO_2$ and a reduction in fuel swelling. This parametric study reviews the aspects of radioactive decay chains of uranium and thorium. It founded the simulation using approved nuclear codes, such as SERPENT for neutron physics calculations and the FRAPCON code, which defines the licensing process. The results show that thoria based fuel has a higher performance than $UO_2$ fuel in regular operation and can improve safety margins.

## 1. INTRODUCTION

Today, nearly 100% of the currently operating nuclear power reactors are thermal reactors. The largest share of 61% comprised pressurized water reactors (PWR), followed by boiling water reactors (BWR) at 21%. The standard fuel, $UO_2$, requires enrichment levels up to about 4.8% of U-235 for light-water reactors (LWR). The nuclear industry has a vast knowledge of mechanical, thermal, and irradiation properties of $UO_2$ accumulated throughout the fifth years of experience in manufacturing the fuel for reactors. However, as the industry researches new GEN-IV reactor designs, researching different fuels is essential because $UO_2$ is limited by its low thermal conductivity, especially at high temperatures. Two main concerns driving the research and development of fuel technology are safety and waste.

In a less quantifiable manner, safety and waste characteristics also play significant roles in the public option and approval of the use of nuclear energy systems [1]. This investigation focuses on fuel designs using $ThO_2$-$UO_2$ so-called Th-MOX used in pressurized power reactors. Table 1 illustrates the physical properties of pure $UO_2$ and $ThO_2$ at room temperature.

**Table 1: Comparison of various properties of $UO_2$ and $ThO_2$**

| **Properties** | **$UO_2$** | **$ThO_2$** |
|---|---|---|
| Weight Molar (g/mol) | 270 | 264 |
| Thermal conductivity (W/m-°C) | 8.0 | 10.3 |
| Melting Point (°C) | 2850 | 3650 |
| Theoretical density (g $cm^3$) | 10.96 | 10.0 |
| Lattice Parameter (angstrom) | 5.4704 | 5.5974 |
| Thermal Expansion (µm/m°C) | 9.8 | 8.9 |
| Heat Capacity (J/kg-K) | 328 | 266 |
| Young modulus (GPa) | 200±5 | 233 ± 5 |
| Shear modulus (GPa) | 78 ± 8 | 94 ± 8 |
| Poisson rate (unitless) | 0.31 | 0.28 |

Civilian nuclear power reactors focused on the application of $(U\text{-}Pu)O_2$ mixed-oxide (MOX) fuels using many fractions of $PuO_2$ to improve performance [2-3]. However, neutron kinetics study appoints that MOX fuel might suffer from a definite void reactivity coefficient. Though much debate to exist on the analysis of these concepts, they have spurred investigations into the use of MOX fuels. Mixed-oxide fuel is atypical by isotopic composition, as it contains both uranium and plutonium oxide [4-5]. The MOX fuel recovering a portion of the remaining energy from spent fuel and to prevent plutonium use in weapons systems. Today, the global stockpile of plutonium weighs 520 metric tons [6-8].

Thorium-based fuels became subject too much research, and a wide range of applications focused on the replacement of uranium dioxide ($UO_2$) fuel. The estimated global thorium reserves are about four times that of uranium reserves. $ThO_2$ is chemically stable compared to $UO_2$ and more cost-effective because uranium prices have reached higher values over the last decade. There are a few isotopic wastes divided by half-life, such as high, medium, or low-level, depending on the half-life of the emitted isotopes. Isotopes emit alpha, beta, and gamma radiation, including Pu-238, U-234, Np-237, and Am-241. Longer half-lives can reach millions of years. Therefore, reducing nuclear waste is a critical problem to solve. The fissionable isotopes used in nuclear reactors include U-233, U-235, Pu-239, and Pu-241. The fertile isotopes are U-238 and Th-232.

With replacing $UO_2$ by $ThO_2$ must consider all effects such as the neutron absorption cross-section, radioactive decay, and transmutation chains. However, several experiments performed concluded that thorium and uranium combined fuels can exhibit a similar response to $UO_2$ fuel.

The nuclear fuels contain up of one or more fissile isotopes such as U-235, Pu-239, and U-233 also the fertile isotope U-238 or Th-232. The fission of U-233, produced through neutron capture from Th-232, results in neutrons yields higher than those of U-235 and Pu-239. Th-232 reduces fission actinides produced when compared to U-235 and Pu-239. Thorium is a beneficial option because of decreasing radiotoxicity of spent fuel. Also, the thermal

conductivity of $ThO_2$ is about 10% higher than the conductivity of $UO_2$ [9]. One concept that shows promising performance is the use of $ThO_2$-based fuel. When analyzing the thoria-urania $(Th\text{-}U)O_2$ cycle, there is evidence of better safety conditions. Thoria fuels have a higher melting point, reduced fuel temperature, reduced gas pressure, and a better mechanical response when compared with $UO_2$ fuels. The $(Th\text{-}U)O_2$ fuel uses Th-232 as a fertile isotope loaded into the core reactor, where nuclei of Th-232 absorb a neutron and transmute into nuclei of Th-233. The returns offered by breeding capacity establish the concept of sustainable fuel.

### 1.1. Neutronic Analysis of Thoria-based Fuel

Reactor physics and burnup calculation used the Serpent code that is a multi-purpose continuous-energy tool. Serpent code has exhibited a high-fidelity to decipher neutron physics in simulations. The aim of $(Th\text{-}U)O_2$ is producing the maximum amount of fissile U-233 at the end of the cycle. Serpent code can provide a kinetics neutron analysis coupled with isotopic results, also the temperature coefficient of reactivity. Figure 1 illustrates a normalized isotopic distribution during the irradiation cycle.

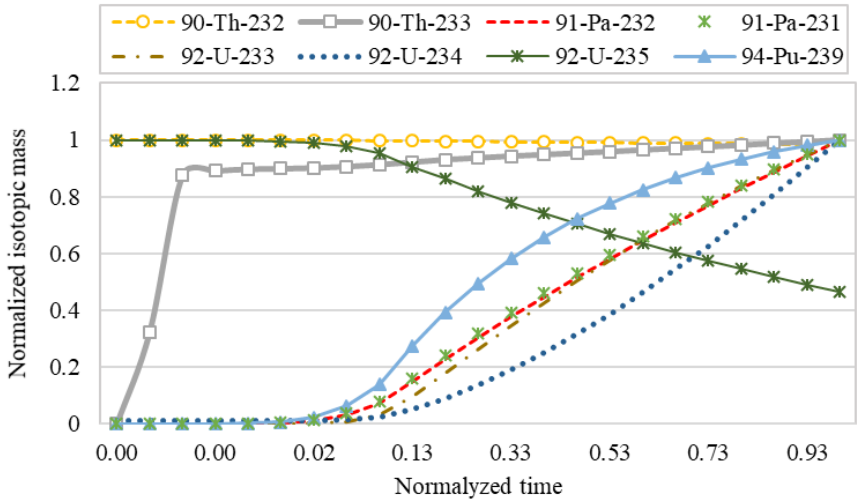


**Figure 1: Normalized isotopic concentration during irradiation cycle**

### 1.2. Fuel Performance Code

The FRAPCON is fuel rod analysis tools, also considered enough the conservative developed by the Pacific Northwest National Laboratory (PNNL) and subsidized by the National Regulatory Commission (NRC) for licensing and auditing nuclear reactors in the United States [10]. The fuel code can predict fuel performance of $UO_2$ and MOX pellets with zirconium-alloys (zircaloys) as cladding [11]. The computer code, FRAPCON, enjoys global acceptance used for fuel assessments and to foresee accident scenario behavior. FRAPCON suffered several changes to account for $(Th\text{-}U)O_2$ properties to analyze the performance of steady-state irradiation in a PWR core [12]. FRAPTRAN is a transient fuel performance code developed by PNNL to model LWR fuel behavior, up to 65 MWd/kgU [13].

Fuel codes can solve system equations via one-dimensional heat conduction through a composite cylinder, including the gap between the pellet and cladding with the finite difference method. The material property library (MATPRO) defines all thermal and mechanical features of nuclear materials at temperatures ranging from room temperature to melting temperature [14].

## 2. MATERIALS AND METHODS

### 2.1. Fuel Properties

The sintering process of $UO_2$ can improve pellet density, which is essential in the burn cycle. Density can impact heat transport, porosity level, fuel stoichiometry, and irradiation effects [15-16]. Equation 1 represents the density correlation used in composite fuel. Thoria density is 7% less dense than $UO_2$.

$$\rho(Th_{1-x}U_x)O_2 = \frac{4(M_{(UO_2)} + x(M(ThO_2) - M(UO_2))}{NA(a_{(ThO_2)} + x(a_{(UO_2)}^3 - a_{(ThO_2)}^3)} \quad (1)$$

where $M_{(UO2)}$ and $M_{(ThO2)}$ are the molecular weights of $UO_2$ and $ThO_2$ respectively, $a_{(UO2)}$ and $_{(ThO2)}$ represents the lattice parameters, $x$ represents the molar fraction of $UO_2$, and $NA$ is the Avogadro constant.

There are several correlations calculated from Vegard's law to Th-MOX fuel, are functions of $UO_2$ molar fractions. The metallurgical process can produce several levels of compression and homogenization. Figure 2 illustrates the fuel density of $UO_2$, $ThO_2$, and Th-MOX.

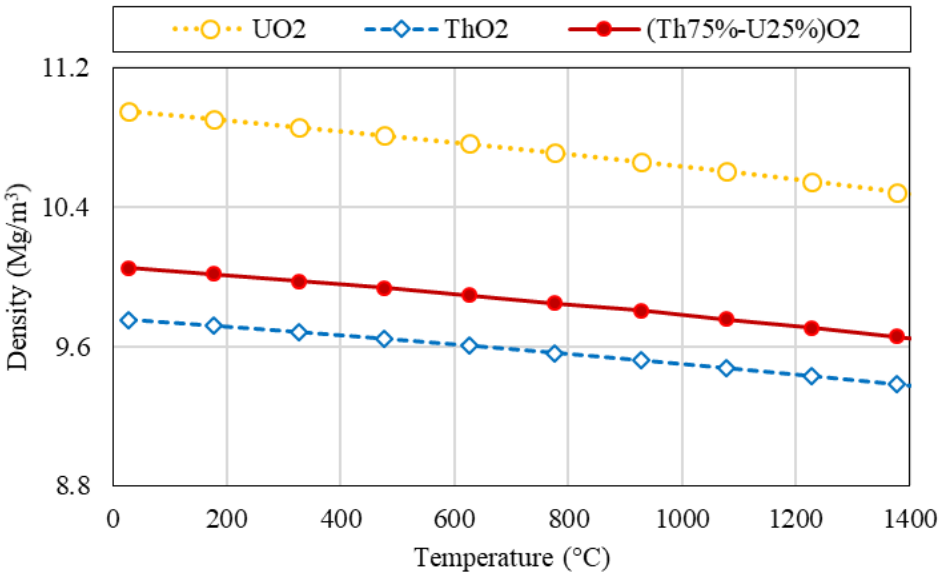


**Figure 2: Numeric fit of $ThO_2$, $UO_2$, and (Th-U)$O_2$ densities**

Fuel performance code FRAPCON defines the density of $UO_2$ of 10.96 g/cm$^3$. Theoretically, using the molecular weight of $UO_2$ and the lattice parameter of 0.54704 nm could calculate the fuel density. Equation 2 shows the correlation adapted from Belle and Berman [17]. Equation 3 displays the theoretical density of $ThO_2$-$UO_2$ fuel as a function of the weight fraction of $UO_2$. This function is valid in the temperature range of 298 to 1600 K [18-19].

$$\rho_{(T)} = 9.9981 + 0.0094(x) - 8.7463\times10^{-6}(x)^2 + 1.1192\times10^{-7}(x)^3 \quad (2)$$

$$D_{(ThO_2-UO_2x)} = 10.087 - 2.891\times10^{-4} - 6.354\times10^{-7}(x) + 5111\times10^{-6}(x)^2 \quad (3)$$

where $x$ is the molar fraction of $UO_2$.

### 2.1.1. Thermal expansion

The rule of mixtures is popular in calculating the thermal expansion of composite fuel discussing in several reports. At temperatures of 298 K, the thermal expansion coefficient of $UO_2$ is 10.2 ± 1.25 µm/m, and $ThO_2$ is 9.20 ±1.35 µm/m, estimated at 7% [20-21]. The pellet expansion will be a beneficial factor that delays the gap closure, avoiding a failure. The fuel thermal expansion FTHEXP routine used in FRAPCON code containing mixed fuel expansion properties. Figure 3 exemplifies the fractional linear thermal expansion of $ThO_2$, $UO_2$, and $(Th75\%\text{-}U25\%)O_2$.

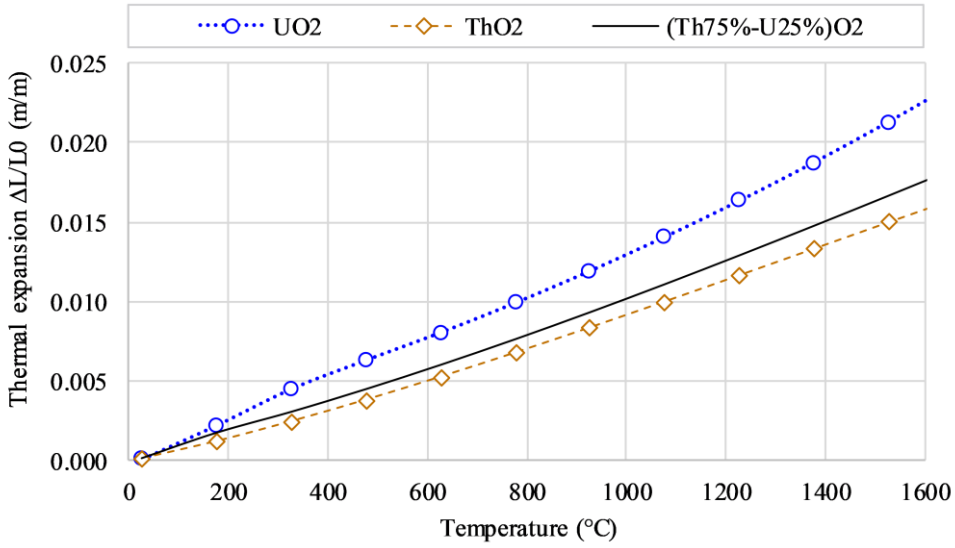


**Figure 3: Thermal expansion of $ThO_2$, $UO_2$, and $(Th_{0.75}U_{0.25})O_2$**

### 2.1.2. Thermal conductivity of (Th-U)$O_2$

Heat transfer models used in fuel codes use thermal conductivity as a critical physical property to estimate fuel temperature [9]. We find the conductivity of ceramic fuels because of the

extrapolation of the thermal capacity obtained by the increase in enthalpy [7]. Equation 4 displays the thermodynamic relationship of the thermal diffusivity.

$$k = \alpha \times Cp \times \rho \tag{4}$$

where $\kappa$ is conductivity (w/m-k), *Cp* is heat capacity (kJ/kg-K), and ρ represents density (kg/m$^3$). The density of a pellet decreases with increasing temperature for volumetric thermal expansion, while the heat capacity of fuel increases with increasing temperature. Equation 5 shows the thermal conductivity of mixed fuel.

$$\kappa_{(ThUO_2)} = \kappa_{(ThUO_2)95}\beta\left(\frac{D}{(1+0.5.(1-D)}\right) \tag{5}$$

where $\beta$, an empirical factor used to correct fuel density for $UO_2$, is 1.0789, and *D* is the theoretical density of fuel commonly estimated between 92% and 98%.

The thermal diffusivity of the pellets decreases as a function of the increasing temperature. Fuel codes use five thermal factors that have a dependence on conductivity, such as burnup, porosity, chemical contents, and theoretical density (TD) percentages [16]. Equation 6 represents a polynomial fitting for $ThO_2$, $UO_2$, and (Th-U)$O_2$ using 75% of $ThO_2$. Table 2 lists the coefficients of the polynomial regression shown in equation 6.

$$k_{(Fuel)} = A \times T^4 + B \times T^3 + C \times T^2 + D \times T + E \tag{6}$$

where, $\kappa$ is thermal conductivity in W/m-K, and the temperature is in *K*, with a valid range of 873 K to 1880 K.

**Table 2: Coefficients of the polynomial fitting used in the thermal conductivity of fuels**

| Fuels (TD) | A($\times10^{-13}$) | B($\times10^{-9}$) | C($\times10^{-6}$) | D($\times10^{-2}$) | E |
|---|---|---|---|---|---|
| K($UO_2$) | 2.6045 | -2.3586 | 8.7500 | -1.5170 | 12.2340 |
| K($ThO_2$) | 1.8897 | -1.2611 | 3.2773 | -4.0984 | 23.8470 |
| K(Th-U)$O_2$ | 1.8282 | -1.2227 | 3.1904 | -4.0022 | 23.2640 |

### 2.1.2. Enthalpy and heat capacity

The Neumann-Kopp rule (NKR) can predict the specific heat capacity of a ceramic composite fuel. In laboratory uses a calorimeter to measure enthalpy increments, which calculate proper functions adapted to heat capacity [22]. Equation 7 expresses the NKR rule used to predict heat capacity.

$$Cp_{(ThU)O_2} = Cp_{(UO_2)}(x) + Cp_{(ThO_2)}(1-x) \tag{7}$$

where *x* is the fraction of $UO_2$.

From open literature, it found that polynomial functions widely used to calculate the heat capacity as a function of temperature. Equation 5 represents polynomial coefficients used for heat capacity in kJ/kg, as a function of temperature in *K*, valid between 300 to 2000 K of the mixed fuel. Table 3 shows the polynomial coefficients of the heat capacity.

$$Cp_{(T)} = A \times T^4 + B \times T^3 + C \times T^2 + D \times T + E \tag{8}$$

where *A*, *B*, *C*, *D*, and *E* are polynomial coefficients, *T* is temperatures in *K*.

**Table 3: Polynomial coefficients of heat capacity of $UO_2$, $ThO_2$, and $(Th_{0.75}U_{0.25})O_2$**

| Polynomial coefficients | A($\times 10^{-11}$) | B($\times 10^{-7}$) | C($\times 10^{-4}$) | D | E |
|---|---|---|---|---|---|
| $UO_2$ | -3.3577 | 2.4487 | -5.6342 | 0.54170 | 122.05 |
| $ThO_2$ | -1.9591 | 1.4024 | -3.4615 | 0.37175 | 153.42 |
| $(Th_{0.75}$-$U_{0.25})O_2$ | -2.3088 | 1.6640 | -4.0047 | 0.41424 | 145.58 |

In open literature exist a considerable scatter related to the specific heat of $ThO_2$, in part because of distinct manufacture methods. The heat capacity of $UO_2$ is higher than $ThO_2$, and mixed fuel shows a fitting curve, according to NKR.

Further can observe that the heat capacity of $UO_2$ shows temperature dependence, and standard $UO_2$ fuel undergoes λ-phase transition around 2670 K. Experimental data suggest that intense investigation is required above the gamma transition point. Figure 4 displays the specific heat of the fuels analyzed.

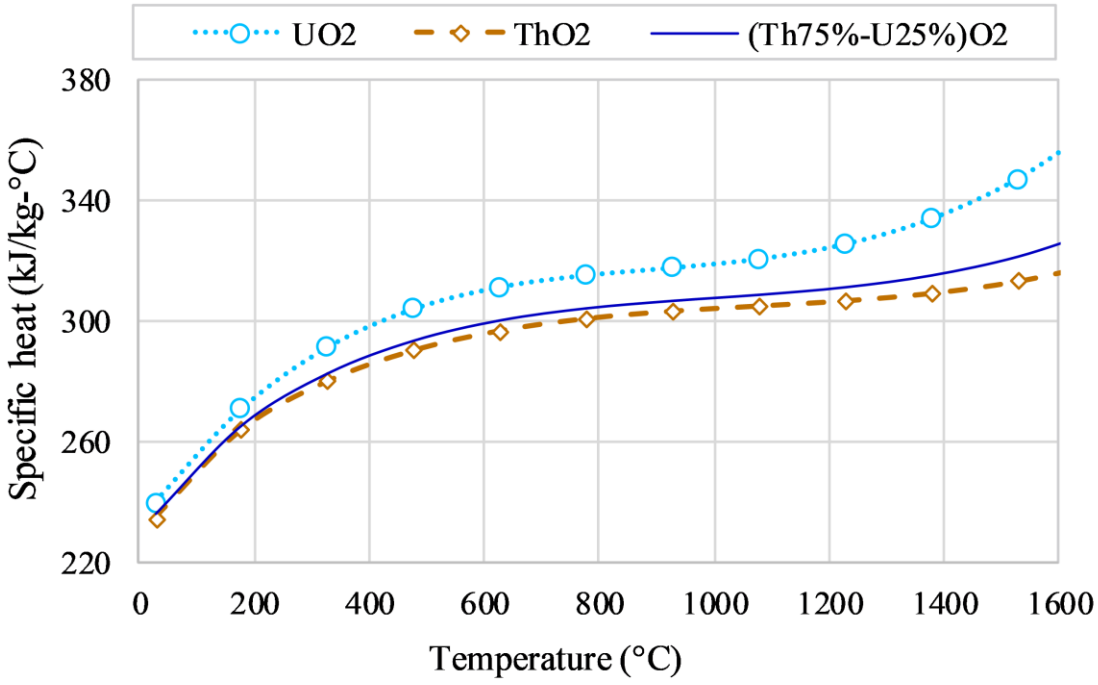


**Figure 4: Specific heat of $ThO_2$, $UO_2$ and $(Th_{0.75}U_{0.25})O_2$**

The fuel heat capacity routine FCP and fuel enthalpy FENTHL used in FRAPCON code are empirical functions. Specific heat function comprising four parameters, fuel temperature, chemical composition, molten fraction, and oxygen-to-metal (O/M) ratio. Figure 5 displays the enthalpy of the fuels investigated in this study.

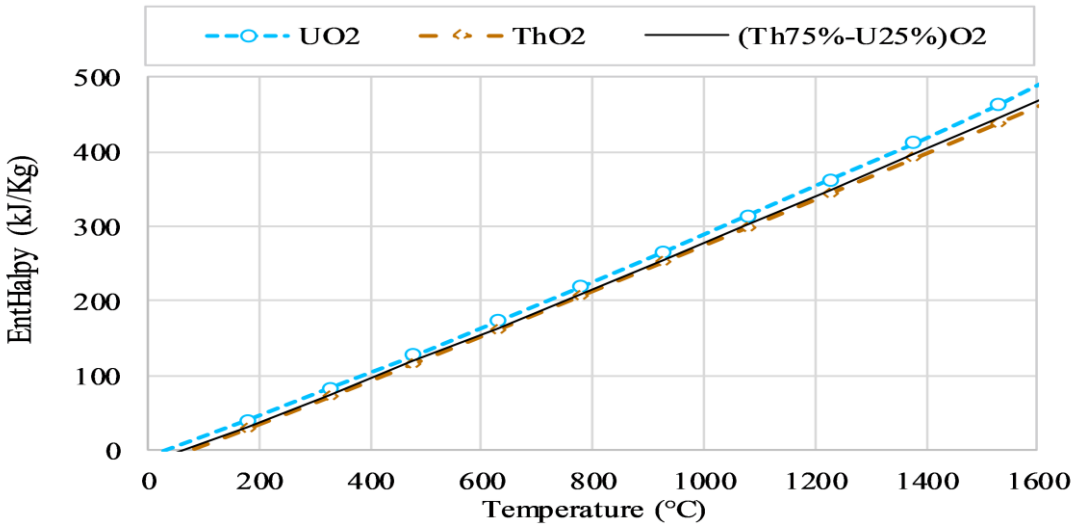


**Figure 5: Fuel enthalpy of $UO_2$, $ThO_2$, and (Th-U)$O_2$**

### 2.1.4. Radial power profile

The radial distribution of U-235 and Pu-239 is the solution of a simple diffusion model for the thermal neutron flux. The model TRANSURANUS burn-up model (TURNB) is the standard axial power profile model used for $UO_2$. Figure 6 illustrates the radial power profile of mixed fuel.

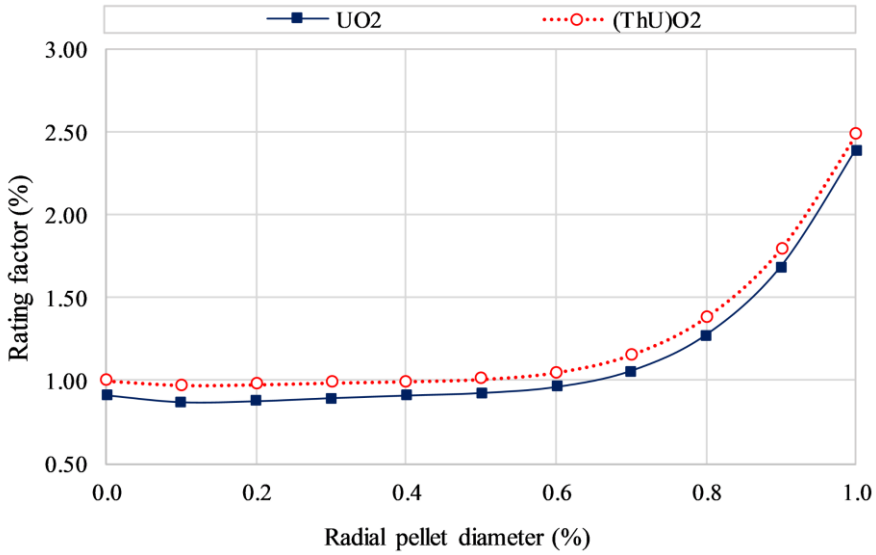


**Figure 6: Radial power profiles following the THUPS model**

TUBRNP uses the one-group diffusion approximation, but TUBRNP is imprecise when describing the radial power profile for (Th-U)$O_2$ fuel. This method does not seem accurate enough in calculations of composite fuel. Therefore, a new model adapted to calculate the radial power profile of thorium-based fuels will come from tests or simulations. The Thoria Urania Power Shapes (THUPS) model reaches satisfactory results for low and medium burnup. The significant differences between these two models are the sets of isotopes, and the implementing of an appropriated shape function because of the broader absorption cross-section of thorium and plutonium, used in the THUPS method [3].

### 2.1.5. Fission gas release model

The onset of FGR depends mostly on the diffusion rate of fission gases in the fuel matrix. Equation 9 can calculate the diffusion coefficient of a composite fuel at low temperatures, while the equation 10 displays the relationship for high temperatures.

$$D_{(ThU)O_2} = 10.57 \exp\left(\frac{-9508}{T}\right) \tag{9}$$

$$D_{(ThU)O_2} = 100^{(Bu-21)/35} \times (2.996 \times 10^{-13}) \exp\left(\frac{-26316.6}{T}\right) \tag{10}$$

where $T$ is the temperature $K$, and $Bu$ represents burnup in MWd/kgU.

The fission gas release used in simulations considers 10% of diffusion coefficients for $UO_2$, reducing FGR rates at high burnup.

### 2.2. Mechanical Models of (Th-U)$O_2$

According to Vegard's law, widely used in multiphase solid solutions showing similar crystallography, the physical properties will be a linear function based on end content fractions of the composition. Equation 11 represents the modulus of elasticity of pure $UO_2$, equation 12 shows the correlation used for $ThO_2$, and equation 11 is the combined rule for the mixture of (Th-U)$O_2$ with 75 w% of $ThO_2$.

$$E_{(ThU)O_2} = 241.29 - 3.3018 \times 10^{-2} T + 7.6415 \times 10^{-2} P.T - 564.81P \tag{11}$$

where $E$ is the modulus of elasticity in GPa, $T$ is the temperature in K, $G$ is shear modulus, and $P$ is porosity, or 1- $TD$% of fuel.

## 3. RESULTS AND DISCUSSION

Detailed full-core simulations of thorium-mixed fuel show the feasibility of use in extended operating periods exceeding 18 months, motivating to consider thorium-mixed fuels as an option in power reactors. Typically, a fuel assembly burns for three fuel cycles in core reactors, at operating periods of 18 months. Figure 7 illustrates the linear heat rate of fuel used in

simulation during the burn cycle, known as an axial power in the center of the fuel rod and at extremes.

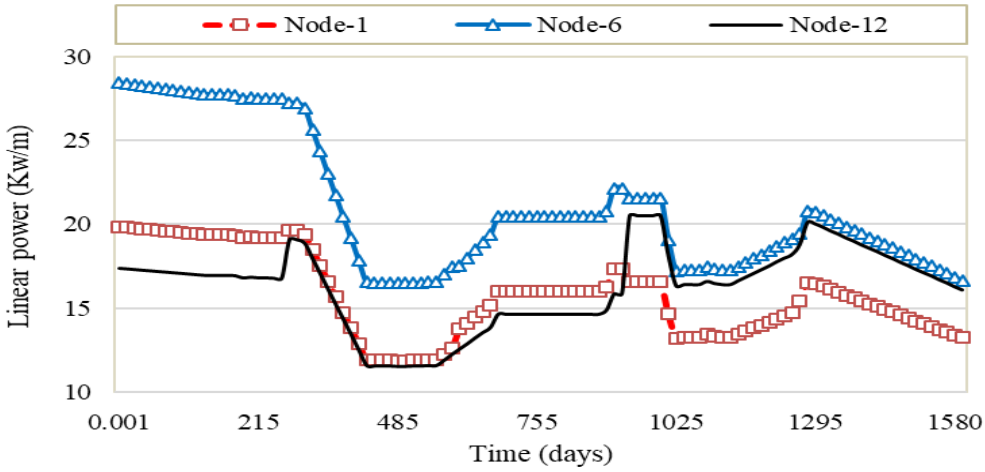


**Figure 7: Axial power used in the simulation at the center node and extremities**

The main features of PWR 17×17 with four loops are a thermal power of 3400 MW, internal core pressure of 15.53 MPa, cladding material of zircaloy, or zirlo, and an outer pellet diameter of 9.5 mm. We based simulation on the FRAPCON code with 1620 days of irradiation, reaching 66 GWd/MTU during three cycles of 16.5 months. $ThO_2$ has desirable features because of the higher thermal conductivity joined with a smaller coefficient of thermal expansion compared to $UO_2$. The higher melting point of $ThO_2$ is beneficial, at 3500 K, compared to $UO_2$ at 2800 K. Figure 8 illustrates the average fuel temperature of $UO_2$ and $ThO_2$ during reactor operation.

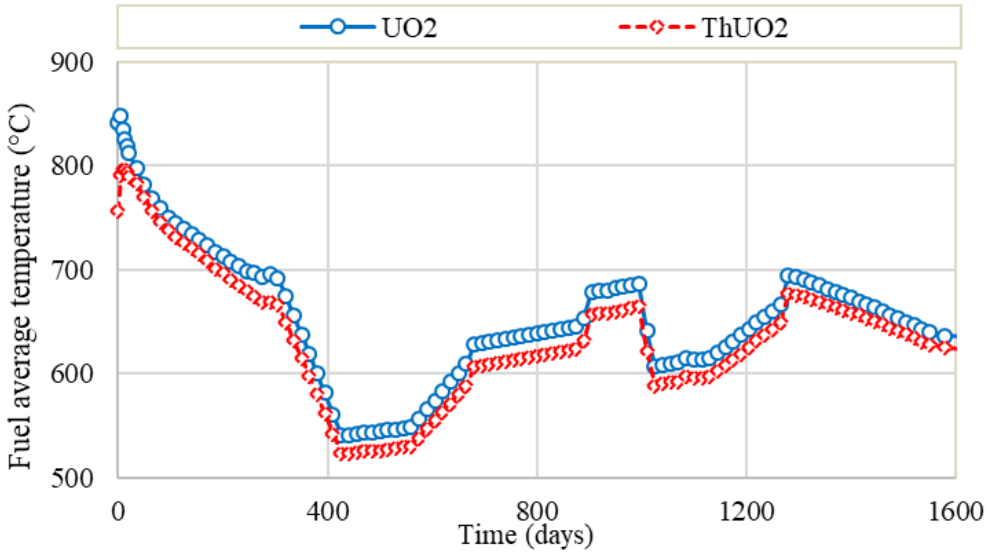


**Figure 8: Average fuel temperature of (Th-U)$O_2$ and $UO_2$**

Thermal expansion induced by anharmonic vibrations occurs in the temperature range of 725 to 1230 °C. Figure 9 illustrates the axial thermal expansion of fuel stacks under irradiation of $UO_2$ and composite fuel.

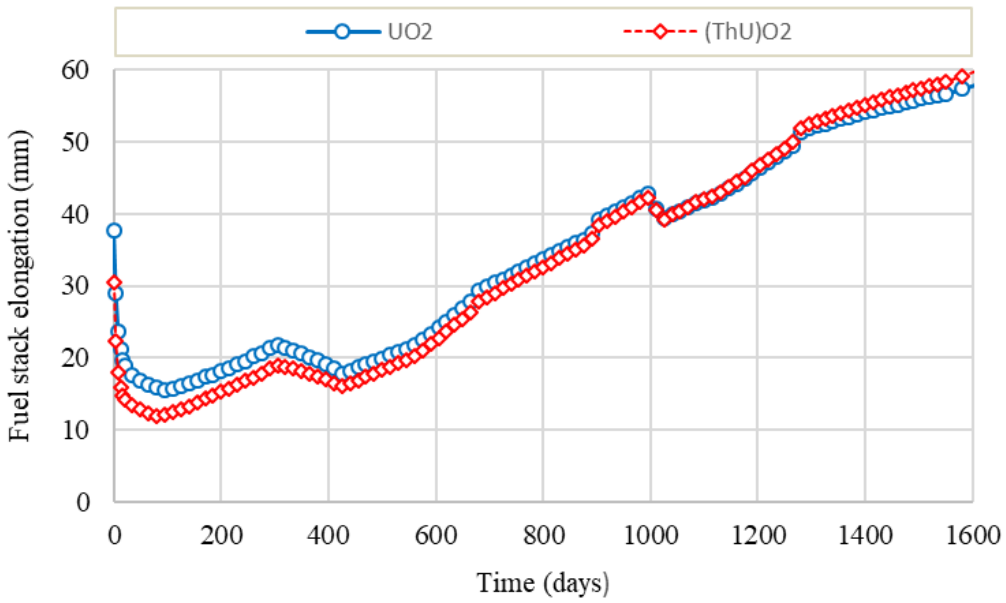


**Figure 9: Fuel stack elongation behavior of $UO_2$ and (Th-U)$O_2$**

The plenum volume accumulates from gaseous fission products and helium, while the internal plenum pressure occurs because of FGR effects, temperatures, and conductivity of gas mixtures. Thorium-based fuels should show a smaller internal plenum pressure because of reduced gaseous products generated, and the contribution of a lower fuel temperature compared to $UO_2$. Figure 10 illustrates internal plenum pressure from the simulation of $UO_2$ and (Th-U)$O_2$.

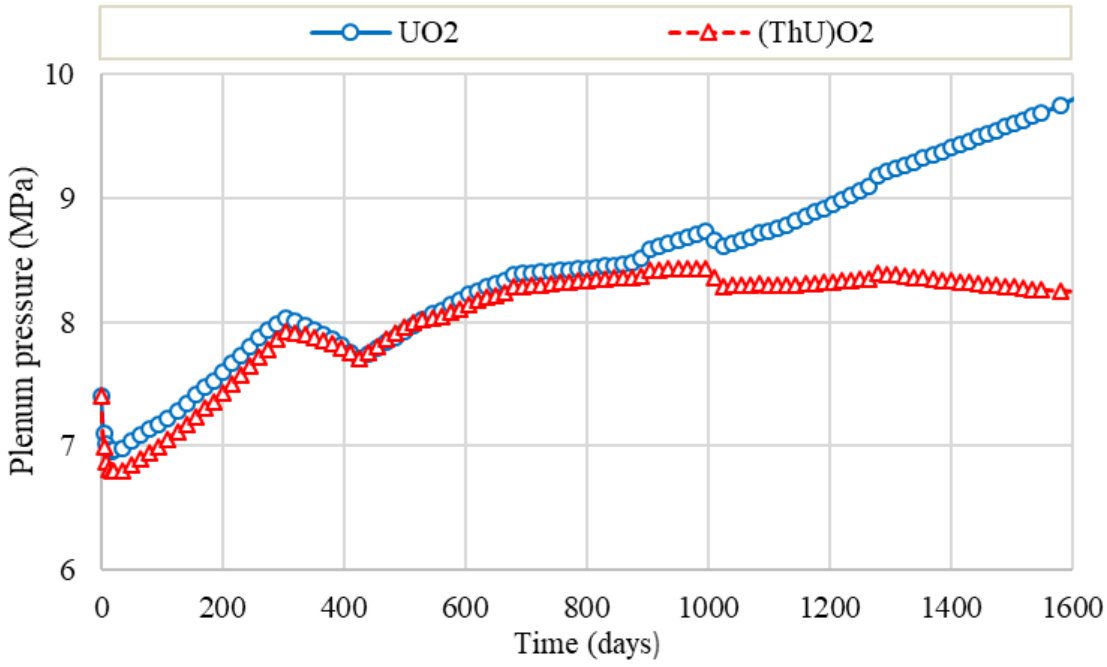


**Figure 10: Plenum pressure of $UO_2$ and (Th-U)$O_2$ fuels.**

There are several gaseous products, Xe, Kr, Cs, I, and He created during irradiation, and solid actinides contents released into fuel. The values collected from the simulations showed a reduced level of gaseous products of the composite fuel, partly because of a diffusion coefficient of $ThO_2$ is around half of the $UO_2$, and lower fuel temperatures. Figure 11 illustrates FGR of 0.5% for $(Th\text{-}U)O_2$ and 3% for $UO_2$, partly attributed to the medium average power rate of 20 kW/m.

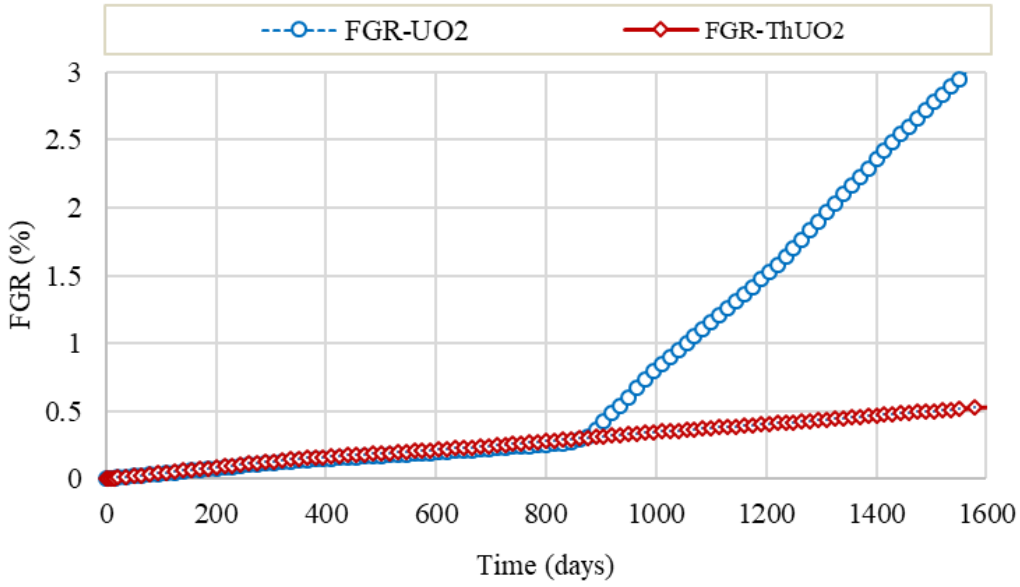


**Figure 11: Fission gas release of $UO_2$ and $(Th\text{-}U)O_2$**

## 4. CONCLUSIONS

This work showed that there is a great interest in the Thorium usage in commercial power reactors because of several beneficial factors. Thorium reserves are abundant, and there are many nations with limited uranium deposits. Many investigations proved that $(Th\text{-}U)O_2$ fueled on PWRs should extend burnup cycles compared to $UO_2$. Besides, reactors working with composite fuel may reduce plutonium proliferation. Also, it shows better radioactive waste management regarding long-lived fission products and high-level wastes. ThO2 has better chemical stability and higher radiation resistance compared to $UO_2$. Because of diffusion coefficients exhibited the mixed fuel has a reduced fission gaseous rate when compared with $UO_2$. The higher thermal conductivity and a smaller coefficient of thermal expansion are desirable features of $(Th\text{-}U)O_2$.

With the appropriated properties, including in fuel code, such as thermal conductivity, thermal expansion, heat capacity, and enthalpy increment, the FRAPCON permitted simulate the mixed fuel. The kernel of adaptation realized int the FRAPCON to support $(Th\text{-}U)O_2$ are FTHCON, FTHEXP, FCP, and FENTHL, also changed the density and melting point of the fuel. The results are compatibles reach lower fuel temperature, reduced fission products, and minor deformations.

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