

BEHAVIOR OF THORIUM PLUTONIUM FUEL ON LIGHT WATER REACTORS

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ABSTRACT

Designs using thorium-based fuel are preferred when used in compliance with sustainable energy programs, which should preserve uranium deposits and avoid the buildup of transuranic waste products. This study evaluates a method of converting uranium dioxide (UO_2) to thorium-based fuel, with a focus on Th-Pu mixed oxide (Th-MOX). Applications of Th-MOX for light water reactors are possible due to inherent benefits over commercial fuels in terms of neutronic properties. The fuel proposed, $(\text{Th-Pu})\text{O}_2$, can be helpful because it would consume a significant fraction of existing plutonium. Aside from the reactor core, the proposed fuel could be useful in existing technology, such as in a pressurized water reactor (PWR). However, licensing codes cannot support Th-MOX fuel without implementing adaptations capable of simulating fuel behavior using the FRAPCON code. The $(\text{Th-Pu})\text{O}_2$ fuel should show a plutonium content that produces the same total energy release per fuel rod when using UO_2 fuel. Thorium is a fertile material and demands a slightly higher plutonium content when used in Th-MOX. Mixed ceramic oxides show thermodynamic responses that depend on the comprising chemical fractions, and there is little information in databases on irradiation effects. The neutronic analysis is carried out using the SERPENT code to quantify transuranic production and compare this production with the original UO_2 fuel assembly. Parameters such as delayed neutron fraction and temperature reactivity coefficient are also determined. Through these analytical methods, the viability and sustainability of the proposed new fuel assembly can be demonstrated in a closed fuel cycle.

1. INTRODUCTION

Thorium-based fuels have been a subject of much research, with a wide range of applications envisioning the replacement UO_2 fuels. This investigation is focused on using thorium-based fuel mixed with plutonium dioxide (PuO_2) for use in light water reactors (LWRs), particularly as a pressurized water reactor (PWR) fuel type [1]. Naturally, thorium is four times more abundant in the Earth's crust than uranium. Therefore, in many countries, thorium might be a readily exploitable resource [2]. The interest in thorium-based fuel is growing because of the increase in uranium prices in 2007 and the decrease in popularity of uranium after the Fukushima disaster in 2011 [3]. Thorium dioxide (ThO_2) is also more chemically stable than uranium dioxide and does not oxidize as quickly. The goal of these investigations is developing a new version of the fuel performance code, known as FRAPCON, which can simulate thorium-mixed plutonium oxide (Th-MOX) fuels under steady-state conditions. This fuel technology represents the confluence of knowledge, performance, and safety margins to improve all aspects of nuclear fuels.

There are many fuel designs with varying composition, geometric configuration, and chemical contents that can make up a nuclear fuel system. This proposal investigates a solution to the problem of radioactive waste build-up, which is a significant drawback in the production of energy via nuclear power. The waste materials produced within a fuel cycle can be categorized as high, medium, or low-level, depending on the radioisotope category [4].

High-level isotopes emit alpha, beta, and gamma radiation, also including Pu-238, U-234, Np-237, and Am-241. The long half-lives of some isotopes, reaching millions of years, make this problem a crucial one to solve. Plutonium generated from U-238 neutron capture is the source of the plutonium weapon proliferation and high toxicity. Thorium-based fuels are a fertile material in nuclear fuel, also reducing large amounts of plutonium produced. The plutonium produced is around 0.77% of the weight of all uranium fuel spent. To 2018 statistics foresee that the global stockpile of separated plutonium reaches about 520 metric tons, of which about 230 metric tons is in military custody, while 290 metric tons is in civilian custody.

1.2 Fuel Performance Code

FRAPCON is a fuel licensing code developed by the Pacific Northwest National Laboratory (PNNL) and is sponsored by the Nuclear Regulatory Commission (NRC). A perfect understanding of the interaction between ThO₂, UO₂, and PuO₂ should be implemented into FRAPCON, which is the preferred fuel performance code of the NRC. FRAPCON will give insight into how fuel comprising thorium and uranium will affect fuel strain-stress response.

1.3 Serpent Monte-Carlo Code

The reactor physics analysis of (Th-Pu)O₂ fuel used Serpent code to calculate multiplication factor and concentrations of fission products. The serpent code is a three-dimensional system widely used to lattice physics that produces similar results compared to CASMO and HELIOS. Figure 1 illustrates a normalized concentration of isotopic from Serpent code for (Th 92% - U 8%)O₂.

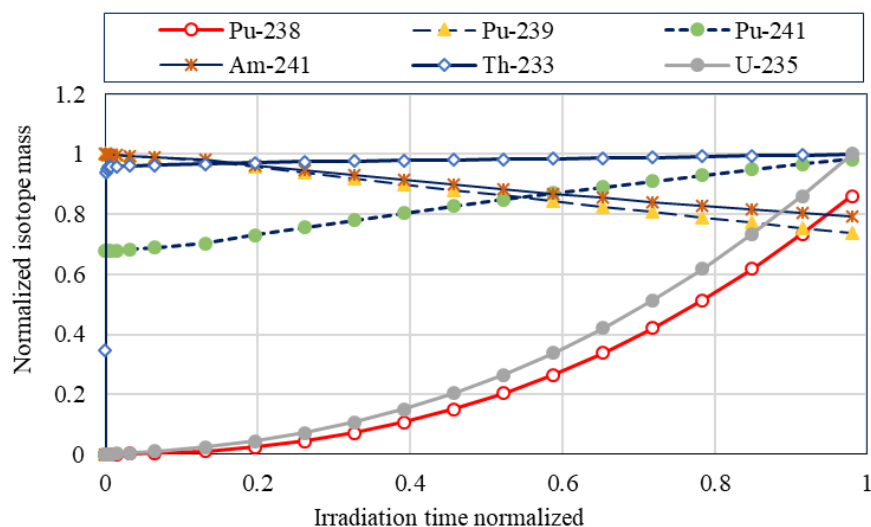


Figure 1: Normalized isotopic distribution performed with Serpent code for (Th-Pu)O₂.

2. MATERIALS AND METHODS

2.1 Fuel Modelling

Implementing mixed fuel within FRAPCON required update material models that could describe the thermophysical properties of (Th-Pu)O₂. Each property represents a subroutine of FRAPCON, such as thermal conductivity and thermal expansion, and models dedicated to swelling, densification, and fission gas release. In the mechanical models, fuel deformation is a consequence of thermal expansion, swelling, and densification. Besides, simulation considers that the pellet is a perfect cylinder with isotropic physical properties.

Plutonium is recovered from spent uranium-based nuclear fuel, and it does not occur naturally. Spent fuel produces several isotopes because of the neutron cross-section capture of ²³⁸U. One neutron capture in U-238 leads to the formation of Pu-239, typically created in nuclear reactors by transmutation. The consecutive neutron captures lead to the formation of the higher plutonium isotopes Pu-240, Pu-241, and Pu-242. Also, the production of Pu-238 results from a chain of neutron captures in U-235. Out of these isotopes, only Pu-239 and Pu-241 are fissile [5]. Figure 2 displays plutonium grades and their isotopic compositions.

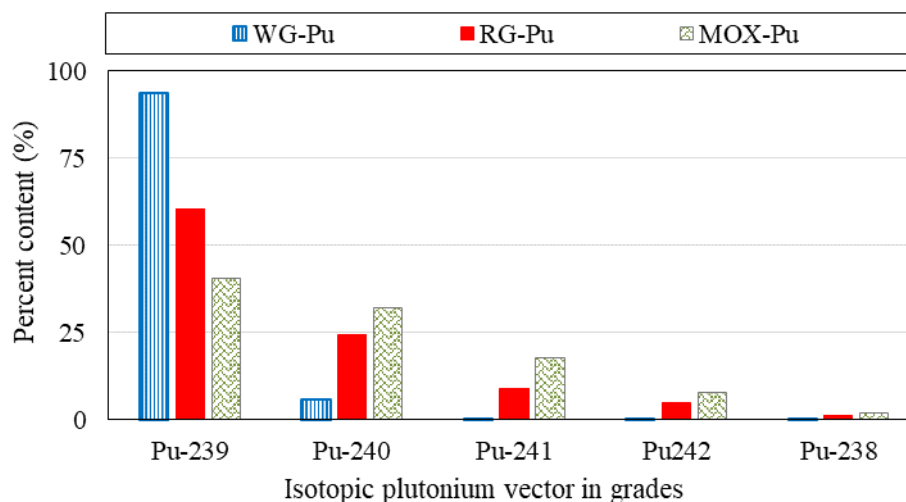


Figure 2: Isotopic plutonium composition for several plutonium grades.

The sources of plutonium are standard grades produced from spent fuels containing different isotopic concentrations. The intention is to use the grid reactor-grade plutonium (RGPu), which contains 60.30% of Pu-239, seem more suitable. However, it can use the weapons-grade plutonium (WGPu), which contains 93.80% of Pu-239, could replace the RGPu sometimes. Usually, the plutonium vector used is the WGPu that contains less than 6% Pu-240, and RGPu with about 24% Pu-240, needs to manufacture of (Th-Pu)O₂. In this investigation opted for RGPu grades, Serpent code analysis agreed that Pu-239 is fissile like U-235 and generates similar fission energy using 92% of ThO₂. The energy in (Th-U)O₂ produced by the fission of ²³⁹Pu is like of UO₂ fuel using U-235 about 4.2% of enrichment.

2.2 Fuel Properties Comparison

The composite fuel (Th-Pu)O₂ inherits properties of the weight fraction compounds, using The Kopp–Neumann (KN) law, or rule of mixtures [6]. Comparatively, the thermal conductivities and heat capacities of the composite fuels are slightly higher than standard UO₂. Table 1 summarizes several physical properties of the primary ceramic fuels discussed in this investigation [7-11].

Table 1: Properties of nuclear fuels

Physical properties of nuclear fuels	UO ₂	PuO ₂	ThO ₂
Theoretical density (kg/m ³)	10963±70	11.440±70	10.600±70
Lattice parameter (pm)	547.02±0.04	539.54±0.04	559.74±0.06
Melting point (°C)	2846±30	2663±30	3650±30
Coefficient of thermal expansion(µm/m-°C)	9.8	6.7	8.9
Thermal Conductivity (W/m-K)	8.65	6.30	14.0
Heat capacity (kJ/kg-K)	328	344	266

The thermal conductivity $k(T)$ of nuclear fuels can be calculated from thermal diffusivity $\alpha(T)$, specific heat capacity $C_p(T)$, and density $\rho(T)$, according to equation 1. Figure 3 shows the thermal diffusivity of UO₂, PuO₂, and ThO₂.

$$k(T) = \alpha(T) \times \rho(T) \times C_p(T) \quad (1)$$

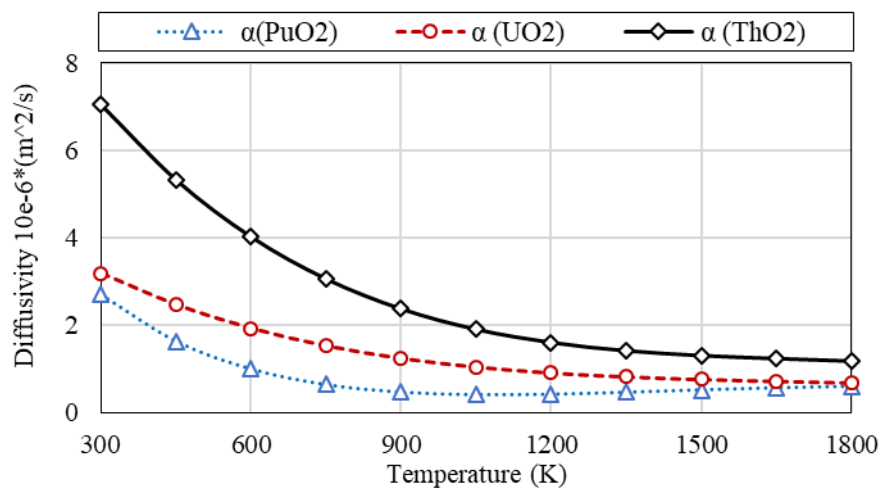


Figure 3: Thermal diffusivity of UO₂, PuO₂, and ThO₂ as a function of temperature.

Theoretically, the thermal conductivity of ThO₂ at room temperature is higher than PuO₂ because of the phonon to phonon scattering. The thermal conductivity represents the efficiency of heat transfer by conduction describing the ratio of the density coupled with the heat transferred through the material. Heat capacity is described as the amount of heat required to raise the unit temperature of the composite unit mass by unit temperature. Because of the small fraction of PuO₂, around 8% and ThO₂ 92% the property trends move toward ThO₂.

However, an increase in the PuO₂ content should cause a small, but a systematic decrease in the thermal conductivities of the composite ceramic (Th-Pu)O₂ solutions.

2.3 Thermal Properties

The primary thermal properties of interest for (Th-Pu)O₂ are thermal conductivity and thermal expansion. The thermal expansion model of UO₂ implemented in FRAPCON are functions of temperature and porosity of the fuel. The swelling model implemented in FRAPCON is burnup dependent for temperatures over 1200°C. Over 1200°C becomes much more temperature-dependent. FRAPCON uses the Material Properties Library (MATRPO) but may lack updates on the coefficients for newly released versions. This model is entirely temperature-dependent for the first 40 GWd/MTU of burnup. However, after 40 GWd/MTU, the thermal expansion of UO₂ fuel can increase. The rule of mixtures or KN formulation produces approximated results for heat capacities and thermal conductivities of (Th 92% - Pu 8%)O₂ fuel. The KN rule assumes that the capacity of a solid mixture is equal to the sum of the heat capacities of the fractions of each element. The model also needs inputs of the specific heat and the thermal conductivity relationship dependence on the temperature of pure oxides, ThO₂, and PuO₂.

2.3.1 Thermal conductivity

Figure 4 expresses the thermal conductivity as a function of temperature for UO₂, and mixed fuel containing 80% UO₂ and 20% PuO₂. Equation 2 shows a correlation using 100% of the theoretical density deduced from the conductivity of ThO₂ and PuO₂ [12].

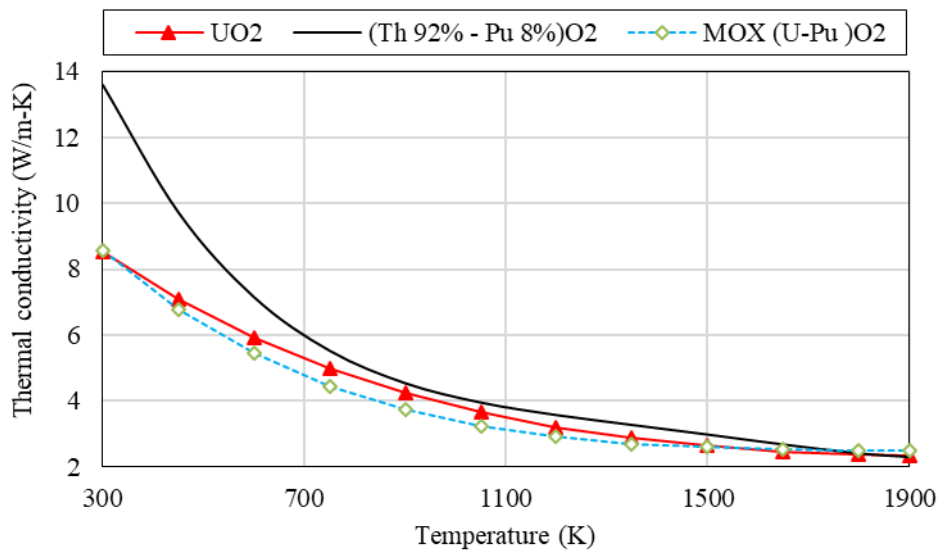


Figure 4: Thermal conductivity of UO₂, U-MOX and (Th-Pu)O₂

$$k_{(Th-Pu)O_2} = -1.079 \times 10^{-9} T^3 + 2.6901 \times 10^{-6} T^2 + 2.9103 \times 10^{-9} T + 2.362 \times 10^{-12} \quad (2)$$

Phonons and electrons are the primary mechanisms of particles or excitations that can explain the heat transport in ceramic materials, such as ThO₂ and PuO₂. Therefore, below 1500 K, a simplified model based on phonon mechanisms can be used to calculate thermal conductivity.

Table 2 lists the coefficients used to calculate the thermal conductivity of fuel using 95% of theoretical density. Equation 3 shows a correlation using both coefficients A and B [13].

$$k(\text{phonon}) = (A + BT)^{-1} \quad (3)$$

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

Table 2: Parameter used to correlation based on phonon mechanism $k = (A + BT)^{-1}$

Parameters	(Th 93%Pu 7%)O ₂	(Th 92% Pu 8%)O ₂	(Th 91% Pu 9%)O ₂
A (WmK) ⁻¹	0.029198	0.050353	0.072839
B (m/W)	0.00025789	0.00024853	0.00024853

The thermal conductivity should decrease monotonically with increasing temperature. The conductivity models calculated by phonon mechanisms suggest that phonons are excited when the temperature rises. At higher temperatures, the phonon scattering stressful the heat transport, and the conductivity suffers reductions [14].

2.3.2 Heat capacity

Heat capacity represents a safety parameter during accident scenarios because of fuel temperatures. The heat capacity of fuel represents a derivate of the enthalpy given as functions of temperature. The heat capacity of UO₂ is slightly lower if compared with PuO₂. Several reports recommended the use of the Kopp-Neumann (KN) formulation. The KN rule that is used to calculate the specific heat of composite fuels but exists other methods. However, Maxwell and Rayleigh showed expressions to measure the thermal conductivities of mixtures. The composites properties can a best-fit using KN rules or Vegard's Law that are equivalents formulations Figure 5 displays the specific heat of the contents of the mixture and (Th-Pu)O₂, following the KN rule.

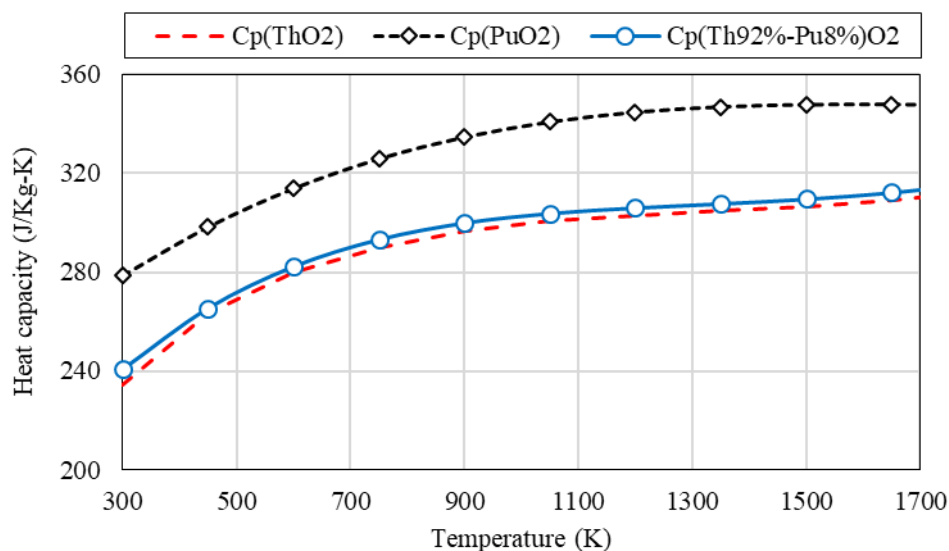


Figure 5: Heat capacity of composite fuel (Th-Pu)O₂, ThO₂, and UO₂.

Equation 4 expresses the relationship of specific the heat of the composite fuel (Th-Pu)O₂. Equation 5 displays the same relationship given as a function of temperature in Kelvin.

$$Cp(Th_xPu_y)O_2 = x.Cp(ThO_2) + y.Cp(PuO_2) \quad (4)$$

where x is the molar fraction or weight fraction of ThO₂, and y is the fraction of PuO₂.

$$Cp(Th-Pu)O_2 = 5.5187 \times 10^{-8} T^3 - 0.00021776 \times T^2 + 0.29392 \times T + 171.93 \quad (5)$$

The FRAPCON code calculates a temperature profile for the coolant and fuel rod of each axial node. The implemented models assume that the fuel rod is a right circular cylinder surrounded by coolant. The heat conduction model adopted by FRAPCON only considers the radial direction because of the sizeable length-to-diameter ratio. Because of this assumption, fuel codes ignore axial heat conduction.

2.3.3 Thermal expansion

Thermal expansion coefficients and specific heats of multiphase materials depend on the properties of the comprising materials. For two-phase composite ceramics, expansion capacity has limits that coincide with the relationship between the unique thermal expansion coefficients and modulus of elasticity. Linear heat rate is a parameter that can control fuel temperature and fission gas release. The addition of PuO₂ on a ThO₂ matrix reduced the thermal conductivity in fuel pellets. However, fuel expansion shows several implications, such as an interaction between pellet fuel and zirconium-based alloys. The coefficient of thermal expansion at room temperature of PuO₂ is 7.8 μm/m-°C, of ThO₂ is 8.9 μm/m-°C, and UO₂ is 9.76 μm/m-°C. Using Vegard's law found that (Th-Pu)O₂ should reduce thermal expansion if compared with UO₂. Figure 6 illustrates the thermal expansion of composite fuels.

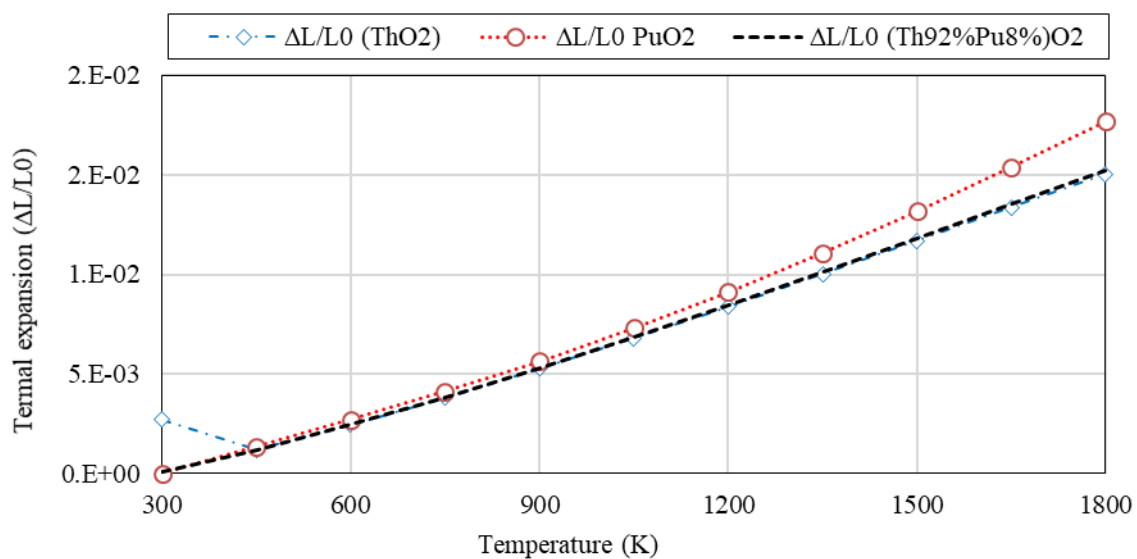


Figure 6: Thermal expansion of ceramic oxides ThO₂, PuO₂, and (Th-Pu)O₂.

2.4 Fission Gas Release

In the FRAPCON code, the diffusion coefficients are a crucial parameter used in the Massih model. The diffusion coefficients for UO_2 within the valid temperature range of 773 to 2300 °C, the coefficient should vary from 10^{-21} to 10^{-16} (m^2/s). The fission gas release of ThO_2 depends on the diffusion coefficient of ThO_2 that represents about 10% of UO_2 . If fuels UO_2 and PuO_2 exhibited a stable fission gas yield, varying of 0.25-0.30%. Xenon is the stable gas the most substantial fraction, approximately 90% of Xenon and 10% of Krypton. A different response occurs with thorium-based fuel because of the effects produced by U-233. The $(\text{Th-Pu})\text{O}_2$ mixed fuel shows similar fission effects of U-233, exhibiting around 82% Xe and 18% of Kr. The fission of U-233 results in 10% more gaseous atoms per fission. Thorium-based fuels must have a lower gaseous diffusion coefficient, which decreases to half the order of magnitude used in UO_2 .

2.5 Mechanical Models

In this study, a molar fraction of PuO_2 8%, and ThO_2 92% used for $(\text{Th-Pu})\text{O}_2$. However, ThO_2 shows insufficient data to predict fuel cracking and relocation to account for creep in the proposed model. The empirical relations used to mix fuel may differ because of the metallurgical methods used to manufacture Th-MOX fuels. The $(\text{Th-Pu})\text{O}_2$ fuel assumes that the swelling models are like the UO_2 model, and diffusion coefficients are trending towards ThO_2 . However, solid and gaseous fission products should show a dependence on burnup. UO_2 loss conductivity and strength response for radiation damage effects, which intensify fragmentation, relocation, and consequentially reduce the thermal conductivity [15]. Equation 6 expresses the modulus of elasticity of ThO_2 .

$$E(\text{ThO}_2) = 2.491 \times 10^{11} (1 - 2.21P)(1.0230 - 1.405 \times 10^{-4} \exp(181/T)) \quad (6)$$

where $E(\text{ThO}_2)$ represents the modulus of elasticity in Pa, P is the fuel porosity of around 5% for fresh fuel, and T is the temperature in K.

The elastic modulus at room temperature of ThO_2 and PuO_2 are respectively 239.88 ± 10 GPa and 257.50 ± 15 GPa. The Poisson coefficient used in the FRAPCON code for UO_2 is 0.316, which does not consider temperature dependence. The Poisson ratio is independent of temperature for ThO_2 . The Poisson ratio for ThO_2 is 0.2 and 0.30 for PuO_2 . Equation 7 represents the modulus of elasticity of composite fuel.

$$E(\text{Th}_x\text{-Pu}_y\text{O}_2) = E(\text{ThO}_2) (1 + 0.0284y) \exp(-1.75x) \quad (7)$$

3. RESULTS AND DISCUSSION

The theoretical exercise performed in this study has as a basis the FRAPCON code, using physical properties of $(\text{Th-Pu})\text{O}_2$, with 92% ThO_2 . The case of interest is a four-loop PWR 17x17 that can produce 3853 MWTh rated thermal power. Industrial assembly designs informed by fuel suppliers contain 264 fuel rods and 25 positions to reactive dynamics control with a current length of 3.65 m.

The core reactor shows three enrichment regions, which shows a progressive fuel enrichment of 2.35 w/o, 3.40 w/o, and 4.45 w/o. The simulation is reproducing a civilian power reactor, with an irradiation period of 1476 active power days, and reaching about 62 GWd/MTU during three cycles of 16.5 months. Table 3 shows the configuration PWR 17X17 with fuel designs used in the fuel performance FRAPCON.

Table 3: Manufacturing parameters of PWR fuel adopted to FRAPCON code

Fuel parameters	Values
Pellet outside diameter (mm)	8.19
Pellet high (mm)	9.83
Fuel density (% of theoretical)	95.50
Rod pitch (mm)	12.60
Cladding outside diameter (mm)	9.50
Cladding thickness (mm)	0.57
Radial gap (μm)	82.55
Total rod length (m)	365.76
Internal pressure (MPa)	15.51
Channel equivalent diameter (mm)	11.8

The cladding material initially planned can be the ZIRLO™, and all standard properties from fuel suppliers are fully compiled into FRAPCON. The pellet has an outer diameter of 9.5 mm. The thermodynamic parameters are common values, such as the reactor operating at a pressure of 15.51 MPa. The coolant inlet temperature is 279 °C, with an outlet temperature of 324 °C. Figure 7 displays the axial power used in the simulation.

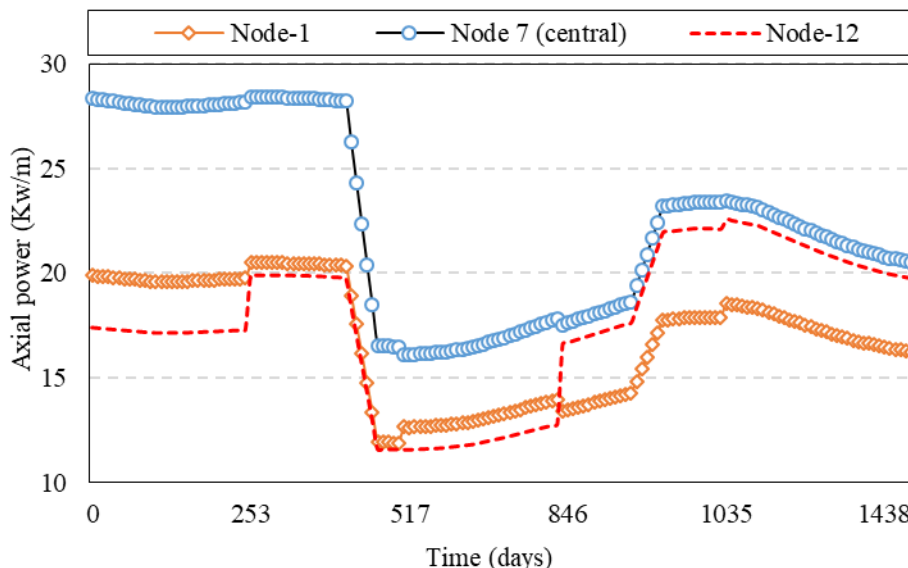


Figure 7: Axial power applied under fuel rod using 12 nodes uniformly distributed.

An experimental series conducted in the Halden reactor in Norway, rod-3 of the IFA-730 test, reported similar results when compared with the FRAPCON code changed to $(\text{Th-Pu})\text{O}_2$ [16].

Test rod-3 used a composite fuel containing 92.1% of Th and 7.9% of Pu, which was sintered at 1650 °C with a hold time of 6 hours. The sintered pellet reached a theoretical density of 97%, with an outer diameter of 5.9 mm. The total active length of the fuel rod is 3.65 m with 12 axial nodes. The (Th-Pu)O₂ fuel was subject to a radiation cycle of 300 days, completing a burnup of 25 GWd/MTU [16]. Figure 8 illustrates the fuel temperatures.

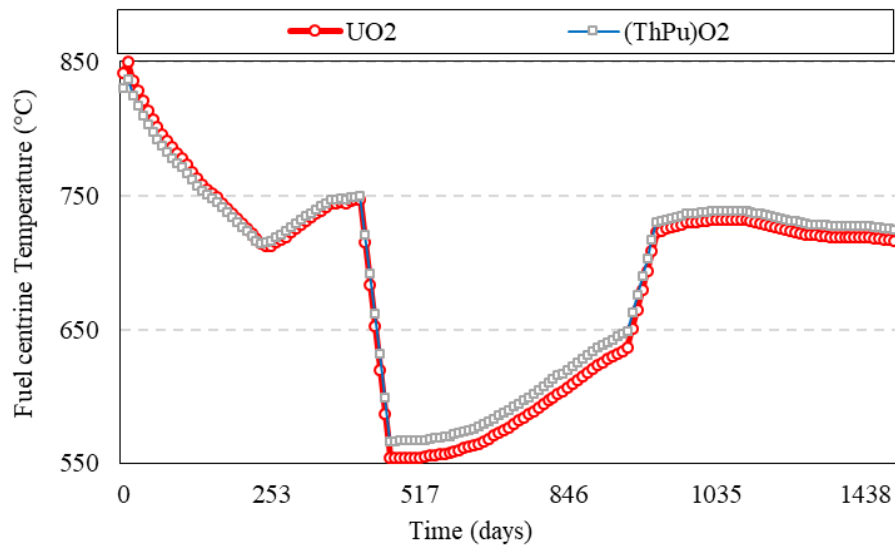


Figure 8: Fuel centerline temperature from UO₂ and (Th-Pu)O₂

The axial power density can explain power curves at the start of the cycle because of the asymmetric shape used. The fresh fuel power peak factor is around 1.4 localized in the middle of the rod. Following the power peak factor, drop to 1.2 after six months. Nearly to the end cycle when fuel reaches 12-month power peak drops to a level of 1.1

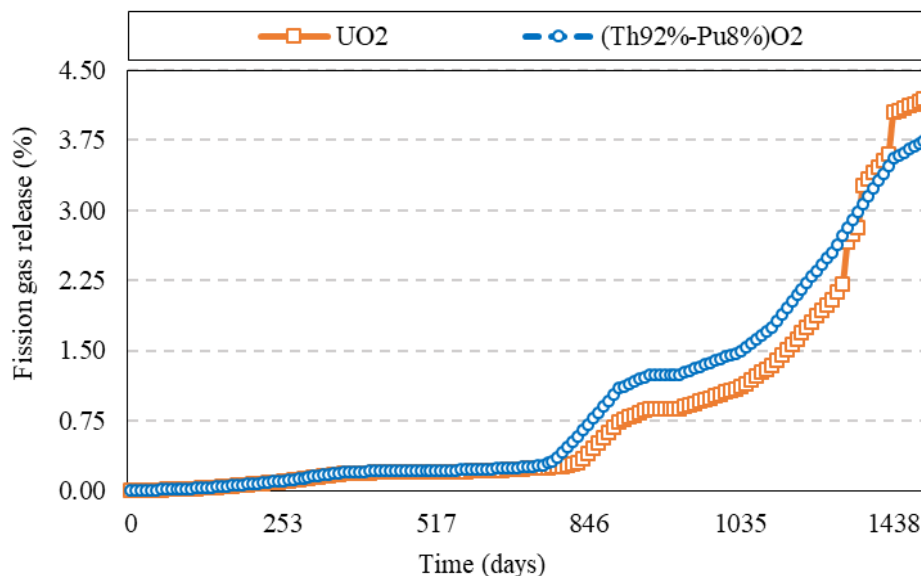


Figure 9: Fission gas release from UO₂ and (Th-Pu)O₂

The mixture of ThO₂ and other actinides can produce a composite material exhibiting inhomogeneous physical properties, in part because of the sintering process. Fission fragments increase under irradiation causes a rapid loss of mechanical properties. Figure 9 shows the fission gas release produced by (Th-Pu)O₂, compared with UO₂. Rapid temperature changes, such as experienced during start-up and shutdown conditions, cause the fuel pellet to crack. Fuel cracking is a consequence of the thermal hoop stresses and differential thermal expansion generated across the fuel pellet.

Fission gas released model is the safety parameters investigated coupled with fuel temperature, which can specify limit the lifetime of fuel. Therefore, the gaseous release depends mostly on the diffusion rate of fission gases in the fuel matrix. The diffusion coefficient of ThO₂ is about 10% at low temperatures and 50% at higher temperatures if compared with UO₂.

4. CONCLUSIONS

The plutonium vector used in this study was RGPu, widely considered the most common, recovered from PWR spent fuels. The fuel performance code FRAPCON should contain a harmonious integration of the physical models used to ThO₂ and PuO₂. The mixed fuel properties came from the physical features of each content based on practical rules using the Koop-Neumman rule and Vegard's Law. The Serpent code is a Monte-Carlo code used to calculate the fission products and isotopic distribution. Also, the Serpent foresaw the radial power profile of fuel assemblies. Serpent code analyzed fuel and reactor core behavior regarding neutron kinetics and fuel burn cycle used.

The simulations performed with FRAPCON showed results compatible with the features estimated on IFA-730 tests. The set of routine updates, such as FTHCON, FHTEXP, FCP, FENTL, MASSIH, which are the kernel of code adaptations. The mechanical response suffered updates such as the modulus of elasticity, Poisson coefficient, and shear modulus. However, the combination of all physical models into FRAPCON needs efforts because of irradiation effects. Results from simulation can summarize that the safety margins exhibited are comparable to the behavior of UO₂. Finally, we can conclude that (Th-Pu)O₂ could replace UO₂ with slight restrictions proposed to fuel designs.

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