

Advanced Nuclear Fuels Based on Thorium Mixed Oxides

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1. Introduction

Thorium-based fuels are a more sustainable nuclear fuel for electricity generation. Exist analysis of composite fuel (Th-U)O₂ used in many types of nuclear reactors. In this study, the physical properties and models of advanced fuel systems formed ThO₂ 75 wt.% and UO₂ 25 wt.%, which worked with 19.5% enrichment of U^{235} . It analyzed the physical properties of mixed fuels using the composition of mixtures, such as the lattice parameters, thermal conductivity, specific heat, mechanical strength, and fission gas release. The codes FRAPCON-4.0 and FRAPTRAN-2.0 adapted can calculate the composite fuel response compared with uranium dioxide fuel used for light water reactors. In addition, the increased diffusion coefficient produced lower fuel swelling compared with UO₂.

Thorium fuels had included an extensive range of applications, such as pressure-tube heavy water reactors (HWRs), light water reactors (LWRs), and thorium molten salt reactors. Advanced reactors, such as sodiumcooled fast reactors, can support the thorium mixed oxide fuel [1]. Early the Shippingport reactor, in Pennsylvania, USA, was a light water breeding reactor that operated with thorium as fuel during 1977–1982 [2]. Thorium is at least three times more abundant than uranium [3]. The natural isotopic distribution of thorium is 100% of Th²³² and is not fissile, but it is a fertile material, like U²³⁸. Thorium requires fissile materials, such as U²³⁵ and Pu²³⁹, to begin the reaction. Today, exist a few mixed fuel cycles based on thorium use (Th²³²+Pu²³⁹), (Th²³²+U²³³), (Th²³²+U²³⁵), and other formulations, including dopant additions [4]. On the other hand, it researches innovative fuels, such as uranium nitride (UN) and uranium carbide (UC), which have several advantages over UO₂, such as increased burnup capabilities and higher thermal conductivities [5]. Therefore, The UN and UC have restrictions on underwater environments. The physical properties of ceramic fuels at 293.15 K and 0.1 MPa are listed in Table I. Thorium fuel stands out for its high thermal conductivity.

Physical Properties	UO ₂	PuO ₂	ThO ₂	U ₃ Si ₂	UC	UN
Density (g/cm ³)	10.96	11.46	10.30	12.2	13.63	14.30
Thermal Conductivity (W/K)	8.68	6.3	14.07	8.08	25.3	13.0
Thermal Expnsion (µm/m)	9.76	7.8	9.43	15.5	10.1	7.2
Heat Capacity (J/kg K)	235	240	234	201	200	190

Table I: Physical properties of traditional fuel materials used for thermal reactors

This study included a simulation of mixed ceramic oxides using (Th-U)O₂ and (Th-Pu)O₂ using FRAPCON code. The strategic composition utilized (Th-75 wt%, U-25 wt%)O₂, homogeneously distributed into core reactor, with U^{235} enrichment of 19.5%. On the other hand, exist (Th-92 wt%-Pu-8 wt%)O₂ that improve Pu destruction rates. Also, any Pu is generated from thorium. (Th-Pu)O₂ has higher thermal conductivity, and reactivity may use burnable poison due to higher reactivity at begin. Therefore, Pu-239 is a result of reprocessing of spent nuclear fuel. Plutonium grades depend on reactor type and burnup cycle. In PWRs plutonium grades achieving 56% Pu²³⁹, 23% Pu²⁴⁰, 12.8% Pu²⁴¹ and 5.3% of Pu²⁴². The weapon-grade produced at low burnup by reduction extraction process to separate and show until 93.8% of Pu²³⁹. Early, the manufacturing route used to mix thorium fuels used the traditional powder route.

Over the years, options arose, such as pellet impregnation, sol-gel microsphere palletization, and vibratory compaction or Vi-pack.

2. Methodology

The FRAPCON fuel code can predict the fuel performance of UO_2 and $(U-Pu)O_2$ pellets coated with zirconium alloys (Zircaloys) [6]. Thus, FRAPCON must update to account for the $(Th-U)O_2$ and $(Th-Pu)O_2$ physical properties to analyze steady-state irradiation performance in a typical PWR 17×17 .

Composite fuels would show thermal properties as intermediates between the pure contents if the production route produced a homogeneous system [7]. Predict the physical properties using an empirical rule of mixtures or Vegard's law, which assumes that components have a similar crystal structure. Vegard's law is a practical approach resembling the Koop–Neumann rule (KNR) to compile mixture properties. Thermal and mechanical properties, such as the melting point, linear coefficient of thermal expansion, thermal conductivity, and creep rate, are widely used in fuel performance codes. The library of properties of FRAPCON has subcodes for each physical feature. Composite fuel has physical properties that intermediate between the pure contents, but it must assume the metallurgical route to produce a homogeneous system.

The thermal conductivity of ThO₂ is 10% higher than that of pure UO₂ at around 1000 K, coded in the FTHCON subroutine. FTHCON computes burnup effects, porosity, fission products as Xe and Kr [8],[9]. However, the impact of Pu fraction variation is much more substantial for $(Th-Pu)O_2$ than $(Th-U)O_2$ because PuO₂ contents become hyper stoichiometric. Therefore, the linear thermal expansion of ThO₂ is slightly lower than UO₂ described in FTHEXP using an interpolation across a compositional range based on Vegard's law [9]. Volumetric changes affect the pellet-to-cladding gap size, which is a safety parameter. Figure 1 shows the thermal conductivity of mixed fuels.



Figure 1: Thermal conductivity of UO₂,(Th75%-U25%)O₂ and (Th92%-Pu8%)O₂.

A few subcodes FCP and FENTHL describe the specific heat capacity and enthalpy of thorium fuels have the same form, based on the Koop Neumann rule. The thorium has lower heat capacity, and enthalpy than UO_2 and PuO_2 and no exist burnup dependence. The subcodes containing elastic properties include new correlations, such as FELMOD calculates Young's Modulus and FPOIR for Poisson ratio.

But Young's modulus of PuO_2 and ThO_2 fuels are slightly superior to UO_2 , and PuO_2 moduli achieve about 2.84% larger than ThO_2 . The Poisson ratio of UO_2 is 0.316 higher than both ThO_2 and PuO_2 , which have the same numerical value of 0.28.

The radial distribution of power adopted in the FRAPCON code is the TRANSURANUS Burn-up (TUBRNP) model used for UO₂ to calculate the local concentration of actinides, the primary fission products [10]. The substitute for TUBRNP is the Thoria Urania Power Shapes (THUPS) model. THUPS includes a distinct set of isotopes and an appropriate shape function.

3. Results and Discussion

To simulate the performance of the (Th-U)O₂ and (Th-Pu)O₂ fuels, using the parameters of the PWR AP1000 reactor, a 17×17 configuration type, capable of generating 1117 MWe. Table II shows fuel parameters used for simulation.

Parameter	AP1000-system
Fuel (planed UO ₂)	$(Th-U)O_2/(Th-Pu)O_2$
Fuel rod outer diameter (mm)	9.5
Pellet outer diameter (mm)	9.83
Pellet length (mm)	0.082

Table II: Fuel parameters based on AP-1000 reactor design

Fuel centerline temperatures can reduce by 2-8% for (Th-U)O₂ mixed fuel and result in enhanced thermal conductivity of ThO₂ compared with UO₂. During irradiation, produce gaseous fission products, such as Xe, Kr, Cs, I, and He. (Th-Pu)O₂ shows a similar temperature to UO₂. Figure 2 shows fuel centerline temperature for 1200 days.



Figure 2: Comparison of fuel center temperature during the full burn cycle.

4. Conclusions

Both options used (Th-75 wt.%-U-25 wt.%)O₂ and (Th-92 wt%-Pu-wt.%)O₂ showed beneficial for reducing plutonium risk. Therefore, constraints exist related to the plutonium grades used and configuration based on the homogenous distribution of fuel. The fuel centerline temperature is compatible with UO₂ for (Th-Pu)O₂. The most key features of pure ThO₂ for the safety analysis are the enhanced thermal conductivity, higher melting point, and lower thermal expansion. These beneficial features permit a longer burn cycle and operation at a higher power level, reducing the FGR compared with UO₂. In addition, isotopes U²³³ and Th²³² have detailed responses in terms of core reactivity. Reactivity control depends on which fission isotope is predominant, U²³⁵, U²³³, or Pu²³⁹, and the time response of U²³³ makes dynamic fission control possible with little difficulty. Thorium has the advantage of lower radiotoxicity but produces a reduced amount of U²³², an emitter of gamma rays. ThO₂ spent fuel has the benefit of chemical inertness. In the past decade, there has been a growing risk of nuclear proliferation. Plutonium is a fissile isotope eliminated after a chain reaction, which minimizes the risk of making weapons with it. Using thorium has economic advantages because of lower and stable international prices and substantial natural reserves spread worldwide.

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References

- [1] Y. Long, Modeling the performance of high burnup thoria and urania PWR fuel. Doctoral thesis, Massachusetts Institute of Technology (PhD-MIT), (2002).
- [2] IAEA, Thorium Fuel Cycle-Potential Benefits and Challenges. IAEA TECDOC Series No. 1450. International Atomic Energy Agency, (2005).
- [3] D. S. Gomes, G. S. Laranjo, F. B. V. Oliveira. Analysis of a pressurized power reactor using thorium mixed fuel under regular operation. In: INAC, Oct. 21-25. pp. 4996-5009, (2019).
- [4] D. S. Gomes, A. T. Silva, F. B. V. Oliveira, G. S. Laranjo. The Behavior of thorium plutonium fuel on light water reactors. In: INAC, October 21-25, Santos, SP, pp. 4984-4995, (2019).
- [5] Z. Xu, Design strategies for optimizing high burnup fuel in pressurized water reactors. Massachusetts Institute of Technology, (2003).
- [6] K. J. Geelhood, W. G., P. Luscher, I. Raynaud. Porter, FRAPCON-4.0: A computer code for the calculation of steady-state. Thermal-Mechanical Behavior of Oxide Fuel Rods for High Burnup, (2015).
- [7] A. K. Nayak, B. R. Sehgal, Thorium–Energy for the Future. Springer Singapore, (2019).
- [8] International Atomic Energy Agency. Thermophysical properties database of materials for light water reactors and heavy water reactors. IAEA TECDOC, 1496, (2006).
- [9] Y. Long, Y. Yuan, M. S. Kazimi, R. G. Ballinger, E. E. Pilat. A fission gas release model for high-burnup LWR ThO₂-UO₂ fuel. Nuclear Technology, 138, pp. 260-272, (2003).
- [10] A. J. Mieloszyk, M. S. Kazimi. Fuel performance analysis of a (ThU)O₂-fueled, reduced moderation boiling water reactor. Nuclear Technology, 191, pp. 268-281, (2015).