



## Assessment of the UO<sub>2</sub>-Graphene Composite Fuel Proposed for Nuclear Reactors

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### 1. Introduction

Uranium dioxide (UO<sub>2</sub>) is the ceramic nuclear fuel technology currently used in light water reactors (LWRs). Since the Fukushima Daiichi disaster, international efforts to avoid accidents have involved searching for more tolerant fuels. The solution proposed in this study comprises the dispersal of graphene nanoparticles with high thermal conductivity throughout the UO<sub>2</sub> matrix, using the addition of a second-phase material with enhanced physical properties. A considerable thermal gradient can be created using a standard pellet fuel based on UO<sub>2</sub> and treated as a thermal insulator. Tolerant fuels must exhibit a low thermal gradient, coupled with good reactivity in the reactor core. The UO<sub>2</sub>-graphene composite shows a minor impact on the moderate neutron flux. The sintering method works with a powder mixture of graphene, and UO<sub>2</sub> powder manufactured quickly using spark plasma sintering (SPS).

In light water reactors (LWRs), 443 are currently in operation, and 51 are under construction, producing about 393.241 GWe. Over the last two decades, studies have focused on enhancing the thermal conductivity of UO<sub>2</sub>. They are using techniques based on adding a second phase dissolved in the UO<sub>2</sub> fuel matrix, such as beryllium oxide (BeO), graphite, or silicon carbide (SiC). UO<sub>2</sub>-BeO with 9.6 vol.% BeO can increase thermal conductivity by 10% [1], while experiments employed UO<sub>2</sub> with 10 vol.% SiC have achieved 30% of increasing [2]. Composite fuels with the addition of nanoparticles such as carbon nanotubes (CNTs), UO<sub>2</sub>-SiC, UO<sub>2</sub>-diamond, and UO<sub>2</sub>-graphene have several experiments [3]. However, under irradiation, the addition of SiC resulted in enhanced cracking, and though UO<sub>2</sub>-diamond had shown a lower fission gas release (FGR). Accident-tolerant fuel (ATF) is a term used to describe a technical concept that further enhances nuclear fuel safety and performance. The U.S. Department of Energy's (DOE) Office of Nuclear Energy (N.E.) awards funding to industry partners, universities, and national labs to develop ATF plans. The global nuclear market comprises pressurized water reactors (PWRs), representing 64%, and boiling water reactors (BWRs), with 18% of all reactors. It most increases the thermal conductivity to minimize the fuel pellet cladding mechanical interaction (PCMI), using additives, such as graphene or SiC to achieve this.

UO<sub>2</sub>-graphene can enhance thermal conductivity by 33% with 10 vol.% graphene. It incorporates the mechanically robust graphene nanoplatelets into the UO<sub>2</sub> fuel matrix to improve its thermal-mechanical properties. High-density UO<sub>2</sub>-SiC and UO<sub>2</sub>-diamond composite fuel pellets produced on a small scale by spark plasma sintering (SPS)

The Sintering, the mixture of graphene nanoparticle additions to UO<sub>2</sub> powder, uses the SPS method [4-5]. Thus, the UO<sub>2</sub>-graphene pellets were prepared by SPS at 1500 °C for 5 min under an applied pressure of 40 MPa in a vacuum. SPS generates grain sizes of 12 ± 1 μm, compared with the typical grain size range of 10–15 μm attained by standard sintering [6]. Graphene exists as a two-dimensional planar sheet that displays the basic structure of graphite, CNTs, and fullerenes. It also shows high thermal conductivity (5000 W/(m-K)) and a large heat capacity. In additions, the graphene has excellent mechanical stiffness, Young's modulus of about 1 TPa, and mechanical strength of 130 GPa [7]. Graphite consists of hundreds of thousands of graphene layers while opening carbon nanotubes in the longitudinal direction produces graphene layers [8]. Nanocomposites are materials that have at least one phase of less than 100 nm in size [9]. Carbon nanotubes are one-dimensional, graphene is two-dimensional, and diamond and graphite are three-dimensional [10].

The hybridization of carbon is beneficial for improving the mechanical properties of composite fuel. Carbon nanotubes comprise only sp<sup>2</sup> hybridized bonds. Graphene also shows sp<sup>2</sup> hybridization connected to a hexagonal lattice structure. At room temperature of 293.5 K and 0.1 MPa, the sp<sup>2</sup> bond is more thermodynamically stable than the sp<sup>3</sup> hybridization. Graphite is a fully trigonal sp<sup>2</sup> network, while diamond is fully tetrahedral sp<sup>3</sup> hybridized, each carbon atom show connection with three other carbons. In graphite, each carbon atom contains one non-bonded outer electron, allowing it to conduct electricity.

## 2. Material and Methods

A Computer Code for Thermal-Mechanical Nuclear Fuel Analysis under Steady-state and Transients (FAST) is the last version of the fuel performance code. Fast following the FRAPCON is a fuel rod analysis tool that is sufficient for the conservative analysis, development of the Pacific Northwest National Laboratory (PNNL). The U.S. National Regulatory Commission (USNRC) recommends the FAST and FRAPCON code for the licensing and auditing nuclear reactors in the United States.

FAST code can simulate UO<sub>2</sub> and mixed oxide (MOX) fuel, also support zirconium alloys (zircalloys) as cladding [11]. FAST underwent several modifications to account for UO<sub>2</sub>-graphene properties to analyze its steady-state irradiation performance in a PWR core.

The concept fuel sintered with 5 vol.% carbon nanotubes or graphene powder exhibited a 28% increase in thermal conductivity but limited the grain growth size. In experiments had founded UO<sub>2</sub>-SiC with 10 vol.% SiC showing a 50% conductivity increased if compared with standard UO<sub>2</sub>. In the SPS method, the heated powder is placed under pressure and subjected to direct pulsing at low voltage (5–15 V) and high current (10 kA), allowing nanostructured materials and composites. UO<sub>2</sub> fuel properties result from the microstructure's fuel quality, which affects the temperature profile, fracture, fission gas release, and volumetric changes defined in the sintering process. The sintering of UO<sub>2</sub>-graphene must reduce the pores in a granular material by atomic diffusion, resulting in compact homogenization. The conventional method for UO<sub>2</sub> is the industrial method without external pressure and a lower temperature ramp rate of 2–10 °C/min, as opposed to SPS, which employs a ramp rate of 50–200 °C/min. Table I shows a few fuel properties.

Table I: Summarizes the properties of the carbon compounds blended into the UO<sub>2</sub> matrix.

Properties	UO <sub>2</sub>	Graphene	CNT	SiC
Melting point (°C)	2880 ± 30	4727±1000	4727±1000	2450±250
Density (g/cm <sup>3</sup> )	10.960	2.670	2.600	3.21
Thermal conductivity (W/m·k)	8.68	2500±1200	3000±1000	275±30
Thermal expansion (10 <sup>-6</sup> m/m)	9.76	-4.8	4.25±8	3.5±1
Specific heat (J/kg K)	235	950	425	655
Elastic modulus (GPa)	192	1022	1055	418

The concept fuel sintered with 5 vol.% carbon nanotubes or graphene powder exhibited a 28% increase in thermal conductivity but limited the size of the grain growth. The conventional method for UO<sub>2</sub> is the industrial method without external pressure and a lower temperature ramp rate of 2–10 °C/min, as opposed to SPS which employs a ramp rate of 50–200 °C/min. Uranium carbide has a cubic crystalline structure and a specific heat of 240 J/(kg·K), though carbon composites show a higher specific heat than UO<sub>2</sub>. Thermal conductivity investigations use theoretical models, such as density functional theory, the Boltzmann-transport-equation, and the molecular dynamics simulation approach. Carbon compounds adding to the UO<sub>2</sub> matrix depends on the temperature, porosity, burnup, oxygen/uranium (O/U) ratio, and other factors. The thermal conductivity of the graphene-free defects was 4500 to 5200 W/(m·k). The Neumann-Kopp law can use mixed oxides, such as (Th-U)O<sub>2</sub> to estimate thermal properties.

However, UO<sub>2</sub>-graphene follows a nanocomposite model that uses Hasselman and Johnson's approach in place of the Neumann-Kopp estimation. Code implementation used to simulate the composite fuel dispersion

of graphene (5–10 vol.%) corrects the original subroutines in Fortran of the FRAPCON. Thus, subcodes changed are the thermal conductivity FTHCON, thermal expansion FTHEXP, specific heat capacity FCP, and fuel enthalpy FENTHL. Table II lists the nuclear parameters used in the simulation.

Table II: Nuclear fuel parameters of PWR 17 x 17

Nuclear characteristic parameter	Values
Cladding outer diameter (mm)	0.9144
Pellet outer diameter (mm)	0.7844
Cladding thickness (mm)	0.0571
Pellet length (mm)	11.0007

Under the steady-state operation, simulate a test based on single-rod fuel performance using the licensing code FRAPCON. Material property models used for standard  $\text{UO}_2$  are in the Materials Property (MATPRO) package adopted by the USNRC [12].

### 3. Results and Discussion

The base irradiation of the full-length rod comprised six irradiation cycles, corresponding to an adequate full power of 2064 days. During the cycles, the average linear generation heat rates were 37.5, 28.0, 22.0, 18.0, and 18.0 KW/m. Figure 1 displays a comparison between the average fuel temperature using  $\text{UO}_2$  and the composite fuel  $\text{UO}_2$ -graphene.

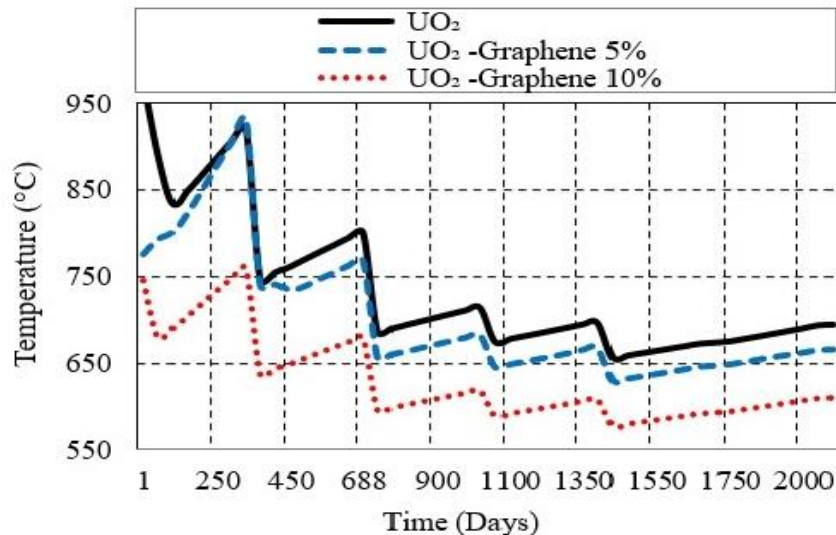


Figure 1: Fuel average temperature for  $\text{UO}_2$ -5 vol% Graphene and  $\text{UO}_2$  10 vol% Graphene

### 4. Conclusions

This study analyzed the  $\text{UO}_2$ -graphene behavior using an adaptation related to the composite fuel and graphene properties. The models applied must provide the essential relationship that describes the thermal and mechanical responses of the fuel rod. Thus, the nuclear PWR of  $17 \times 17$  could support graphene mix fuel. There are many potential benefits of  $\text{UO}_2$ -graphene composite pellets, including enhanced thermal transport capacity combined with the lower neutron absorption cross-section of graphene. Graphene additions produce a notable fuel temperature reduction and improve safety margins.

Also, it can conclude that the SPS method is faster than conventional sintering, with an application time between 5 and 10 mins. It also reaches higher densities at lower temperatures and minimizes grain growth. Composite pellets also exhibit a slight reduction in grain size, with grains of  $12 \pm 2 \mu\text{m}$  compared to  $15 \pm 5 \mu\text{m}$  for pure  $\text{UO}_2$ , with the smaller grain size promoting the release of fission gas. The sintering process improved the fuel density of  $\text{UO}_2$  with 5% vol of graphene of  $10.55 \text{ g/cm}^3$ , and  $\text{UO}_2$  composite fuel with 10% of graphene is  $10.09 \text{ g/cm}^3$ . Theoretically, the composite pellets will suffer reduced cracking because of their higher mechanical strength and reduced thermal expansion. This simulation produced results comparable to those previously reported for mixed fuels based on carbon compounds. Therefore, exist the disadvantage of FGR produced.

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### References

- [1] Davidson. E. E., Franceschini. F., Kim. K. S., “ATF Benchmark Problems.” Oak Ridge National Lab.(ORNL), (2019).
- [2] Gomes, D. S., A. Abe, Silva. A. T., Muniz, R. O. R Muniz, Giovedi, C., Martins, M. R., “Assessment of high conductivity ceramic fuel concept under normal and accident conditions.” In: *Technical Meeting on Modelling of Fuel Behaviour in Design Basis Accidents and Design Extension Conditions*, May 13-16, Shenzhen, China (2019).
- [3] Gomes, D. S., Silva, A. T., “Performance analysis of  $\text{UO}_2$ -SiC fuel under normal conditions.” In: (INAC) October 21-25, Santos, SP pp. 5056-5069, (2019).
- [4] Gomes, D. S., Muniz, R. O. R., Giovedi. C., “Improving performance with accident tolerant fuels.” In: (INAC), October 22-27, Belo Horizonte, MG, (2017).
- [5] Yao. T., Xin, G., Scott. S. M., Gong. B., Lian. J., “Thermally Conductive and Mechanically Robust Graphene Nano platelet Reinforced  $\text{UO}_2$  Composite” *Nuclear Fuels. Scientific reports*, vol. 8(1), 2018, pp. 1-9.(2018).
- [6] Tulenko, J., Subhash, G. “Development of Enhanced Accident tolerant fuel pellets-Innovative High Thermal Conductivity  $\text{UO}_2$  Ceramic Composite Fuel Pellets with Silicon Carbide/ $\text{Cr}_2\text{O}_3$  Using Spark Plasma Sintering.” (No. NEA-NSC-DOC--2013-9), (2013).
- [7] Tulenko. J., G. Subhash, G. “Development of Innovative Accident Tolerant High Thermal Conductivity  $\text{UO}_2$ -Diamond Composite Fuel Pellets, (2016).
- [8] Bhushan, B. Luo. D., Schricker. S. R. Sigmund. W., Zauscher. S. *Handbook of nanomaterials properties*. Springer Science & Business Media, (2014).
- [9] Tanaka. K., Iijima, S. *Carbon nanotubes and graphene*. Newnes (2014).
- [10] Cartas. A., Wang, H., Subhash. G., Baney, Tulenko. J., “Influence of carbon nanotube dispersion in  $\text{UO}_2$ -carbon nanotube ceramic matrix composites utilizing spark plasma sintering.” *Nuclear Technology*, vol. 189(3), pp. 258-267,(2015).
- [11] Geelhood, K. J., Luscher, W. G., Raynaud, P., Porter. I., “FRAPCON-4.0: A Computer Code for the Calculation of Steady-state.” Thermal-Mechanical Behavior of Oxide Fuel Rods for High Burnup, (2015).
- [12] Hagrman, D. L., et al., “MATPRO-Version 11: A handbook of materials properties for use in the analysis of light water reactor fuel rod behavior.” Idaho Falls (USA), (2015).