Title: Heat Transfer Simulations of the UO₂ Particle-Graphite System in TREAT Fuel

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Abstract

In this study, a heat transfer simulation of a UO₂ particle-graphite system in highly-enriched nuclear fuel at the Transient Reactor Test Facility (TREAT) was performed using the finite element method. Different factors that can impact fuel performance were modeled and implemented in the simulated micro-scale UO₂ particle-graphite system. The fission fragments caused an irradiation-induced degradation of the thermal conductivity of the graphite, which added major heat resistance to the irradiated system. The effect of graphite quality and irradiation on the UO₂ particles has also been evaluated, but neither has an impact as pronounced as the fission fragment damage to the graphite. By combining these factors, the dynamic temperature profiles were obtained, and the limitations on particle size in the irradiated and unirradiated UO₂ particle-graphite systems have been determined.

Keywords: TREAT, finite element analysis, uranium oxide, graphite, heat transfer, COMSOL

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1. Introduction

After the Fukushima Daiichi nuclear accident, development of advanced accident-tolerant fuels for Light Water Reactors (LWRs) has become an urgent goal of the nuclear industry in order to improve the safety margin of nuclear plants based on experience gained from the accident (Zinkle et al., 2013; Zinkle and Was, 2013). In comparison with the traditional fuel (UO_2 with Zircaloy cladding) used in LWRs, accident-tolerant fuels are expected to tolerate extreme environments in off-normal conditions for longer periods of time (Wachs, 2012), i.e. exhibit a longer “grace period” (IAEA, 1991). To date, many advanced techniques have been proposed to improve the fuel’s resistance to damage, melting, and chemical reaction during accident scenarios (Farmer et al., 2014; Lindemer et al., 2014; Mo et al., 2014; Nelson et al., 2014; Ott et al., 2014; Terrani et al., 2014; Yacout et al., 2013; Yueh and Terrani, 2014; Zinkle and Was, 2013). To examine the performance of advanced fuels, a test reactor that can simulate the neutron and thermal conditions during transients is necessary. The Transient Reactor Test Facility (TREAT) is a large air-cooled reactor system located at Idaho National Laboratory (INL). During service from 1959 to 1994 (INL, 2009), the reactor generated several thousand reactor transients, and hundreds of experiments have been performed for a wide variety of reactor development programs (Crawford et al., 1998). TREAT is now being prepared to restart its operation to assist advanced fuel development by providing a method to perform transient testing (Wachs, 2012).

TREAT has not operated in the 20 years since 1994. To manufacture new fuel for the reactor, fuel specifications are needed as a guideline to provide limits on both physical and thermal properties. TREAT was designed to simulate transients in off-normal conditions, including transients seen in severe accidents. The core consists of a 19×19 array of assemblies, each containing a 1.22 m (4 ft) high active section composed of graphite- UO_2 fuels. A dispersion of

fine UO_2 particles (typically tens of micrometers in diameter) is embedded in the graphite. While the test specimens during a TREAT transient experience conditions which can lead to fuel damage, including melting and cladding failure, the reactor's fuel itself was designed to maintain its integrity even when generating very large power bursts.

In parallel with TREAT restart preparations, a program is underway to develop a similar replacement core in which the UO_2 contains only low-enriched uranium. As part of the design of the low-enrichment core, analyses are being conducted to evaluate the rate of heat transfer from the UO_2 particles to the surrounding graphite matrix during the most severe transients anticipated to be performed with the core. The current study was based on the original fuel design of TREAT, in which highly-enriched uranium (HEU) was used. For the recent low-enriched uranium (LEU) core design, both carbon/uranium atomic ratio and enrichment will be significantly lower than those in the HEU core design. The LEU core will need to generate power transients about 20-30% higher in power and energy than the HEU core did. However, the modeling procedures and related discussion in this study are independent of the changes in fuel properties and power generated during transients and are therefore still valid for the LEU fuel.

In this study, the heat transfer in the UO_2 -graphite system used in the TREAT fuel was modeled using the finite element method to evaluate the fuel performance during transients. The heat produced by individual UO_2 particles was scaled from the reactor power-time histories measured in previous high-peak-power transient tests in TREAT. The model takes into account multiple factors that may influence fuel performance: (1) degradation of the thermal conductivity of graphite after fission recoil fragment irradiation; (2) degradation of the thermal conductivity of UO_2 after irradiation; and (3) dependence of the graphite matrix thermal conductivity upon the degree of graphitization of the carbon. This study aims to develop a better understanding of the

contributions of the aforementioned factors to heat transfer in the micro-scale UO_2 particle-graphite system and provide some baseline information for the current low-enriched fuel design for TREAT.

2. Computational set up

2.1 Thermal properties of UO_2

The required thermal properties for the current simulation include the thermal conductivity k , heat capacity c_p , and density ρ . For heat transfer simulation in the UO_2 particle-graphite system, the most important material property is the thermal conductivity. Many different models for the temperature-dependent thermal conductivity of UO_2 have been interpreted from experimental data (Carbajo et al., 2001; Duriez et al., 2000; Fink, 2000; Fink et al., 1981; Harding and Martin, 1989; Lucuta et al., 1996; Lucuta et al., 1994, 1995; Martin, 1982; Philipponneau, 1992; Ronchi et al., 1999). The difference in thermal conductivity among these models is insignificant over a temperature range from 300 to 3000 K. Here, we selected the equation given by Ronchi et al. (Ronchi et al., 1999):

$$k = \frac{100}{6.548 + 23.533t} + \frac{6400}{t^{5/2}} \exp\left(\frac{-16.35}{t}\right), \quad (1)$$

where $t = T(\text{K})/1000$ and k is the thermal conductivity in $\text{W m}^{-1}\text{K}^{-1}$.

The temperature dependence of the heat capacity of UO_2 was modeled by Fink et al. (Fink, 2000; Fink et al., 1981) based on the available experimental data. The following equation was selected for the current simulation:

$$c_p(T) = 52.1743 + 87.951t - 84.2411t^2 + 31.542t^3 - 2.6334t^4 - 0.7139t^{-2}, \quad (2)$$

where $t = T(\text{K})/1000$ and c_p is in $\text{J mol}^{-1}\text{K}^{-1}$. Fig. 1 shows the plot of c_p as a function of temperature from 300 to 3000 K (The units of c_p were converted to $\text{J kg}^{-1}\text{K}^{-1}$ with the molar weight of 267.2 g/mol in order to implement the equation in the finite element model).

(insert Figure 1)

Because the temperature dependence of the UO_2 density is very small below the melting point (Fink, 2000), the constant value of 10.963 kg/m^3 was selected for this simulation. Note that the UO_2 particle is assumed to be of full density in the present study; however, some uncertainty may exist in the results for very large particles ($> 100 \text{ um}$) due to possible retained porosity.

The most concerning irradiation effect on the UO_2 in this simulation is the irradiation-induced degradation of the thermal conductivity. Lucuta et al. developed an analytical expression for the thermal conductivity of irradiated UO_2 based on the analysis of the SIMFUEL (simulated extended burnup UO_2 fuel) data:

$$c_p = k_{1d}(\beta, T)k_{1p}(\beta, T)k_{2p}(p, \sigma)k_{3x}k_{4r}(T)\lambda_0(T), \quad (3)$$

where β is the burnup in atom %; T is the temperature in K; p is the volume fraction of pores and bubbles (assuming 5 volume % in this study); and σ is the pore shape factor (assuming 1.5 for spherical bubbles). The expressions for all components of Eq. 3 are given in Table 1. Detailed analysis for this model can be found in the work by Lucuta (Lucuta et al., 1996).

(insert Table 1)

Note that this model is established based on the assumption that the lattice vibration, rather than the electronic process, is the major contributor to the thermal conductivity. This assumption is

valid below 1900 K, but the contribution of the electronic process will take over above this temperature (Lucuta et al., 1996). In this study, we assumed that, at temperatures higher than 1900 K, the temperature dependence of the thermal conductivity of UO_2 after irradiation is similar to that without irradiation. Figure 2 shows the comparison of the thermal conductivities of unirradiated and irradiated UO_2 as calculated by Eqs. 1 and 3, respectively.

In addition to the thermal conductivity, neither the thermal capacity nor the density is very sensitive to irradiation up to 1% burnup (Fink, 2000; Lucuta et al., 1995), which would likely only be achieved after many years of TREAT operation. Therefore, we used the same sets of values for these two parameters in the simulations for both the unirradiated and irradiated systems.

(insert Figure 2)

2.2 Thermal properties of graphite

The thermal conductivity of graphite is dependent on its quality and irradiation history. The quality of graphite used in the original TREAT fuel is not comparable to the nuclear grade graphite that was proposed for use in the graphite moderated high-temperature reactor, i.e. the Very-High Temperature Gas-cooled Reactor (VHTR). The graphite in the original TREAT fuel is actually mixed with molded carbon and graphite, and contains some interior pores and cracks after manufacturing. A large difference in thermal conductivity was found between these two types of graphite. Figure 3 shows the thermal conductivity for both graphite used in the original TREAT fuel (Handwerk and Lied, 1960) and nuclear grade graphite (Burchell, 2012; Gallego and Burchell, 2011). The thermal conductivity of nuclear grade graphite is about 7 times that of the TREAT graphite at room temperature, and about 3 times that of the TREAT graphite at

1100°C.

In the TREAT fuel, irradiation of the graphite is mainly due to two sources: fission fragments and neutrons. The irradiation damage from the fission fragments accumulates within a short range near UO_2 particles, whereas the damage from neutrons is more homogeneous within the entire UO_2 particle-graphite system. Many studies of the neutron irradiation effects on graphite have been conducted since the 1950s (Bonal et al., 2009; Burchell, 2012; Gallego and Burchell, 2011; IAEA, 2000; Kelly, 1967, 1981; Simmons, 1965; Snead, 2008; Snead and Burchell, 1995; Snead et al., 2005; Sumita et al., 2009). The thermal conductivity of graphite after fast neutron irradiation (Snead and Burchell, 1995) is very close to that of the original TREAT graphite (Handwerk and Lied, 1960). It is very difficult to quantify the neutron irradiation effect on the original TREAT graphite, since the graphite's intrinsic defects (such as interior pores and cracks) outweigh the effects of neutron irradiation. This simulation is focused on the effect of fission fragment recoil damage near UO_2 particles. Compared to a great number of studies on the fast neutron irradiation damage of graphite, very few studies focus on fission fragment damage. Hunter (Hunter, 1959) reported experiments that measured the thermal conductivity degradation of impregnated/molded graphite as a result of irradiation by fission recoil fragments. He observed that the thermal conductivity at high temperatures decreased from 41.8 W/(m·K) to a saturated value of ~1.046 W/(m·K). This value did not further decrease with more irradiation, and thus was considered the saturated thermal conductivity after fission fragment irradiation. In the current study, we assume the temperature dependence of the thermal conductivity after fission fragment irradiation is insignificant. The assumption is based on the observations of neutron-irradiated graphite (Burchell, 2012; Gallego and Burchell, 2011) and the unirradiated TREAT graphite (Fig. 3), both of which show a very small temperature dependence on thermal

conductivity. A constant thermal conductivity of 1.046 W/(m*K) (Hunter, 1959) is used for the fission fragment irradiated graphite (Fig. 3).

(insert Figure 3)

Despite the great difference in the thermal conductivity, the heat capacities of nuclear grade graphite and the original TREAT graphite are comparable in the range of room temperature to 1000°C. The heat capacity for the original TREAT graphite was reported by Handwerk and Lied (Handwerk and Lied, 1960), and is plotted in Fig. 4. The analytical expression of the heat capacity (unit: J/kg*K) for nuclear grade graphite is given in the ASTM standard C781-08 (ASTM, 2008):

$$c_p = \frac{1}{11.07T^{-1.644} + 0.0003688T^{0.02191}} \quad (4)$$

The density of the original TREAT fuel (Handwerk and Lied, 1960) is $\sim 1.72 \text{ g/cm}^3$ compared to the value of $\sim 1.8 \text{ g/cm}^3$ for nuclear grade graphite (Burchell, 2012). The irradiation effect on both the heat capacity and the density of graphite is assumed to be insignificant in this simulation (this assumption is based on the references: (Gallego and Burchell, 2011; Simmons, 1965; Snead and Burchell, 1995)).

(insert Figure 4)

2.3 UO₂ particle-graphite models

The graphite-uranium block used in a HEU TREAT fuel element was designed to be 10.16 cm × 10.16 cm × 122 cm (Handwerk and Lied, 1960). To fabricate the graphite-uranium block, 0.248 weight percent U₃O₈ powder (containing 93.1% enriched uranium) was loaded into the fuel mix.

During fabrication, the U_3O_8 powder was presumably reduced to UO_2 . The volume ratio of UO_2 to graphite is estimated to be 1:2571.

The UO_2 particle-graphite model developed in this work utilizes the commercial finite-element code COMSOL Multiphysics[®] (COMSOL, 2013). With the capability to couple multiple physical models within a single model domain, COMSOL has been used extensively in various engineering areas and has recently been introduced to research in nuclear engineering (Aufiero et al., 2013; Bell and Lewis, 2012; Cammi et al., 2012; Di Marcello et al., 2012; Kim et al., 2013; Mourgues and Sanchez, 2012; Sreedhar et al., 2013). The simulation in this study employed the “heat transfer” module to solve for the time-dependent heat transfer in the UO_2 particle-graphite system. The fuel in TREAT was simplified to be a UO_2 particle-graphite system based on the assumption that all UO_2 particles are uniformly dispersed within the graphite matrix. Figure 5 shows the basic model for the unirradiated UO_2 particle-graphite system. Both the graphite matrix and the UO_2 particle embedded in the matrix are modeled to be spherical, with a radial ratio of 1/13.7. This model is referred to as the “cell calculation:” each fuel particle is associated with a “cell” whose dimensions are prescribed by the overall fuel-to-moderator ratio and the average size of the fuel particles (Hetrick, 1953). To accelerate the computation while maintaining sufficient accuracy, the system is modeled as having 2D-axisymmetric geometry, where the axisymmetric boundary is placed across the center of the particle. The outer boundary of the graphite matrix is a symmetric boundary. This model was developed to simulate the unirradiated UO_2 particle-graphite system in the highly-enriched fuel for TREAT.

(insert Figure 5)

After long-term reactor operation, the irradiation effect on both the UO_2 particles and the graphite must be taken into account in the model. The focus of this simulation was on the fission fragment irradiation of graphite near UO_2 fuel particles. To determine the range of the fission fragment damage in graphite, SRIM (Stopping and Range of Ions in Matter) computer code was used to calculate the interaction of the fission fragments with the graphite. SRIM is often used to calculate the interaction of ions with matter based on the Monte Carlo simulation method (Biersack and Haggmark, 1980; Ziegler, 1977-1985). Here, 100 MeV Xe ions were modeled to bombard the graphite. The bombardment produces a damaged region with a depth of $\sim 15\ \mu\text{m}$ in the graphite. On that basis, the range of damage that results due to fission fragment irradiation in graphite was estimated to be $\sim 15\ \mu\text{m}$. For a conservative estimation, we homogenized the irradiation damage within this range and assumed that the degraded thermal conductivity of the region reached a saturated value, an assumption that was based on a study by Hunter (Hunter, 1959). Hunter observed that the decrease in the thermal conductivity of graphite due to fission fragment irradiation saturates when the dose reaches a certain value. This saturation phenomenon of the thermal conductivity of graphite has also been observed in neutron-irradiated graphite for even lower doses (Burchell, 2012; Gallego and Burchell, 2011; Snead and Burchell, 1995). The $15\ \mu\text{m}$ irradiation-damaged graphite layer (blue region in Fig. 6) was incorporated in the system between the UO_2 particle (red region in Fig. 6) and the massive graphite matrix (grey region in Fig. 6).

(insert Figure 6)

2.4. Model inputs

Three core temperature-limited natural burst transients from the original TREAT experiments are shown in Fig. 7. The left y-axis in Fig. 7 is the total power produced in TREAT as a function of time. The transients labeled “Test EOS-2,” “Test E3,” and “Test E4” represent three transients of

different severities. Among them, Test EOS-2 (Tylka et al., 1977) used the highest power transient able to be generated in TREAT using the original highly-enriched fuel. Tests E3 and E4 represent the lower-peak-power, broader burst transients (Deitrich, 1975; Doerner et al., 1971). The total TREAT power was scaled to the power of a single highly-enriched UO_2 fuel particle with respect to the volume of that particle. The right y-axis in Fig. 7 shows the scaled power of the UO_2 particle with a 44 μm diameter (the typical diameter of the UO_2 fuel particles in the original TREAT fuel). For both the unirradiated and irradiated models, heat was assumed to homogeneously generate within the UO_2 fuel particle embedded in the graphite matrix.

(insert Figure 7)

3. Results

3.1. Unirradiated TREAT fuel

The UO_2 particle-graphite system in the unirradiated original TREAT fuel (Fig. 5) was used as the baseline for this study. Figure 8 shows the change in temperature at different critical boundaries/locations in the system (shown in Fig. 5) during the transients. For all three transients, the temperature in the center of the particles reaches a maximum value slightly after the moment of peak power. For the EOS-2 transient, the highest temperature in the center of the UO_2 particle was found to occur ~0.17 seconds after the initiation of the transient. The temperature profile of the system at this moment is shown in Fig. 9. The temperature gradient from the center of the particle to the graphite-particle boundary is ~260°C. For the E3 and E4 transients, the change in temperature of the systems was not as profound as in the EOS-2 transient. The maximum temperatures in the UO_2 particle during the EOS-2, E3, and E4 transients are ~560, ~235, and ~150°C, respectively (Fig. 8). The equilibrium temperatures of the system during the EOS-2, E3, and E4 transients are ~340, ~200, and ~140°C, respectively. For all three transients, the heat

release of the UO_2 particles takes ~ 0.3 seconds, after which the temperature of the particle finally reaches the equilibrium temperature of the system.

(insert Figure 8)

(insert Figure 9)

3.2. Irradiated TREAT fuel

Figure 10 shows the temperature development at different critical boundaries/locations in the irradiated UO_2 particle-graphite system (Fig. 6). The maximum temperatures in the UO_2 particle during the EOS-2, E3, and E4 transients were ~ 1700 , ~ 600 , and $\sim 295^\circ\text{C}$, respectively (Fig. 10). In the EOS-2 simulation of irradiated fuel, as in the unirradiated simulation, the highest temperature in the center of the UO_2 particle was found to occur ~ 0.17 seconds after the initiation of the transient. At that moment, the temperature differences from the particle center to the particle-graphite boundary and from the particle-graphite boundary to the irradiation-damaged graphite boundary were 520°C and 880°C , respectively (Fig. 11). The equilibrium temperatures of the system during the EOS-2, E3, and E4 transients are almost identical to the equilibrium temperatures of the unirradiated TREAT fuel. Note that the equilibrium temperature is determined by the power input and the heat capacity of the material, both of which are considered to be unaffected by the irradiation. The time required to reach an equilibrium temperature is similar for the irradiated and unirradiated systems.

(insert Figure 10)

(insert Figure 11)

3.3. Limitations on UO₂ particle size

The power output of the particle is determined by its size/volume when assuming a homogeneous power density within the TREAT fuel. Starting from the baseline simulation with the UO₂ particle with a 44 μm diameter, UO₂ particles of different sizes were modeled, and their temperature developments during different transients were simulated. Figure 12 shows the maximum temperatures at different critical boundaries/locations of the unirradiated UO₂ particle-graphite system during the transients. Note that the larger the particle, the larger the temperature gradient from the particle center to the graphite. This indicates that the heat produced in the large UO₂ particles was not as efficiently released during the transients. Here, the critical particle size was defined as the maximum size of the UO₂ particle whose highest temperature during the transient was below the UO₂ melting point of $\sim 2,865^{\circ}\text{C}$. In other words, the particle center would melt if the particle size exceeded the critical size. In the unirradiated system, the critical particle sizes for the EOS-2, E3, and E4 transients were found to be 95 μm , 160 μm , and 255 μm , respectively (Fig. 12).

(insert Figure 12)

After irradiation, the decrease in the thermal conductivity of both the UO₂ particle and the graphite near the UO₂ particles significantly impedes the heat transfer within the system. Figure 13 shows the maximum temperatures at different critical boundaries/locations of the irradiated UO₂ particle-graphite system during the transients. For UO₂ particles of the same size, those in the irradiated system attain a much higher temperature than those in the unirradiated system. Therefore, the critical particle size in the unirradiated system is much smaller than that in the irradiated system, and is valued at 60 μm , 115 μm , and 200 μm for the EOS-2, E3, and E4 transients, respectively.

(insert Figure 13)

3.2. Influence of graphite quality

The thermal conductivity of nuclear grade graphite is much better than that used in the original TREAT core (Fig. 3). Thus, the heat produced in UO_2 particles is expected to release faster when the UO_2 particles are embedded in nuclear grade graphite. During the simulation of the EOS-2 transient, the maximum temperature attained in the center of the particle is $\sim 460^\circ\text{C}$ (Fig. 14), whereas this temperature is $\sim 560^\circ\text{C}$ for the case of the UO_2 particle embedded in the original TREAT graphite (Fig. 8(a)). The influence of the superior thermal conductivity of the nuclear grade graphite can also be seen in the temperature gradients from the particle-graphite boundary to the graphite matrix. This temperature development is represented by the red and green curves in Figs. 8 and 14. Almost no temperature gradient exists in the case of the nuclear grade graphite (Fig. 14), whereas there is about an 80°C gradient in the case of the original TREAT graphite (Fig. 8(a)). The improved thermal conductivity of nuclear grade graphite allowed for larger critical particle sizes because more heat could be released through the graphite during the transients. The critical particle sizes for the EOS-2, E3, and E4 transients are $110\mu\text{m}$, $180\mu\text{m}$, and $290\mu\text{m}$, respectively.

(insert Figure 14)

Although using nuclear grade graphite in the system accelerates the heat release from the UO_2 particles, the effect is less significant in the irradiated system. Figure 15 shows the temperature development of the irradiated UO_2 particle-graphite system during the EOS-2 transient. The peak temperature in the center of the UO_2 particle is $\sim 1640^\circ\text{C}$, a value that can be compared to the peak temperature of $\sim 1650^\circ\text{C}$ for the UO_2 particle in the original TREAT graphite (Fig. 10 (a)). Therefore, the increase in the critical particle size through the use of nuclear grade graphite in the irradiated system is not as profound as that in the unirradiated system. The critical particle sizes

for the EOS-2, E3, and E4 transients are 65 μ m, 120 μ m, and 205 μ m, respectively, all of which are only about 5 μ m larger than the critical particle sizes in the original TREAT graphite.

(insert Figure 15)

4. Discussion

As previously mentioned, the outer boundary of the graphite matrix is modeled to be symmetric. The system is therefore adiabatically bounded. Since the present study is based on natural burst transients occurring over a sub-second time period, the heat transferred from fuel element to cladding, and further to the outside environment, is insignificant within the time frame. Another assumption in the model is the homogeneous size and space distribution of fuel particles within the graphite. Under this assumption, the packing fraction of the UO₂ is dilute enough that the temperature pulses resulting from different particles do not interact. In reality, although some aggregations of small UO₂ particles were found in the fuel elements (Handwerk and Lied, 1960; Hunter, 1959), the majority of fuel particles were well distributed within the graphite.

Although the high thermal conductivity of nuclear grade graphite can improve heat transfer, the contribution of good-quality graphite to the irradiated system is not greatly beneficial due to the large thermal resistance of the irradiation-damaged graphite layer. Here, it should be noted that the quality of the graphite might not be as high as that of nuclear grade graphite due to the fabrication procedures involved in manufacturing the original TREAT fuel. The thermal conductivity measured from the original graphite may not be overly applicable to the graphite in the new TREAT fuel, which presumably contains fewer cracks and less porosity. The thermal conductivity of the graphite in the new TREAT fuel is expected to be between the values for nuclear grade graphite (black curve in Fig. 3) and the original TREAT graphite (red curve in Fig.

3), even though the graphitization ratio may increase in the new fuel design. Combining the factors of irradiation damage and graphite quality, the upper size limit of the UO_2 particle can be determined based on the highest-power transient (the EOS-2 transient) performed with the highly-enriched original TREAT fuel. This critical particle diameter is $60\mu\text{m}$, assuming the system with the original TREAT graphite (lower thermal conductivity) was irradiated to 1% burnup. Most of the particles within the fuel should not exceed this value, otherwise UO_2 particle melting would likely occur. Subsequently, the melted fuel particles would solidify, and the newly-formed uranium oxide could have different stoichiometry and thermal properties. Meanwhile, a gap between the uranium oxide particle and the graphite could be created during the melting-resolidification process. All these consequences following particle melting should be avoided during reactor operation.

From the SRIM calculations, the fission damage boundary was estimated to be about $15\mu\text{m}$ from the particle. Thus, if the UO_2 particle-to-particle distance were smaller than $30\mu\text{m}$, most of the graphite would experience irradiation damage from fission fragments in addition to damage from neutrons. When the total volume of UO_2 particles is determined, the distance between the particles is smaller for smaller mean particle sizes. Figure 16 compares the fission-fragment irradiation damaged region (in blue) produced by differently-sized particles (in red). For fine particles, the heat produced by each particle is very small, so the risk of melting would not be a concern. However, the fission fragments will damage a large fraction of the graphite matrix, thereby the general thermal conductivity of the graphite will decrease during reactor operation. Previous experiments on $1\mu\text{m}$ UO_2 -impregnated graphite show a dramatic decrease in the thermal conductivity following reactor radiation exposure (Hetrick et al., 1953). The ratio of the thermal conductivity of the graphite before and after irradiation was reported as being in the range of 1/30 to 1/15. Therefore, if fine UO_2 particles are to be employed, the thermal

conductivity degradation by fission fragments must be included in the thermal calculation of the fuel elements and the reactor core.

(insert Figure 16)

In this study, the UO_2 particle was modeled as being embedded in graphite, i.e., in intimate contact with the graphite. This was an optimized case of this phase boundary, as there was no gap between the particle and the graphite. With good quality control in fuel fabrications, it is unlikely to see a large amount of UO_2 particles pulled out of the graphite matrix. However, one must note that if a gap (either filled with air or vacuum, dependent on the fabrication approach) between the particle and the graphite exists, this gap may significantly impede the heat release from the UO_2 particle, and can result in much higher temperatures compared to the system without the gap.

Another concern for the UO_2 particle-graphite system is the possible interaction between graphite and UO_2 . This interaction may produce CO/CO_2 and uranium carbide, depending on the operating temperature and pressure, and may change the physical properties of the system. However, the “threshold” temperature for this interaction is $1700\text{K} \pm 150\text{K}$. Below this temperature, the interaction rate will be negligible (Campbell, 1976). Thus, this interaction may only occur in the large UO_2 particles with high burnup during a transient (note that the temperature of the particle decreased rapidly in the transients), and will not be a major issue for fuel elements in general.

5. Conclusions

In this study, the heat transfer in a UO_2 particle-graphite system in TREAT fuel was simulated with the finite element method. This analysis, developed for highly-enriched UO_2 particles, is equally appropriate for use with low-enriched particles. It can determine particle size limits for use in design specifications and fabrications requirements of TREAT fuel. After studying different factors that can influence heat transfer within the system, the following conclusions were reached:

- a. After fission fragment irradiation, the thermal conductivity of graphite is significantly degraded. The heat produced by the UO_2 particle cannot easily transfer to the graphite, leading to a significant increase in the temperature of the UO_2 particle.
- b. Graphite with better qualities (especially better thermal conductivity) can improve the heat transfer within the fuel. However, the major heat resistance in the system after long-term operation is due to the thin fission fragment irradiated graphite layer. Thus, the contribution of better-quality graphite is negligible for the irradiated system.
- c. The limitations on the fuel particle size in order to avoid particle melting in the particle center were determined. The 44 μm UO_2 particle size in the original HEU TREAT fuel is confirmed by the present analysis to be appropriate for use, and has been demonstrated by the satisfactory performance of that fuel during TREAT's many years of operation.

6. Acknowledgements

This work was sponsored by the U.S. Department of Energy, Office of Global Threat Reduction (NA-21), National Nuclear Security Administration, under Contract No. DE-AC-02-06CH11357 between UChicago Argonne, LLC and the Department of Energy.

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Figure captions

Fig. 1: Heat capacity as a function of temperature calculated by Eq. 2 (Fink, 2000).

Fig. 2: Thermal conductivity of UO_2 as a function of temperature as calculated by Eqs. 1 and 3 (Lucuta et al., 1996; Ronchi et al., 1999). The irradiated UO_2 was assumed to achieve 1% burnup.

Fig. 3: Thermal conductivity of graphite in different conditions (Burchell, 2012; Gallego and Burchell, 2011; Handwerk and Lied, 1960; Hunter, 1959)

Fig. 4. Heat capacity of graphite in different conditions (ASTM, 2008; Handwerk and Lied, 1960)

Fig. 5: Schematic of the unirradiated UO_2 particle-graphite model. The UO_2 particle (red) is embedded in the graphite matrix (grey).

Fig. 6: Schematic of the irradiated UO_2 particle-graphite model. The UO_2 particle (red) was embedded in the graphite matrix (grey). A fission fragment irradiated zone (blue) was built between the two phases.

Fig. 7: TREAT core temperature-limited natural burst transients (left y-axis) and the scaled power inputs of a UO_2 particle with a 44 μm diameter (right y-axis) as functions of time.

Fig. 8: The simulated temperature development of the unirradiated UO_2 particle-graphite system during the Test (a) EOS-2, (b) E3, and (c) E4 transients

Fig. 9: The temperature profile of the unirradiated UO_2 particle-graphite system at ~0.17 seconds after initiation of the EOS-2 transient

Fig. 10: The simulated temperature development of the irradiated UO_2 particle-graphite system during the Test (a) EOS-2, (b) E3, and (c) E4 transients

Fig. 11: The temperature profile of the irradiated UO_2 particle-graphite system at ~0.17 seconds after initiation of the EOS-2 transient

Fig. 12: The maximum temperatures of differently-sized particles at various critical boundaries/locations in the unirradiated UO_2 particle-graphite system during the Test (a) EOS-2, (b) E3, and (c) E4 transients

Fig. 13: The maximum temperatures of differently-sized particles at various critical boundaries/locations in the irradiated UO_2 particle-graphite system during the Test (a) EOS-2, (b) E3, and (c) E4 transients

Fig. 14: The simulated temperature development of the unirradiated UO_2 particle-graphite system during the EOS-2 transient. The UO_2 particle with a 44 μm diameter was embedded in the nuclear grade graphite.

Fig. 15: The simulated temperature development of the irradiated UO_2 particle-graphite system during the EOS-2 transient. The UO_2 particle with a 44 μm diameter was embedded in nuclear grade graphite.

Fig. 16: The irradiation-damaged zone near differently-sized UO_2 particles

(Figure 1)

(Figure 2)

(Figure 3)

(Figure 4)

(Figure 5)

(Figure 6)

(Figure 7)

(a)

(b)

(c)

(Figure 8)

(Figure 9)

(a)

(b)

(c)

(Figure 10)

(Figure 11)

(a)

(b)

(c)

(Figure 12)

(a)

(b)

(c)

(Figure 13)

(Figure 14)

(Figure 15)

(Figure 16)

Table 1. The components of the thermal conductivity model of irradiated UO₂ (Eq. 3), (Lucuta et al., 1996).

Component	Expression	Effects for the quantifications
k_{1d}	$k_{1d}(\beta) = \left(\frac{1.09}{\beta^{3.265}} + \frac{0.0643}{\sqrt{\beta}} \sqrt{T} \right)$ $\arctan \left(\frac{1}{1.09/\beta^{3.265} + (0.0643/\sqrt{\beta})\sqrt{T}} \right)$	Dissolved fission product
k_{1p}	$k_{1p} = 1 + \frac{0.019\beta}{3 - 0.019\beta} \frac{1}{1 + \exp(0.0643/\sqrt{\beta})\sqrt{T}}$	Precipitated solid fission product
k_{2p}	$k_{2p} = \frac{1 - p}{1 + (\sigma - 1)p}$	Pore and fission-gas bubbles
k_{3x}	$k_{3x}=1$	No deviation from stoichiometry under normal operation conditions
k_{4r}	$k_{4r} = 1 - \frac{0.2}{1 + \exp((T - 900)/80)}$	Radiation damage
λ_0	Eq. (1)	Thermal conductivity of unirradiated UO ₂

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