INL/JOU-19-54936-Revision-0

Impact of Grain Size on Performance Degradation of TREAT LEU

May 2020

Sebastian Schunert, Adam X Zabriskie, Daniel Schwen, Vincent M Laboure, Mark D DeHart

Changing the World's Energy Future

Idaho National Laboratory

INL/JOU-19-54936-Revision-0

Impact of Grain Size on Performance Degradation of TREAT LEU

Sebastian Schunert, Adam X Zabriskie, Daniel Schwen, Vincent M Laboure, Mark D DeHart

May 2020

Idaho National Laboratory
Idaho Falls, Idaho 83415

http://www.inl.gov

Prepared for the
U.S. Department of Energy
Under DOE Idaho Operations Office
Contract DE-AC07-05ID14517

Annals of Nuclear Energy 139 (2020) 107294

Contents lists available at ScienceDirect

Annals of Nuclear Energy

journal homepage: www.elsevier.com/locate/anucene

Impact of grain size on performance degradation of TREAT LEU

Sebastian Schunert *, Adam Zabriskie, Daniel Schwen, Vincent Laboure, Mark DeHart

Nuclear Science and Technology Directorate, Idaho National Laboratory, P.O Box 1625, Idaho Falls, ID 83415, United States

ARTICLE INFO

Article history:
Received 13 August 2019
Received in revised form 27 November 2019
Accepted 27 December 2019
Available online 10 January 2020

Keywords:
TREAT LEU conversion
Multiphysics simulation
Graphite
Radiation damage

ABSTRACT

We argue that radiation damage induced degradation of thermal conductivity does not set a lower limit on fuel grain sizes for the low enriched uranium fuel design of the Transient Reactor Test Facility (TREAT). Earlier work reports that smaller grains cause a larger degradation of thermal conductivity than larger grains constraining the smallest feasible size of fuel grains. This work assesses TREAT's transient performance in the presence of radiation damage. The difference between the two studies is in treating damaged and fresh graphite as serial (this work) or parallel (previous) heat resistors. We use a multiphysics model of TREAT fuel grains to compute the reduction in transient capability measured by the total deposited energy as a function of irradiation dose. We find that radiation damage has a negligible effect on energy deposition.

1. Introduction

The transient reactor test facility (TREAT) at Idaho National Laboratory allows testing of current and future fuel concepts under severe accident conditions (Freund et al., 1960). TREAT is a graphite moderated reactor with high enriched uranium (HEU) fuel particles with an estimated radius between 10 and 20 μm distributed in the moderator. A common TREAT experiment, referred to as a self-limiting pulse, places fuel in the experiment cavity and inserts reactivity into the reactor, increasing the reactor power which in turn increases the reactor temperature. The increase in reactor temperature reduces its reactivity, ultimately shutting the reactor down. During the power excursion, a fraction of the reactor energy is deposited into the experiment, producing desired accident conditions while thermal feedback shuts the reactor down. This work exclusively investigates conditions during self-limiting TREAT pulses.

To reduce the proliferation risk, an effort is currently underway to convert TREAT to low enriched uranium (LEU). The LEU design maintains a similar fuel concept to the HEU design; the design differences include a decrease of the enrichment of U-235 from 94.5 mol-% to 20 mol-% and a decrease of the graphite to fuel volume ratio from 2572:1 to 626:1. The goal of these changes is to maintain roughly the same amount of U-235 in the LEU core (Connaway et al., 2016). It is also expected that TREAT's LEU fuel design will have significantly smaller fuel grains than the HEU design.

There are two main reactivity feedback mechanisms relevant to this work: Doppler broadening and spectral shift. An increase in the U-238 content in LEU fuel grains introduces additional Doppler temperature feedback that is sensitive to the fuel temperature, whereas spectral shift, the only relevant feedback mechanism for the HEU core, is sensitive to graphite temperature only (Zabriskie et al., 2019). Given a sufficient difference between the average fuel grain and graphite temperatures during a transient, the additional feedback could severely limit the LEU TREAT's operational envelope. The particular concerns are:

1. poor heat removal from the fuel to the graphite by large fuel grains.
2. creation of an insulating layer by fission product induced radiation damage in the graphite immediately outside the fuel grains.

Mo et al. (2015) investigates the evolution of fuel temperature for measured TREAT power traces, and Mo et al. (2017) investigates the bulk heat conductivity of TREAT fuel as a function of irradiation. These studies find that for smaller fuel grains, heat removal from the fuel grains is more efficient, but thermal conductivity degradation is more severe. The studies conclude that radiation damage effectively limits the minimum grain size for LEU fuel to 10 μm.[1]

* Corresponding author.
E-mail address: sebastian.schunert@inl.gov (S. Schunert).

[1] Mo et al. (2017) is not clear on whether size refers to diameter or radius. Earlier work of the same author (Mo et al., 2015) suggests that size refers to diameter, hence the minimum radius is found to be 5 μm in Mo et al. (2017).

https://doi.org/10.1016/j.anucene.2019.107294
0306-4549/

This work re-examines the importance of radiation damage for LEU TREAT and its impact on grain size selection using a multiphysics model of a TREAT fuel grain. In contrast, Mo et al. (2017) makes its conclusion based on a bulk thermal conductivity of irradiated graphite, i.e. the thermal conductivity of a block of fueled graphite with two different temperatures maintained at the left and right edge. In our opinion, this does not represent the relevant physical process in TREAT, because it treats fission-fragment irradiated and essentially undamaged graphite far away from fuel grains as alternative paths for heat flow. In TREAT, heat is generated in fuel grains and then needs to pass both irradiated and un-irradiated graphite regions. In essence, Mo et al. (2017) uses a parallel heat resistance model as figure of merit when a serial heat resistance model better represents the process.

In addition, Mo et al. (2017) quantifies the thermal conductivity of fission-product damaged graphite by an average burnup making it independent of grain radius. We show that the dose (measured in displacement per atoms) of graphite immediately outside a fuel grain is larger for larger grains.

In this work, we first clarify the effect of the grain size on the dose in the surrounding graphite (Section 2), then a multiphysics fuel grain model is introduced in Section 3, in Section 4 a worst case estimate is used to demonstrate that the effect of thermal conductivity degradation of graphite is negligible on TREAT performance, and in Section 5 the possibility of fuel grain melting is explored. Our conclusions are presented in Section 6.

2. Effect of grain size on graphite radiation damage

This Section demonstrates that the dose of fission fragment damage in the graphite surrounding fuel grains increases with increasing grain size. The received dose increases with the number of fission fragments escaping from the fuel, but decreases with the irradiated graphite volume. With increasing grain size, the number of fission fragments born in the grain and the volume of irradiated graphite increases, while the escape probability from the fuel decreases. We examine the influence of these three factors on the magnitude of the dose received in the graphite surrounding fuel grains.

Mo et al. (2017) evaluates the thermal conductivity of graphite at the average burnup, i.e. the energy deposited per unit volume, neglecting the effect of grain size on the received dose. This Section demonstrates the relationship between graphite dose and fuel grain radius. We investigate under which conditions Mo et al.'s (2017) approach is valid and if these conditions are met for the TREAT LEU design.

2.1. Analytical model of graphite radiation damage around fuel grains

We first focus on a simplified discussion of the mechanisms leading to an increase of dose rate with increasing grain size. The underlying assumption of this simple model is that the received dose is proportional to the fission fragment rate density, $j(R)$, defined as the number of fission fragments that leave the fuel grain divided by the irradiated graphite volume. An analytical formula is derived for $j(R)$ under the assumption that fission fragments are born uniformly and isotropically in the fuel and have fixed, but different ranges in the fuel and graphite, respectively. The resulting formula exposes the mechanism by which larger fuel grains accumulate more radiation damage in the graphite surrounding them.

If we denote the fission rate density by F, the fuel grain radius by R, the probability that a fission fragment born uniformly and isotropically in the fuel grain will leave the fuel grain by $\eta(R)$, and the affected graphite volume by $V_{\mathrm{G,irr}}$, then $j(R)$ is given by[2]:

$$j = \frac{8}{3}\pi F R^3 \frac{\eta(R)}{V_{\mathrm{G,irr}}}, \tag{1}$$

where F does not depend on R because the total volume of fuel in the reactor is constant. As the grain radius increases, the number of fuel grains in the reactor decreases to keep the total fuel volume constant. For a given reactor power, the fission rate density is computed as the ratio of total reactor power and total fuel volume and hence does not depend on the grain radius. The volume of irradiated graphite is computed by:

$$V_{\mathrm{G,irr}} = \frac{4}{3}\pi\left(\left(\tau_g + R\right)^3 - R^3\right) = \frac{4}{3}\pi\left(\tau_g^3 + 3\tau_g^2 R + 3\tau_g R^2\right), \tag{2}$$

where τ_g is the range of fission products in graphite. One can derive an expression for $\eta(R)$ by assuming that fission fragments are born uniformly and isotropically, and have a constant range τ_f in the fuel (Harrison, 1953):

$$\eta(R) = \frac{3\tau_f}{4R}\left(1 - \frac{\tau_f^2}{12R^2}\right). \tag{3}$$

Fission fragments are more likely to be born in the outer shells of a fuel grain than close to the center due to these shells' proportionally larger volume and consequently the escape probability only decreases linearly with grain radius R.

Combining Eqs. (1)–(3) gives:

$$\frac{j}{F} = \frac{3\psi}{2}\frac{1 - \frac{\psi^2 x^2}{12}}{x^2 + 3x + 3}, \tag{4}$$

where $x = \frac{\tau_g}{R}$ and $\psi = \frac{\tau_f}{\tau_g}$. The ratio of the fission fragment ranges in fuel and graphite, ψ, can be estimated using the Bragg-Kleeman rule (Knoll, 1979):

$$\psi = \frac{\rho_g}{\rho_f}\sqrt{\frac{A_f}{A_g}} \approx 0.7, \tag{5}$$

where $\rho_g = 1.72\,\frac{\mathrm{g}}{\mathrm{cm}^3}$ and $\rho_f = 10.96\,\frac{\mathrm{g}}{\mathrm{cm}^3}$ are graphite and fuel density, and $A_f \approx 238$ and $A_g \approx 12$ are mass numbers of the fuel and graphite, respectively. Expanding j/F given by Eq. (4) into a Taylor series and truncating it after the linear term results in a formula valid for small values of x:

$$\frac{j}{F} = \psi\left[\frac{1}{2} - \frac{x}{2}\right] + \mathcal{O}(x^2). \tag{6}$$

For sufficiently small fission fragment ranges τ_f and/or large fuel grains, the linear term in Eq. (6) can be neglected and the specific dose rate becomes independent of R; it is in this limit that Mo et al.'s (2017) treatment is valid. However, when $x/2$ becomes comparable in magnitude to $1/2$, then this assumption is not applicable. For TREAT, τ_f and R are of the same order of magnitude, and hence the linear correction is not negligible and Mo et al.'s (2017) assumption does not apply. Furthermore, it can easily be seen from Eq. (4) that j/F monotonically decreases with increasing value of x; an increase in x corresponds to a decrease in grain radius. Therefore, larger fuel grains incur larger dose rates in the graphite.

[2] A factor of 2 is multiplied to account for the production of two fission fragments per fission event.

2.2. Numerical investigation of graphite radiation damage around fuel grain

While the analytical model exemplifies the physical mechanism leading to more damage around larger grains, it neglects that most vacancy-interstitial pairs are produced by secondary knock-on atoms and not by the primary fission products. To assess the importance of secondary knock-on atoms, we perform three-dimensional binary-collision Monte-Carlo (BCMC) (Ziegler et al., 1996) simulations for a single spherical UO_2 fuel grain and sufficient graphite such that no fission fragment or knock-on reaches the outer boundary of the simulation domain. Fuel grain separation in TREAT is significantly larger than the range of knock-on atoms, so that the fuel grain can be considered isolated. The BCMC calculations are performed with the three-dimensional BCMC code *MyTRIM* (Schwen et al., 2009; Schunert et al., 2017). Fission sites are sampled uniformly within the fuel grain and the number of vacancies in the fuel and graphite are tallied. We use a displacement threshold of 30 eV throughout because it has been found to represent both graphite and UO_2 reasonably well (McKenna et al., 2016; Dacus et al., 2019). It is noted that the accuracy of the displacement threshold is not important for the conclusions of this work because of the way in which the BCMC results are used. In this section, we show that larger grains incur larger damage in the graphite surrounding it. In the later development, the dose rate distributions are used to determine which portion of the graphite receives fission fragment damage. The displacement threshold does not affect either of these results significantly.

The local dose in displacements per atom (dpa) is obtained as a function of the distance from the fuel grain center. To this end, displacement events are tallied on a uniform spherical mesh. Fig. 1 shows the duration of TREAT operation it takes for the dose to reach 0.01 dpa, instead of plotting the dpa rate density versus distance. The threshold of 0.01 is chosen because it marks the onset of significant degradation of thermal conductivity in graphite (Snead and Burchell, 1995); the conversion to operation time is based on the historical values of 2,600 GJ of TREAT reactor energy release over 35 years of operation. The onset of degradation of thermal conductivity immediately outside of the fuel grains may be expected after 4, 6, and 10 months of operation for grains of radius 20, 10, and 5 μm, respectively, assuming TREAT's operational parameters do not change. This demonstrates that larger fuel grains accumulate radiation damage in the graphite immediately surrounding them faster than smaller grains.

The intensity of radiation damage around TREAT fuel grains significantly depends on their size: higher doses are received in the graphite immediately outside of larger fuel grain. This conclusion is in contrast to Mo et al. (2017), where the degradation of thermal conductivity is taken to depend only on the average burnup but not on the size of the fuel grains.

3. Fuel grain model

We develop a multiphysics model of the transient response of a fuel grain. This model computes the time evolution of temperature and power density for a given irradiation damage state and initial reactivity insertion. Degradation of thermal conductivity is a possible cause of the reduction in TREAT's performance. In practice, however, degradation of thermal conductivity is only important because it reduces TREAT's operational envelope, i.e. reduces possible energy deposition into an experiment, or increases fuel grain temperatures beyond safety limits. Therefore, it is more useful to quantify the importance of irradiation effects on the LEU TREAT design based on energy depositions and peak fuel temperature, and not based on the reduction of thermal conductivity itself.

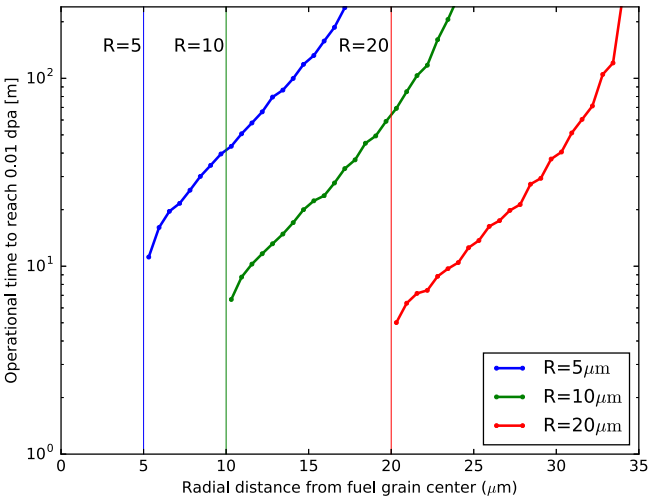

Fig. 1. Cumulative operational time (in months) for the graphite around the fuel grain to receive a dose of 0.01 dpa for grain radii of 5, 10, and 20 μm.

The model developed in this work is patterned after the model proposed by Zabriskie et al. (2019) and is implemented using the MOOSE-based Rattlesnake radiation transport code (Gaston et al., 2015; Wang et al., 2019). The model utilizes a zero-dimensional point-kinetics representation of the neutron population dynamics and a one-dimensional, spherically symmetric representation of heat transfer of the average fuel grain. By definition, the average fuel grain receives a power that is equal to the total reactor power divided by the number of fuel grains. Focusing on a single representative grain has two important ramifications: first the influence of temperature distribution on feedback is neglected, and second information about the hot and cold grains are lost. By comparison against measured HEU TREAT transients, this section shows that predicted power levels are accurate to within measurement uncertainties and that predicted peak timing is about 25% off compared with an experimental timing uncertainty of roughly 10%. However, the ultimate conclusion of this work does not hinge on the exact numerical values obtained from the computational model, but on the fact that adverse effects of radiation on small fuel grains is insignificant for plausible TREAT operating parameters even when generous uncertainty margins are considered.

A point-kinetics model (Duderstadt, 1976) is a system of ordinary differential equations describing the evolution of TREAT's power density $p(t)$ over time t:

$$\begin{aligned}
&\frac{dp}{dt} = \frac{\xi(T_f,T_g)-\beta}{\Lambda}p(t) + \sum_{i=1}^{6}\lambda_i C_i \\
&\frac{dC_i}{dt} = -\lambda_i C_i + \frac{\beta_i}{\Lambda}p(t) \\
&p(0) = p_0 \\
&C_i(0) = \frac{\beta_i}{\Lambda\lambda_i}p_0,
\end{aligned} \tag{7}$$

where C_i is the delayed neutron precursor concentration of group i, β_i is the fraction of neutrons born as delayed neutrons from precursor in group i, $\beta = \sum_{i=1}^{6}\beta_i$, λ_i is the decay constant of precursor group i, $\xi(T_f, T_g)$ is the reactivity that depends on the fuel and graphite temperatures T_f and T_g, respectively, and Λ is the neutron mean generation time.

Temperature feedback to the point kinetics equations is rendered through the reactivity ξ that depends on fuel and graphite temperatures. The temperature field is described by the heat conduction equation:

$$\rho c_p(r,T)\frac{\partial T(r,t)}{\partial t}-\frac{1}{r^2}\frac{\partial}{\partial r}\left[r^2k(r,T)\frac{\partial T(r,t)}{\partial r}\right]=\dot{q}'''(r,t), \tag{8}$$

where ρ is the density, $c_p(r,T)$ is the specific heat capacity, $k(r,T)$ is the thermal conductivity, and $\dot{q}'''$ is the volumetric heat source stemming from fission reactions. Initial and boundary conditions for the heat equation are given by:

$$\begin{aligned} T(r,0) &= 300\text{K (assumed room temperature)} \\ \tfrac{\partial T}{\partial r} &= 0 \text{ at } r=0 \text{ and } r=R_{\max}, \end{aligned} \tag{9}$$

where $R_{\max}$ is the outer radius of the domain:

$$R_{\max}=\sqrt[3]{\sigma+1}R, \tag{10}$$

and $\sigma=626$ is the ratio of graphite to fuel volume. The adiabatic boundary at $r=R_{\max}$ implies that fuel grains do not interact with each other. This is a valid assumption for short transients less than 10 s in length. The fuel grain and graphite temperatures are computed as averages over their respective subdomains:

$$\begin{aligned} T_f(t) &= \frac{3\int_0^R r^2T(r,t)dr}{R^3} \\ T_g(t) &= \frac{3\int_R^{R_{\max}} r^2T(r,t)dr}{\left(R_{\max}^3-R^3\right)}. \end{aligned} \tag{11}$$

The material properties in Eq. (8) with the exception of the thermal conductivity in graphite, are taken as un-irradiated from Mo et al. (2015). Denoting fuel with subscript f and graphite with subscript g : $\rho_g=1.72\frac{\text{g}}{\text{cm}^3}$, $\rho_f=10.96\frac{\text{g}}{\text{cm}^3}$. The heat capacities $c_{p,f}$ and $c_{p,g}$ are computed by:

$$\begin{aligned} c_{p,f} &= 52.1743+87.951\omega-84.2411\omega^2+31.542\omega^3-2.6334\omega^4-0.7139\omega^{-2} \\ c_{p,g} &= \frac{1}{11.07T^{-1.644}+0.0003688T^{0.0219\text{T}}}, \end{aligned} \tag{12}$$

where $\omega=T/1000$, and k_f is given by:

$$k_f=\frac{100}{6.548+23.533\omega}+\frac{6400}{\omega^{5/2}}\exp\left(\frac{-16.35}{\omega}\right). \tag{13}$$

The reduction of thermal conductivity in the fuel grain is neglected in this work because it is small compared with the reduction of graphite's thermal conductivity. A sensitivity study for a recorded TREAT power trace showed that including the degradation of fuel thermal conductivity leads to an increase of the predicted maximum fuel temperature of less than 50 K. This difference has no significant impact on the conclusions of this work. The thermal conductivity of graphite, k_g, is independent of temperature but reduces under fission product irradiation. The effect of fast neutron irradiation in the graphite is neglected because the thermal conductivity of neutron irradiated graphite is very close to that of fresh TREAT graphite and intrinsic defects "outweigh the effects of neutron irradiation (Mo et al., 2015)."

The point kinetics properties in Eq. (7), i.e. $\beta_i,\lambda_i,\Lambda$, are computed using Rattlesnake's improved quasi-static capability (IQS) (Prince and Ragusa, 2019) for the TREAT model described in Zabriskie et al. (2019).

The heat source q''' is computed in exactly the same way as described in Zabriskie et al. (2019). Energy from fission is released as short-range fission fragments (86.8%) with ranges on the order of micrometers, and as electrons, photons, and neutrons (13.2 %) with ranges on the order of millimeters and centimeters. The latter fraction of the energy release is thus deposited uniformly in the domain. Energy released as fission fragments is not uniformly distributed in the fuel grain, because fission fragments ballistically relocate from the fission site. This mechanism is termed ballistic heat transfer and is important if the graphite surrounding the fuel is significantly damaged by fission products (Zabriskie et al., 2019).

The most important difference of this heat source compared to Mo et al.'s (2015) model is that electron, photon, and neutron energy release from fission is uniformly distributed in the whole domain, not just in the fuel. This is a more realistic model because it takes into account the relatively long mean free path of electrons, neutrons, and photons. The range of electrons is of the order of several millimeter and thus about ten times larger than the expected spacing between fuel grains ($100-350$ μm), and the mean free path of photons and neutrons is of the order of a several centimeter and thus about one hundred times larger than the grain spacing. Therefore, the heat source distribution originating from these long-range fission products is essentially uniform. Calculations show that depositing all energy in the fuel grain leads to an increase of the prediction of the peak temperature of the EOS[3] transient in the current TREAT HEU configuration by 327 K (1673 compared with Mo et al.'s (2015) value of 2000 K Mo et al., 2015). The margin to fuel melting may therefore be significantly larger than predicted by Mo et al. (2015).

A linear reactivity feedback model is used exclusively within this work. It is given by:

$$\xi(T_f,T_g)=\xi_0+c_f(T_f-300)+c_g(T_g-300), \tag{14}$$

where c_f and c_g are the fuel and graphite feedback coefficients of reactivity, respectively. The approximation of the linear feedback model is to use feedback coefficients of reactivity that are independent of T_f and T_g. The reactor feedback is sensitive to the core configuration (size, assembly configuration), details of the temperature distribution in the core, and the deployed experiment. In the absence of any of these details for the LEU configuration, an attempt to improve accuracy of the feedback model would result in adding spurious information rather than a better match to reality.

The adequacy of the PKE model is demonstrated by re-creating the measured EOS power trace provided by Mo et al. (2015). We use $c_g=2.25\times10^{-4}\text{K}^{-1}, c_f=0$, $\xi_0=0.043$, and the value of the initial power density $p_0=14.24\text{W/cm}^3$; these are found by fitting an exponential to the measured EOS curve before significant feedback occurs and taking $t\to0$. The two power traces are compared in Fig. 2. The prediction of the model developed for this work matches the measured EOS power well within its limitations. The magnitude of the power peak is predicted to within 2 %, while the time of the power peak is predicted to within 25 % (0.12 versus 0.16 s). TREAT measurements carry significant uncertainties both on power level and timing, namely 20% and 10%, respectively (DeHart et al., 2017). The predicted peak power is well within the measurement uncertainty, while the predicted peak time is not.

The reason for the relatively poor prediction of the peak timing is conjectured to be the feedback model and not the point kinetics data. Both power traces match well during the initial reactor period before feedback becomes significant, but diverge once feedback becomes important. The feedback model introduces two significant assumptions, first it only includes linear temperature terms and second it represents the entire reactor by a single power density and average temperature, i.e., the "average" grain. As argued before, these shortcomings are of little consequence and do not invalidate any conclusions of this study.

[3] The EOS transient is a historical transient performed with the HEU TREAT configuration. When the EOS transient is discussed in this work, we always refer to the TREAT HEU configuration. The HEU configuration is used only when modeling the EOS transient.

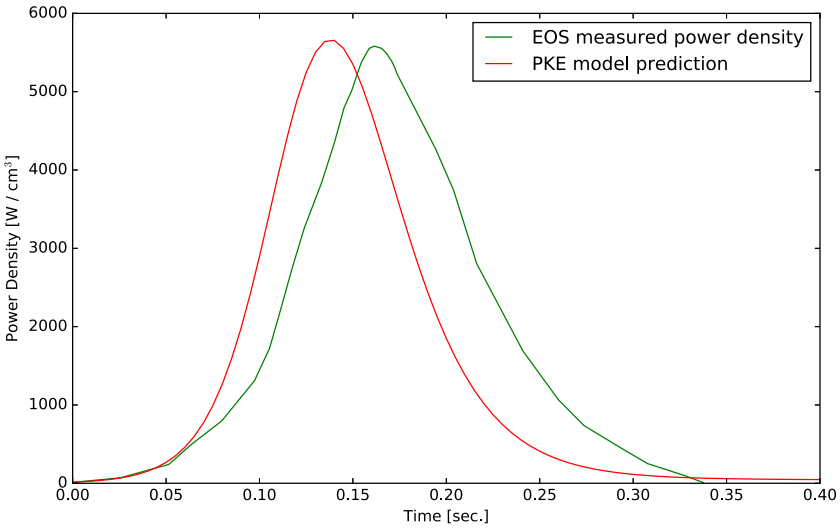

Fig. 2. The measured power trace of the EOS transient from Mo et al. (2015) compared with the prediction of the model developed for this work.

4. Asymptotic degradation of TREAT's transient capabilities

This Section investigates the limits of reduction of LEU TREAT's transient capabilities. TREAT's performance cannot deteriorate below a saturation value, because thermal conductivity of graphite has a lower saturation value below which it cannot decrease (Gallego and Burchell, 2011). An effective lower limit of thermal conductivity of graphite under irradiation implies that transient capabilities cannot decrease further once the asymptotic value of thermal conductivity is reached everywhere in the fission fragment affected region; radiation damage from fast neutrons far away from a grain does not amount to significant levels of dose and is therefore neglected. In this Section, we posit that a sufficient dose is deposited everywhere in fission fragment irradiated regions depicted in Fig. 1 to saturate the degradation of thermal conductivity.

The degradation of graphite's thermal conductivity depends on the dose and the irradiation temperature. Both the saturated thermal conductivity and the dose at which it is reached are reduced with irradiation temperature. The saturation thermal conductivity is reported by Mo et al. (2015), Mo et al. (2017), Snead and Burchell (1995), Bell et al. (1962), Gallego and Burchell (2011) and Vreeling et al. (2008) giving values of 2.5, 20.0, between 10 and 14, 1.4, and 20% of the un-irradiated values, respectively. Within the list of reviewed works, (Bell et al., 1962; Gallego and Burchell, 2011) report the largest asymptotic reduction of thermal conductivity to 1.4% of the un-irradiated value. We therefore assume that graphite retains at least 1% of its thermal conductivity regardless of the received dose.

We characterize TREAT's transient capabilities by the total energy density e deposited during a transient, because energy deposition relates proportionally to energy deposition into the experiment, denoted by E. The ultimate purpose of TREAT is to generate a prescribed E for a given experimental setup. Total energy density e is defined as

$$e = \int_{t_b}^{t_e} p(t)dt, \tag{15}$$

where t_b and t_e are beginning and ending times of a transient. Total energy density is directly related to the energy deposited in an experiment by the power coupling factor μ:

$$E = \mu V e, \tag{16}$$

where V is the reactor volume. The exact value of the power coupling factor depends on the experiment being conducted, but it is reasonably insensitive to changes in e as long as the experiment geometry is fixed; under this assumption E is proportional to e. In the absence of exact geometrical specification of the experiment and core, we use e as a surrogate figure of merit in lieu of E. The reduction of performance is measured as the difference of energy deposition for un-irradiated fuel, denoted by subscript u, and saturated irradiated state, denoted by subscript s:

$$\Delta e = 1 - \frac{e_s}{e_u}. \tag{17}$$

We perform transient simulations over a realistic range in parameter space varying the grain radius, saturation value of thermal conductivity, reactivity insertion, and the fuel feedback coefficient. Increasing the grain radius or decreasing k_s/k_u increases Δe because of less effective heat rejection from the fuel grain and hence larger fuel temperatures; increasing the reactivity insertion increases Δe because it creates a faster pulse that deposits large amounts of energy in the fuel grain leading to larger fuel temperatures; increasing the fuel feedback coefficient increases the strength of the Doppler feedback and thus penalizes large fuel temperatures.

Reactivity insertions ξ_0 are varied between 0.01 to 0.061; the upper limit of $\xi_0 = 0.061$ is obtained by requiring that TREAT's LEU configuration matches the HEU configuration's energy deposition at its maximum reactivity insertion of $\xi_{0,\text{HEU}} = 0.05$ for un-irradiated graphite. Various fuel grain radii, $R = 5, 10, 20\mu$m, are considered and the strength of the Doppler feedback is varied between $c_f/c_g = 0, \ldots, 1$. It is unrealistic for c_f to be larger than c_g because of the comparatively small amount of fuel. Numerical simulations found that $c_f \approx 0.2 c_g$ for LEU (Zabriskie et al., 2019). The graphite feedback coefficient for LEU TREAT is computed to be roughly $c_g \approx 2 \times 10^{-4}\text{K}^{-1}$ for a realistic LEU TREAT geometry (Johnson and Ortensi, 2019) using the MC21 Monte-Carlo (Griesheimer et al., 2015) code. This feedback coefficient is of the same order of magnitude as for the current HEU TREAT geometry. By numerical experiment, the thermal conductivity in the un-irradiated graphite volume is un-important for transient performance because it is much larger than the thermal conductivity of irradiated graphite. We follow Mo et al. (2015) and fix the thermal conductivity of fresh graphite at $k_u = 41.8 \frac{\text{W}}{\text{Km}}$ and limit the saturated value k_s to be larger than or equal to 1 % of this value.

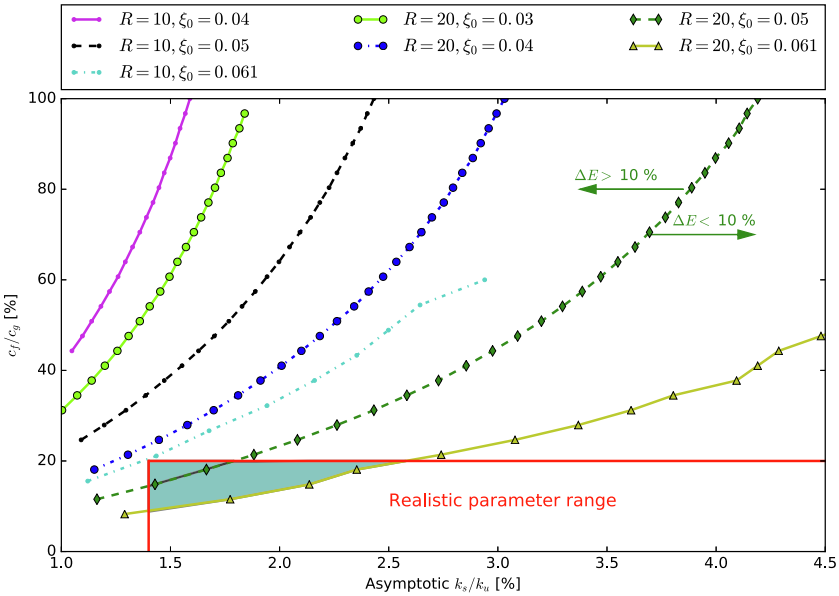

Fig. 3. Level curves for $\Delta e = 10\%$ plotted versus k_s/k_u and c_f/c_g for different grain radii R and reactivity insertions in percent.

The results of the performed study indicate that the asymptotic reduction of transient energy deposition, Δe, is likely to be less than 10%. Level curves for $\Delta e = 10\%$ are plotted in Fig. 3 versus k_s/k_u and c_f/c_g for various combinations of grain radii and initial reactivity insertions. The area to the right of each curve experiences less than 10% of reduction in energy deposition, while the area to the left of each curve sees more than 10% of reduction in energy deposition. A reasonable range of the parameters k_s/k_u and c_f/c_g is marked by the red box.

Even under the assumption that thermal conductivity reduces to 1.4% of its un-irradiated thermal conductivity (smallest value reported in Gallego and Burchell (2011)) and $c_f \approx 0.2c_g, \Delta e \geqslant 10\%$ is only realized if the grain size is quite large, $R = 20\mu$m, and the reactivity insertion is large, $\xi_0 \geqslant 5\%$. The region of the parameter space that can realistically see degradation of $\Delta e \geqslant 10\%$ is shaded in teal in Fig. 3; for the case of $\xi_0 = 0.061$ we require $R = 20$ µm, $c_f > 0.1c_g$ and $k_s < 0.025k_u$.

Reduction of grain size to $R = 5\mu$m avoids significant degradation of TREAT's transient capabilities with radiation damage. In the conducted numerical experiment, models with $R = 5\mu$m do not exhibit significant performance degradation within a reasonable parameter space, $k_s/k_u \geqslant 1\%$ and $c_f \leqslant c_g$ and are therefore not shown in Fig. 3. As expected from the discussion in Section 2, resistance to radiation-induced performance degradation increases with decreasing grain radii. In practice, any grain radius $R \leqslant 10\mu$m would be expected to perform adequately because the assumption of the presented study are conservative: fully saturated thermal conductivity, reduction to very low asymptotic thermal conductivity of graphite, and strong Doppler feedback.

5. Possibility of fuel grain melting

This Section discusses the possibility of fuel melting in the scenarios hypothesized in Section 4. The largest fuel temperature is obtained with large reactivity insertion, large grains, small asymptotic thermal conductivity of graphite, and small fuel feedback coefficient. In this Section, we investigate maximum fuel temperatures during transients for a reactivity insertion of $\xi_0 = 0.061$ under the condition that radiation damage has reduced the thermal conductivity to $k_s = 0.01k_u$; these are the most restrictive conditions for these parameters. This study includes radii of 5 µm and 20 µm and fuel feedback coefficients of $c_f = 0$ and $c_f = 0.2c_g$ to explore the dependence of the maximum fuel temperature on these parameters.

Table 1
Maximum fuel grain temperatures in Kelvin and homologous temperature (temperature divided by UO_2 melting temperature, in parenthesis) during a transient with $\xi_0 = 0.061$ and irradiated graphite with $k_s = 0.01k_u$.

	$c_f = 0.2$	$c_f = 0$
R = 5 µm	728 (0.23)	820 (0.26)
R = 20 µm	946 (0.3)	1764 (0.56)

In Table 1 maximum fuel grain temperatures and associated homologous temperatures (temperature divided by melting temperature of UO_2) for 5 µm and 20 µm and fuel feedback coefficients of $c_f = 0$ and $c_f = 0.2c_g$ are presented. The maximum temperature in LEU TREAT even under extremely conservative parameter choices does not exceed a homologous temperature of 0.6. Our results differ from Mo et al. (2015), which predict fuel temperatures that are several hundred Kelvin higher, and thus the margins to fuel melting is predicted to be much smaller. The main difference of this work compared with Mo et al. (2015) is the use of a heat source distribution that takes into account the non-local deposition of energy released into electrons, γ-rays, and fast neutrons. Based on these results, we conclude that fuel melting is implausible for grains with $R \leqslant 20$ µm.

6. Conclusions

The main finding of this work is that radiation damage from fission fragments does not set a lower limit on LEU TREAT fuel grain sizes. Earlier work in Mo et al. (2017) reports that smaller grains are susceptible to radiation damage induced reduction of thermal conductivity that can lead to performance degradation and potential fuel melting. We conclude the opposite: smaller grains are more resilient to radiation damage and experience a smaller reduction of thermal conductivity in the graphite surrounding them during fission fragment irradiation.

The results of this study indicate that degradation of LEU TREAT's transient capabilities is more severe for larger fuel grains.

Conduction of heat from the fuel grain into the bulk graphite should be thought of as an arrangement of serial conductors. For serial conductors, the thickness of the fission-fragment irradiated region outside the fuel grains and the radiation dose are relevant, while the volume fraction is not. The thickness of the irradiated layer does not change with fuel grain radius, but the irradiation dose increases with increasing fuel radius. The assumption of size-independent irradiation doses around fuel particles is valid only if the ratio of fission product range and grain radius is sufficiently close to zero. This condition is not satisfied for TREAT.

For the purpose of investigating the impact of grain size on thermal conductivity and TREAT performance, we introduce a multiphysics model consisting of the reactor point kinetics equations and a spherically symmetric heat conduction problem. Radiation damage is simulated by the three-dimensional binary collision Monte-Carlo code *MyTRIM*. Feedback is computed using an average fuel grain and graphite temperature in the form of a linear feedback model. We show that the model is sufficiently accurate for the purpose of this work by comparing predictions to measured EOS transient data.

Within a reasonable parameter space of initial reactivity insertion, fuel grain radius, fuel and graphite feedback coefficients, and thermal properties of graphite, the energy deposition in TREAT cannot deteriorate by more than $10-15\%$. Performance degradation is naturally limited by the lower limit of thermal conductivity of irradiated graphite. Reduction in deposited energy by more than 10% is only possible for outlier cases with large grain radii and large initial reactivity insertions. Maximum fuel temperatures computed using the coupled models do not exceed 60% of the melting temperature of UO_2, and hence fuel melting can be ruled out.

This work comes to the conclusion that radiation damage induced reduction of thermal conductivity does not set a lower limit for fuel grain radii in LEU TREAT as long as the typical separation of grains is large compared with the range of fission fragments in graphite. We find that there is no competition in the choice of the grain radius between radiation damage and heat rejection characteristics. As long as other factors not considered in this work do not set lower limits on the fuel grain radii, this study suggests that TREAT LEU reactor designers have the freedom to reduce fuel grain radii to rule out adverse effects on the reactor's performance from additional Doppler feedback.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

This work was supported through the INL Laboratory Directed Research & Development Program under Grant No. 16-010. This manuscript has been authored by Battelle Energy Alliance, LLC under Contract No. DE-AC07-05ID14517 with the U.S. Department of Energy. The United States Government retains and the publisher, by accepting the article for publication, acknowledges that the United States Government retains a nonexclusive, paid-up, irrevocable, world-wide license to publish or reproduce the published form of this manuscript, or allow others to do so, for United States Government purposes.

References

Bell, J.C., Bridge, H., Cottrell, A.H., Greenough, G.B., Reynolds, W.N., Simmons, J.H.W., 1962. Stored energy in the graphite of power-producing reactors. Philos. Trans. R. Soc. London. Series A, Math. Phys. Sci. 254. https://doi.org/10.1098/rsta.1962.0002.

Connaway, H.M., et al., 2016. Analysis of the treat leu conceptual design, Tech. Rep. ANL/RTR/TM-16/1, Argonne National Laboratory..

Dacus, B., Beeler, B., Schwen, D., 2019. Calculation of threshold displacement energies in UO_2. J. Nucl. Mater. 520, 152–164. https://doi.org/10.1016/j.jnucmat.2019.04.002. URL: http://www.sciencedirect.com/science/article/pii/S0022311518312571.

DeHart, M.D., Baker, B.A., Ortensi, J., 2017. Interpretation of energy deposition data from historical operation of the transient test facility (treat). Nucl. Eng. Des. 322, 504–521. https://doi.org/10.1016/j.nucengdes.2017.07.019. URL: http://www.sciencedirect.com/science/article/pii/S0029549317303515.

Duderstadt, J.J., Hamilton, L.J., 1976. Nuclear Reactor Analysis. John Wiley & Sons Inc.

Freund, G., et al., 1960. Design summary report on the transient reactor test facility (treat), Tech. Rep. ANL-6034, Argonne National Laboratory.

Gallego, N., Burchell, T., 2011. A review of stored energy release of irradiated graphite, Tech. Rep. ORNL/TM-2011/378. Oak Ridge National Laboratory.

Gaston, D.R., Permann, C.J., Peterson, J.W., Slaughter, A.E., Andrs, D., Wang, Y., Short, M.P., Perez, D.M., Tonks, M.R., Ortensi, J., Zou, L., Martineau, R.C., 2015. Physics-based multiscale coupling for full core nuclear reactor simulation. Ann. Nucl. Energy 84, 45–54. https://doi.org/10.1016/j.anucene.2014.09.060.

Griesheimer, D., Gill, D., Nease, B., Sutton, T., Stedry, M., Dobreff, P., Carpenter, D., Trumbull, T., Caro, E., Joo, H., Millman, D., 2015. Mc21 vol 6.0 – a continuous-energy monte carlo particle transport code with integrated reactor feedback capabilities. Ann. Nucl. Energy 82, 29–40. https://doi.org/10.1016/j.anucene.2014.08.020. joint International Conference on Supercomputing in Nuclear Applications and Monte Carlo 2013, SNA + MC 2013. Pluri- and Trans-disciplinarity, Towards New Modeling and Numerical Simulation Paradigms. http://www.sciencedirect.com/science/article/pii/S0306454914004058..

Harrison, R., 1953. Effect of particle size fission-fragment damage for particles of fissionable material dispersed in a matrix, Tech. Rep. TID-10017. Battelle Memorial Inst.

Johnson, M., Ortensi, J., 2019. Analysis of treat leu design using mammoth, internal Memorandum..

Knoll, G.F., 1979. Radiation Detection and Measurement. John Wiley & Sons Inc.

McKenna, A., Trevethan, T., Latham, C., Young, P., Heggie, M., 2016. Threshold displacement energy and damage function in graphite from molecular dynamics. Carbon 99, 71–78. https://doi.org/10.1016/j.carbon.2015.11.040. URL: http://www.sciencedirect.com/science/article/pii/S0008622315304498.

Mo, K., Yun, D., Yacout, A., Wright, A., 2015. Heat transfer simulations of the UO_2 particle-graphite system in treat fuel. Nucl. Eng. Des. 293.

Mo, K., Miao, Y., Kontogeorgakos, D., Connaway, H., Wright, A., Yacout, A., 2017. Effect of reactor radiation on the thermal conductivity of treat fuel. J. Nucl. Mater. 487.

Prince, Z.M., Ragusa, J.C., 2019. Multiphysics reactor-core simulations using the improved quasi-static method. Ann. Nucl. Energy 125, 186–200. https://doi.org/10.1016/j.anucene.2018.10.056. URL: http://www.sciencedirect.com/science/article/pii/S030645491830584X.

Schunert, S. et al., 2017. Heat source characterization in a treat fuel particle using coupled neutronics binary collision monte-carlo calculations. In: M&C 2017 International Conference on Mathematics & Computational Methods Applied to Nuclear Science & Engineering. Korean Nuclear Society.

Schwen, D., Huang, M., Bellon, P., Averback, R.S., 2009. Molecular dynamics simulation of intragranular Xe bubble re-solution in UO_2. J. Nucl. Mater. 392 (1), 35–39. https://doi.org/10.1016/j.jnucmat.2009.03.037.

Snead, L., Burchell, T., 1995. Thermal conductivity degradation of graphites due to neutron irradiation at low temperature. J. Nucl. Mater. 224.

Vreeling, J., Wouters, O., van der Laan, J., 2008. Graphite irradiation testing for HTR technology at the high flux reactor in petten. J. Nucl. Mater. 381.

Wang, Y., Schunert, S., Laboure, V., 2019. Rattlesnake theory manual, Tech. Rep. INL/EXT-15-37337, Idaho National Laboratory..

Zabriskie, A., Schunert, S., Schwen, D., Ortensi, J., Baker, B., Wang, Y., Laboure, V., DeHart, M., Marcum, W., 2019. A coupled multiscale approach to treat leu feedback modeling using a binary-collision monte-carlo-informed heat source. Nucl. Sci. Eng. 193 (4), 368–387.

Ziegler, J., Biersack, J., Littmark, U., 1996. SRIM – The Stopping and Range of Ions in Solids. Pergamon Press.