Determining the minimum required uranium carbide content for HTGR UCO fuel kernels

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ABSTRACT

Three important failure mechanisms that must be controlled in high-temperature gas-cooled reactor (HTGR) fuel for certain higher burnup applications are SiC layer rupture, SiC corrosion by CO, and coating compromise from kernel migration. All are related to high CO pressures stemming from O release when uranium present as UO₂ fissions and the O is not subsequently bound by other elements. In the HTGR kernel design, CO buildup from excess O is controlled by the inclusion of additional uranium apart from UO₂ in the form of a carbide, UC_x and this fuel form is designated UCO. Here general oxygen balance formulas were developed for calculating the minimum UC_x content to ensure negligible CO formation and applied to 15.5% enriched UCO taken to 16.1% actinide burnup. Required input data were obtained from CALPHAD chemical thermodynamic models and the Serpent 2 reactor physics and depletion analysis tool. The results are intended to be more accurate than previous estimates by including more nuclear and chemical factors, in particular the effect of transmuted Pu and Np oxides on the oxygen distribution as the fuel kernel composition evolves with burnup.

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1. Introduction

It has been established that pressure vessel failure [1] and CO corrosion [2-4] as well as tristructural isotropic (TRISO) coating compromise from kernel migration [2, 3] are important failure mechanisms for high temperature gas-cooled reactor (HTGR) fuel. These phenomena can be predicted as a function of CO formation using thermodynamic arguments based on the initial composition of the fuel kernel, where O (liberated when uranium present as UO_2 fissions) reacts with the surrounding carbon if not bound by other elements with higher oxygen affinity [2, 3, 5]. In the uranium-carbon-oxygen (UCO) kernel design, CO formation is mitigated by the inclusion of additional uranium as a carbide, UC_x . Conventionally, the UC_x phase is substoichiometric UC_2 designated UC_{2-x} . However, any form of UC_x in the presence of excess C available in the buffer at HTGR temperatures will form, from the standpoint of thermodynamics, UC_{2-x} (or U_2C_3 depending on temperature) and therefore the arguments presented here in this work apply to UC_x generally.

The added UC_x provides additional material that will form oxides more readily than carbon. Prior efforts in the literature have been made to estimate the required proportion of UC_x relative to UO_2 using simplified chemistry [2, 3]. Advances in computational capabilities and the development of new more sophisticated thermodynamic and burnup models merit revisiting these calculations. By including more nuclear and chemical factors it is the aim of this work to produce higher fidelity predictions. Further, the effect of transmutation products, like Pu and Np, on the oxygen distribution are included for the first time.

One possible benefit of refining the calculation is the option for minimizing the amount of UC_x in the UCO kernel so that more of the metallic fission products are oxidized, which more effectively retains them in the kernel relative to their carbide forms. However, UCO kernel designers must consider possible deviations in kernel properties for a given fabrication process and balance the negative impact of CO formation against any positive impact of reducing the inventory of various metal carbides. The purpose of this work is not to recommend a UCO kernel composition, which would be dependent on reactor design and fabrication control, but rather to outline an approach for determining the minimum UC_x content in UCO-TRISO fuel necessary for mitigating CO production using state-of-the-art methods. This is accomplished by combining CALPHAD (CALculation of PHase Diagrams) thermodynamic modeling [6-9] with the Monte Carlo neutron transport and depletion tool, Serpent 2,[10] used for burnup simulations.

2. Approach

For this paper, we define the limiting lower UC_x content in a UCO kernel to be the minimum UC_x needed for a given burnup to maintain an acceptably low oxygen potential, μ_{O_2} , to avoid excessive CO pressures (p_{CO}), as well as the so-called “amoeba effect” due to thermal migration of UO_2 into the buffer region and ultimately the IPyC and SiC layers. It has been suggested that lanthanide diffusion through SiC is avoided when these elements and Y, collectively referred to hereafter as Ln’s, are bound and

immobilized as oxides; experimental evidence supports this [2, 8]. It has been conjectured that fission product (FP) carbides diffuse much faster through SiC and the carbon layers than their relatively immobile oxide(s) [2, 3, 11, 12]. Therefore, a minimum UC_x could be beneficial by potentially allowing for a sufficiently high μ_{O_2} to oxidize the Ln's while at the same time holding it low enough to avoid significant CO production.

2.1 Burnup simulations

Burnup simulations are fundamental to the approach and, among other influences, depend upon the fuel geometry, composition, and the neutron spectrum over the irradiation period. To illustrate the methodology for determining the initial minimum UC_x content, a prismatic HTGR block with a maximum burnup of 16.1% fissions per initial metal atom (FIMA) was used, the details of which are found in Table 1. In principle, the approach requires iteration using the elemental inventory as a function of burnup together with thermodynamic and mass balance arguments detailed below (assuming the neutron spectrum is fairly constant). However, for this case as shown in Fig. 1, the elemental inventory per fission for the oxide formers depend very weakly on the initial UC_x content over the range of 3.8 to 20 mole % that bound the minimum. Thus, iteration was not deemed necessary. The burnup simulation results for a 3.8 mole % UC_x composition gives a highly accurate approximate result compared to the converged solution from iteration with much less computational overhead.

The Monte Carlo neutron transport and depletion analysis tool Serpent 2 [10] was utilized to generate fission product boundary conditions and inputs for the computational thermodynamic software FactSage® program [8] using the example configuration of a prismatic HTGR block. It is noted that these scoping calculations are not intended to be representative of all possible HTGR designs, but rather to demonstrate this methodology and approach for a reasonably representative configuration. These reactor physics calculations assume a constant temperature and do not account for thermal or thermo-mechanical feedback effects.

The example was taken as a standard Modular High Temperature Gas-cooled Reactor (MHTGR) block geometry with 15.5% enriched UCO fuel [13], similar to the definition of a recently developed benchmark problem [14]. The geometry values for the neutronics and burnup calculations are shown in Table 1. The MHTGR block geometry modeled in the calculations is shown in Figure 2. A periodic boundary condition was used in the calculation with no burnable absorbers. For these scoping calculations, ENDF/B-VII.0 nuclear data was used [15], noting that improvements have been made to both the ¹²C radiative capture cross section and fission yields in the more recent ENDF/B-VII.1 library [16]. The nuclear data is all taken at a temperature of 900 K, with the exception of the thermal neutron scattering data for graphite, which is used at 1000 K. Data for 1341 nuclides was included in the depletion calculations.

Three compositions of UCO were selected for a parametric study spanning the expected parameter space of UO₂ (80%/90%/96.2%) and UC_{1.9} (20%/10%/3.8%) molar content.

An example neutron flux spectrum from the analysis is shown in Figure 3 for Beginning-of-Life (BOL) and End-of-Life (EOL) conditions, considered to be 0% FIMA and 16.1% FIMA fuel burnup in the context of this study.

2.2 Thermodynamic calculations

Due to the abundance of C present in the buffer region that encompasses the fuel kernel in a TRISO-coated particle, the O distribution equations that govern the analysis are based on the Ellingham-Richardson diagram for the carbides (over the temperature range of interest) given in Fig. 4. This figure was generated using the FactSage thermodynamic database [8] and a model for fluorite-structure urania solution, (U,Pu,Ln)O_{2-x}, with dissolved Ln oxides [17-24]. Figure 4 shows that Ln's, excluding Eu, are oxidized first, followed by BaC as BaZrO₃, UC_x phase, SrC as SrZrO₃, EuC₂ and finally ZrC as ZrO₂. The plot also indicates the oxygen-to-metal ratio (O/M) for the urania solution should remain below 1.950 for FIMA of 16.1% to avoid oxidizing SiC (should the inner pyrocarbon layer crack and expose it to the kernel system).

The details of the thermodynamic calculations for the urania solution are given in Table 2. The Ln's and Pu dissolve in UO_{2-x} with a μ_{O_2} below that of Zr up to an O/M of 1.947 resulting in an effective O/M for the Ln's and Pu, or Θ^1 , of 1.55 for oxidation of all Zr. A further Θ increase up to 1.61 would be acceptable to avoid conversion of SiC to SiO₂. This allows for computing bounding compositions for UC_x initial content and anticipated CO pressures as shown in Fig. 5. It should be noted that Θ changes with burnup and the bounds must be computed for each particular case.

2.3 Oxygen distribution equations

To determine the amount of oxygen released and thus the necessary getter material content, in this case UC_x, using the principle of conservation of mass the following equations are operative for full consumption of UC_x:

$$n_O = 2f_{UO_2} + (2 - \Theta)f_{Pu} + f_{Np} - \Theta f_{Ln} - 2f_{Zr} - f(y_{Sr} + y_{Ba}) \quad 1$$

$$n_{UC_x} = n_{UC_x}^i - \frac{n_O}{2} - f_{UC_x} \quad 2$$

$$f = f_{UO_2} + f_{UC_x} \quad 3$$

¹ In reality $\Theta > 1.5$ results from a combination of $O/(Ce+Pu) > 1.5$ and $O/U > 2$. The mathematical simplification presented here is for consistency with [3] T. Tiegs, T. Lindemer, T. Henson, *J. Nucl. Mater.*, 99 (2-3) (1981) 222-34.

$$F = \frac{f}{(n_{UO_2}^i + n_{UC_x}^i)}$$

where n_o is the moles of freed O. The molar fissions per UO_2 and UC_x are f_{UO_2} and f_{UC_x} respectively with f being the total and FIMA represented by F . The y_i are elemental inventory per f (or F) as determined from the codes as a function of f (or F) for the specific kernel composition and burnup. The oxidation state of the Ln's and Pu in the fluorite urania solution is represented by Θ . The initial and running total of UO_2 and/or UC_x in moles are n_j^i and n_j respectively.

The first term in Eqn. 1 represents O release from fissioning of UO_2 (or bred in Pu as an oxide) while the second and third correspond to the freeing of O from the fact that ^{238}U is transmuted to Pu and Np existing as either PuO_Θ or NpO . For simplicity and due to the lack of sufficient thermodynamic data, Np is conservatively taken to only form NpO in this oxygen potential range. The remaining terms represent the binding of O to the Ln's, Sr, and Ba fission products. They are treated as composite totals as a function of f (or F).

The burnup simulations give the result of each fission event corresponding to a U or Pu atom and the y_i are fit using a least squares method to the output to facilitate representing this relation up to 16.1% FIMA. The relevant equations resulting from this approach are summarized in Table 3; note the consistency in that the zero intercept values are essentially identical to the fission-product yield for ^{235}U as they should be.

Combining Eqns. 1 – 4 with the initial moles of U set unity, i.e. $n_{UC_x}^i + n_{UO_2}^i = 1$ such that $f/mole = F$ then solving for n_{UC_x} equal to zero leads to Eqn. 5:

$$x_{UC_x} = F \left(1 + \frac{(2 - \Theta)y_{Pu} + y_{Np} - \Theta y_{Ln} - 2y_{Zr} - (y_{Sr} + y_{Ba})}{2} \right) \quad 5$$

where x_{UC_x} is the minimum mole fraction of UC_x needed in fresh fuel to oxidize all the fission products up to and including Ba as a function of F .

3. Results and discussion

This approach for the present example analysis gives 5.1 to 5.5 mole % UC_x as the minimum required initial content to maintain an acceptably low μ_{O_2} and avoid significant CO production. These values are calculated using $\Theta = 1.55$ and 1.61 respectively. The upper and lower Θ 's correspond to calculated equilibrium oxygen potentials whereby Zr ($\Theta = 1.55$) and SiC ($\Theta = 1.61$) just begin to oxidize. Traditionally, sufficient UC_x has been added to the kernel to avoid above-atmospheric CO pressure and the “amoeba” behavior, thus precluding failure via the predominant mechanisms discussed above.

Evidence, discussed in detail below, suggests that designing a fuel kernel close to the minimum UC_x content range could potentially improve performance of the HTGR TRISO particle with regards to Eu retention. Whether kernels can be produced within appropriate tolerances to exercise this option is still an open question and the subject of on-going research at Oak Ridge National Laboratory.

The behavior of Eu is different from the other lanthanides in that it is present as a carbide instead of an oxide as long as the UO₂-UC_{2-x} equilibrium is present, as is illustrated in Fig. 4. While Eu is not a high yield FP, it has been measured to be one of the highest concentration FPs existing outside of UCO TRISO particles after irradiation testing [25]. A particularly distinctive differentiation between the behavior of the lanthanides Eu and Ce appears in Bullock's [12] measurement of time-dependent release defined as diffusion through the SiC layer. (Note that Horsley et al [26] reported that UO₂ particles irradiated to 5.8% fima were measured to contain CO amounts that they calculated to result in 10.5 MPa CO at 1400°C.) For Eu, the behavior at 1500°C was generally consistent with the thermodynamic predictions in that there was greater release from fuel particles with UC₂ and UC_{0.4}O_{1.6} kernels presumably containing EuC₂, while that from fuel particles with Eu₂O₃ in UO₂ kernels was ~6%. Percentage release of Ce at 1500°C from UC₂ kernels containing CeC₂ was similar to EuC₂ in UC₂ and UC_{0.4}O_{1.6} kernels. However, at the end of burnup, the molar Ce/Eu ratio in the kernel would be ~40, so the molar release of Ce at 1500°C was correspondingly larger.

As mentioned earlier, from a thermodynamic standpoint the resulting UC_{2-x} (from the initial UC_x present) should be consumed before EuC₂ initiates oxidation. This leads to Eqn. 6 which gives an approximation of F_{Eu} , the FIMA at which Eu will no longer exist as a carbide assuming Sr and Ba exist as ternary zirconates.

$$F_{Eu} = \frac{x_{UC_x}}{\left(1 - \frac{\Theta y_{Ln} + 3(y_{Sr} + y_{Ba}) - (2 - \Theta)y_{Pu} - y_{Np}}{2}\right)} \quad 6$$

As calculated, this occurs at 10.9 to 11.7 % FIMA for $\Theta = 1.5$ and 5.13 to 5.50 initial mole % UC_x. The thermodynamic analysis shows that while Pu and Ln dissolution into UO₂ can result in increased oxidation of the fluorite solution, at this burnup a $\Theta > 1.5$ is calculated to have a μ_{O_2} value above that of EuC₂. Therefore, EuC₂ is predicted to begin oxidation with between 554 and 591 micromoles of Eu generated per mole initial U.

The present thermodynamic and nuclear calculations reveal several general characteristics of the simulated irradiation. The ²³⁵U fission naturally dominates initially. This isotope is ~60 % consumed by 16.1 % FIMA. As illustrated in Fig. 6, it contributes to ~30 % of the fissions occurring at 16.1 % FIMA with the balance coming primarily from ²³⁹Pu and ²⁴¹Pu. Also, the integral molar O release with burnup described by Eqn. 1 has two components: (1) the residual Pu and Np having O/M < 2 and (2) fissioning of ²³⁵U, ²³⁹Pu and ²⁴¹Pu and subsequent binding of O from the oxide forming FPs that accompany this event.

Breaking Eqn. 1 into its two components and again setting the initial sum of UO_2 and UC_x to unity, the O release per FIMA from transmutation alone is:

$$y_o^{trans} = [(2 - \Theta)y_{Pu} + y_{Np}] \quad 7$$

where y_i 's are implicit functions of F (see Table 3). This results in a slightly decreasing (see Fig. 1) but substantial source of O release as the irradiation proceeds for low enriched U. Including more precise details of this phenomenon differentiates this work from previous similar analyses.

The second component results from fissioning of the actinide oxides and O uptake by the oxide forming FPs; it is the first term along with the second half of Eqn. 1 and as a function of FIMA it is:

$$y_o^{fission} = \frac{2F_{\text{UO}_2} - \Theta F y_{Ln} - 2F y_{Zr} - F(y_{Sr} + y_{Ba})}{F} \quad 8$$

Note that $y_o^{fission}$ constantly increases with FIMA because there are not enough metallic FPs to bind all the O (see Fig. 1). Thus, UC_x is needed to getter the excess O that is liberated from the sum of these two sources.

Finally, many fissions are from Pu isotopes but from an O release perspective whether or not U is first transmuted to Pu before the event is irrelevant and only affects the O distribution calculations via FP inventories.

4. Conclusions

General oxygen balance relations given by Eqns. 1 – 5 were derived for calculating the minimum amount of UC_x in UCO fuel needed to preclude the failure mechanisms resulting from high O_2 and CO pressures. Input data were obtained from thermodynamics and reactor physics calculations capturing more nuclear and chemical factors compared to past analyses. For example, a CALPHAD model for the urania solution [17-24] with dissolved Pu and Ln's provides a better representation of the μ_{O_2} -O/M relationship. Further, included for the first time were the detailed effects of both burnup and residual concentrations of Pu and Np oxides. The present approach was applied to UCO HTGR fuel enriched to 15.5% FIMA and taken to 16.1% FIMA. The results indicate 5.1 to 5.5 mole % UC_x as the minimum initial content needed to maintain acceptably low CO pressure.

Since Eu has been measured [12] to be one of the FPs with the highest fraction the TRISO particles after irradiation testing, it may be desirable to minimize the UC_x fraction in order to oxidize Eu at the earliest possible burnup when concentrations are as low as possible and thus mitigate Eu transport through the SiC layer. Immobilizing Eu has the

potential to reduce the radionuclide release to an even greater level than is currently observed. This can be viewed as an approach to design optimization and would aid in the production of a superior fuel and as a technical description of fuel behavior. However, the deviations in kernel compositions for a given fabrication process must be taken into account as excessive p_{CO} leads to catastrophic failure.

Past analyses have neglected the effect of transmutation products on determining the required UC_x content. Tiegs et al. [3] give reasonably similar results but the value stated for Θ is too high for low burnup fuel according the thermodynamic calculations presented here. Nevertheless, the fission yields in that work, when used to calculate FP inventory, show qualitative agreement with the present simulation.

Acknowledgements

The authors wish to acknowledge the review, aid, and technical insight of Dr. Fred Montgomery and Dr. Tyler Gerczak at Oak Ridge National Lab. This work was funded by a U.S. Department of Energy Office of Nuclear Energy Advanced Reactor Concept Cooperative Agreement Award led by X-Energy, LLC.

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Table 1. Parameters for the neutronics and burnup calculations [13].

Parameter	Value	Unit
UCO enrichment	15.5	%
TRISO packing fraction	35.0	%
Cylindrical compact radius	0.625	cm
Power density	67.8	watt/gram heavy metal
Large coolant channel radius	0.794	cm
Small coolant channel radius	0.635	cm
Unit cell pitch	1.88	cm
Block flat-to-flat dimension	36.0	cm
Fuel kernel radius	0.02125	cm
Buffer thickness	0.01000	cm
IPyC thickness	0.00400	cm
SiC thickness	0.00350	cm
OPyC thickness	0.00400	cm
UCO density	10.65 - 10.85	g/cm ³
Buffer density	1.0	g/cm ³
PyC density	1.9	g/cm ³
SiC density	3.2	g/cm ³

Table 2. Inputs used to determine the equilibrium oxygen potential for fluorite urania with dissolved fission and transmutation products at 16.1% FIMA as a function of O/M (or Θ). No thermodynamic data for Pm and Eu in solution with UO_2 has been assessed; these were therefore aggregated as Nd to a first approximation.

Element	Total moles at 16.1% FIMA	Mole fraction in (U,Ln) O_{2-x} solution	Comment
U	0.7967	0.8722	Includes remaining un-fissioned U from 90% UO_2 and 10% UC_2 initial molar compositions with all remaining UC_2 oxidized to UO_2
La	0.0097	0.0107	
Nd,Pm,Sm,Eu	0.0364	0.0398	No thermodynamic data for Pm, Sm, and Eu in solution with UO_2 has been assessed. Treated as Nd to a first approximation.
Pr	0.0086	0.0094	
Ce	0.0198	0.0217	
Y	0.0056	0.0061	
Pu	0.0366	0.0401	

Table 3. Comparison of the constant values in [3] and the equations from this work derived from a least squares fit to the elemental inventory per FIMA as a function of FIMA over the range 3% to 16.1% generated for 15.5% enriched 96.2% UO_2 – 3.8% $\text{UC}_{1.9}$ molar composition burnup calculation.

Relation	R^2 value	Tiegs et al.
$y_{Ln} = -0.1929F + 0.5339$	0.989	$y_{Ln} = 0.542$
$y_{Pu} = -1.1663F + 0.4041$	0.989	N / A
$y_{Zr} = -0.2987F + 0.3048$	0.992	$y_{Zr} = 0.3400$
$y_{Sr} = -0.1975F + 0.0955$	0.978	$y_{Sr} = 0.0956$
$y_{Ba} = 0.0637F + 0.0678$	0.951	$y_{Ba} = 0.0677$
$y_{Np} = 0.0898F + 0.0089$	0.969	N / A

Figure 1. Elemental inventory for oxide formers at 20% $UC_{1.9}$ (solid), 10% $UC_{1.9}$ (colored, dashed), and 3.8% $UC_{1.9}$ (dots) and freed O (black, solid) per cumulative fissions as a function of % FIMA. The black dashed lines are computed using the relations in Table 3. The inset is a close up view corresponding to the black box enclosing the Pu inventory from 15 to 16 % FIMA in order to aid the reader in differentiating between the lines that are, for the most part, difficult to distinguish. The open and closed circles are calculated using $\Theta = 1.61$ in Eqns. 7 and 8 showing the O released from fission and transmutation contributions independently to illustrate the importance of including the latter effect in the determination of the total O redistributed with burnup.

Figure 2. MHTGR block modeled in Serpent.

Figure 3. BOL (0% FIMA) and EOL (16.1% FIMA) neutron flux spectrum for an example configuration from the reactor physics analyses (80% UO_2 – 20% $\text{UC}_{1.9}$).

Figure 4 Ellingham-Richardson diagram for the oxide forming FPs in equilibrium with their carbides. Note that the O_2 partial pressure is referenced to one atmosphere and these are the temperatures of interest to HTGR designers. The nominal UC_2 line is calculated either as that compound or U_2C_3 depending on phase stability.

Figure 5 Logarithm of the CO pressures for the oxide forming FPs in equilibrium with their carbides. As in Fig. 4, the nominal UC_2 line is calculated either as that compound or U_2C_3 depending on phase stability.

Figure 6. Instantaneous fraction of fissions during each burnup step for an example configuration from the reactor physics analyses (80% UO_2 – 20% $\text{UC}_{1.9}$).