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

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A B S T R A C T

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 UO_2 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 UO_2 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 for 15.5% enriched UCO taken to 16.1% actinide burnup. Required input data were obtained from CALPHAD (CALculation of PHAse Diagrams) 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.

1. Introduction

It has been established that pressure vessel failure (Kovacs et al., 1985) and CO corrosion (Homan et al., 1977; Tiegs et al., 1981; Minato et al., 1991) as well as tristructural isotropic (TRISO) coating compromise from kernel migration (Homan et al., 1977; Tiegs et al., 1981) 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 (Homan et al., 1977; Tiegs et al., 1981; Lindemer, 2002). 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 (Homan et al., 1977; Tiegs et al., 1981). 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 is included for the first time.

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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 (SGTE; CALPHAD; Bale et al., 2002; Dinsdale, 1991) with the Monte Carlo neutron transport and depletion tool, Serpent 2 (Leppänen et al., 2015) 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_{O2}, 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 (Homan et al., 1977; Bale et al., 2002). It has been conjectured that fission product (FP) carbides diffuse much faster through SiC and the carbon layers than their relatively immobile oxide(s) (Homan et al., 1977; Tiegs et al., 1981; Bullock and Kaae, 1983; Bullock, 1984). Therefore, a minimum UC_x could be beneficial by potentially allowing for a sufficiently high μ_{O2} 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 3.8–20 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 interaction with much less computational overhead.

The Monte Carlo neutron transport and depletion analysis tool Serpent 2 (Leppänen et al., 2015) was utilized to generate fission product boundary conditions and inputs for the computational thermodynamic software FactSage® program (Bale et al., 2002) 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 an example configuration.

Table 1
Parameters for the neutronics and burnup calculations (Pope, 2012).

Parameter	Value	Unit
UCO enrichment	15.5	%
TRISO packing fraction	35.0	%
Cylindrical compact radius	0.625	cm
Power density	67.8	watt/g 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

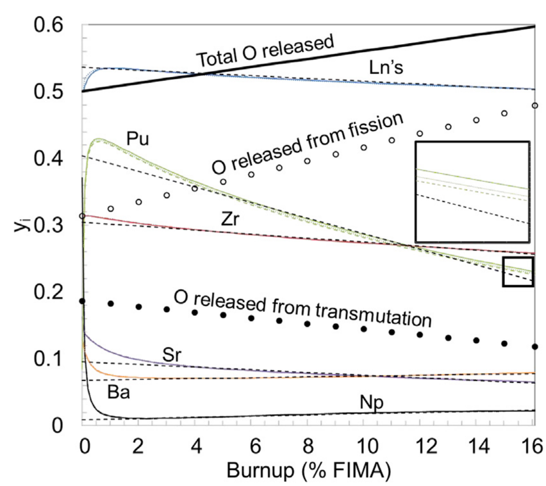

Fig. 1. Elemental inventory for oxide formers at 20% $UC_{1.9}$ (solid), 10% $UC_{1.9}$ (colored, dashed), and 3.8% $UC_{1.9}$ (dots) with released O (black, solid) per cumulative fissions as a function of % FIMA. The black dashed lines are computed using the relations in Table 3. The inset is a close 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 Eqs. (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.

These reactor physics calculations assume a constant temperature and do not account for thermal feedback effects.

The example was taken as a standard Modular High Temperature Gas-cooled Reactor (MHTGR) block geometry with 15.5% enriched UCO fuel (Pope, 2012), similar to the definition of a recently developed benchmark problem (Strydom et al., 2016). 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 Fig. 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 (Chadwick et al., 2006), noting that improvements have been made to both the ^{12}C radiative capture cross section and fission yields in the more recent ENDF/B-VII.1 library (Chadwick et al., 2011). 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.