



Thermochemical assessment of oxygen gettering by SiC or ZrC in PuO_{2-x} TRISO fuel

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ABSTRACT

Particulate nuclear fuel in a modular helium-cooled reactor is being considered for the consumption of excess plutonium and related transuranics. In this work a thermochemical analysis was performed to predict oxygen potential behavior in plutonia TRISO fuel to burnups of 88% FIMA of the Pu²³⁹ content with and without the presence of oxygen gettering SiC and ZrC. The gettering phases are designed to prevent kernel migration, a serious issue in TRISO fuel, and this has been demonstrated with both SiC and ZrC. The phases reduce CO pressure, thus also reducing the peak pressure within the particles by at least 50%, decreasing the likelihood of pressure-induced particle failure. A model for kernel migration based on vapor transport by CO was used to semi-quantitatively assess the effect of controlling oxygen potential with SiC or ZrC and demonstrate the potential dramatic effect of the addition of these phases on carbon transport.

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1. Introduction

The issue of the disposition of spent nuclear fuel has been exacerbated by the recent decision to curtail development of the Yucca Mountain nuclear waste repository. A result of that decision may be a greater emphasis on reprocessing spent nuclear fuel to separate transuranics and fission products. The long-lived transuranics, including the components that are subject to proliferation concerns such as ²³⁹Pu, could then be efficiently consumed in a reactor designed to attain high burnups. The remaining fission products and other components that are shorter-lived would be more easily disposed of through geological or other means having a significantly shorter period in which radiotoxicity is a problem.

It has been proposed that transuranics resulting from reprocessing of light water reactor (LWR) fuel might be profitably consumed in a high temperature gas-cooled reactor, specifically a modular helium reactor (MHR) [1,2]. Specific MHR fuel cycle schemes have been developed using a “Deep Burn” concept where a burnup of 60% fission of initial metal atoms (FIMA) or more can be achieved in a single irradiation and which is now the subject of a US Department of Energy program termed Deep Burn [2]. The fuel form is a TRISO coated particle with an oxide fuel kernel and typical sequence of buffer or low density carbon layer followed by a high density, isotropic pyrolytic carbon layer, a SiC layer and final outer high density, isotropic pyrolytic carbon layer [2]. The fuel compositions are envisioned as largely plutonium with minor neptunium and possibly americium contents, and with target particles that are

not fissile, but contain americium and curium for transmutation to stable or shorter-lived isotopes.

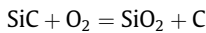
A significant issue for high burnup fuel that can limit life is the increasing pressure within the particle due to production of noble fission gases and CO resulting from oxygen released due to fissioning reacting with carbon in the particle. The total gas pressure has the potential to exceed the yield stress and result in failure. A more insidious problem that is generally observed at high burnup is kernel migration. The fuel kernel with time appears to migrate up the temperature gradient through the buffer layer and eventually the inner pyrolytic layer, and then can damage the SiC layer. This is seen in fuel with oxide kernels whether containing thorium, uranium, or plutonium [3]. The mechanism of apparent kernel movement is carbon transport from the hotter side to the cooler side of the particle. A number of analyses have been performed to understand the phenomenon. Lindemer and Pearson [3] assessed a substantial body of data and mechanisms for a variety of fuels and determined that transport was controlled by solid state diffusion across the oxide fuel kernel, with carbon possibly diffusing through the oxide [4]. Others have concurred that solid state diffusion is a key mechanism in transport, suggesting that the multi-step process is a combination of solid state oxygen transport through the kernel and gas-phase transport of carbon via CO [5–7]. One of the objectives of this work is to contribute to the thermochemical basis for understanding how CO/CO₂ pressure behaves with burnup and to quantify components of a possible mechanism of kernel migration.

The means for controlling kernel migration is to either minimize the thermal gradient or to reduce the oxygen potential (oxygen potential is defined as $RT \ln p(O_2)$ where R is the ideal gas constant, T is absolute temperature, and $p(O_2)$ is a dimensionless

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value defined as the equilibrium oxygen pressure divided by the standard-state oxygen pressure, 101 kPa at 298 K) so that the driving force for solid state transport is substantially reduced and/or the CO pressure is below a practical level for transporting carbon. This has been successfully accomplished by either including an actinide carbide phase in the kernel with a composition euphemistically termed UCO but in reality is a two phase $\text{UO}_{2-x}\text{-UC}_{2-y}$ kernel [3]. Thus the equilibrium between the carbide and oxide fixes or “buffers” the oxygen potential to a low value mitigating kernel migration. Another successfully demonstrated approach is to include SiC as an oxygen getter to buffer the oxygen potential in either the oxide kernel or the low density carbon layer [8]. The oxygen potential is thus maintained at low values by the relationship



Zirconium carbide has since the 1970s been considered a replacement for the SiC layer to achieve higher operating temperatures and for its potentially greater resistance to fission product attack, most notably by palladium [9–10]. The phase can also buffer the oxygen potential to low values and thus control gas pressure and kernel migration, which has been demonstrated by Bullock and Kae [11]. The free energy of ZrO_2 is significantly more negative than that of SiO_2 , and thus will result in even lower oxygen potentials than does SiC. An objective of the thermochemical analysis reported here is to at least semi-quantitatively define the dramatic effect of SiC and ZrC in stabilizing a Deep Burn plutonia fuel kernel in a TRISO configuration to guide development of the fuel form.

2. Computational basis

The approach is to perform thermodynamic equilibrium calculations for the proposed fuel to determine conditions within the particle to high burnups. The MHR fuel considered is a PuO_{2-x} -containing TRISO fuel particle embedded in carbon fuel compacts that are set in prismatic graphite fuel elements. Table 1 provides the basic characteristics of the fuel form. Burnup levels to 88% FIMA, that is consumption of 88 at.% of Pu^{239} , were considered. To avoid difficulties in fuel processing caused by phase transformations at low O/Pu, it is expected that the initial fuel stoichiometry is likely to be ~ 1.7 to retain the fluorite structure of the oxide. That is also the value used in the fuel cycle analysis by Talamo [12] and is therefore assumed for the initial value for the O/Pu of the fuel for the current calculations. Preparation of such a fuel containing ZrC, however, can be problematic as the equilibrium with the zirconium oxide and carbide would drive the O/Pu ratio to ~ 1.6 .

Elemental compositions within the particles undergoing burnup were determined from a set of ORIGEN calculations that provided bred transuranic and fission product concentrations. A curve of %FIMA for Pu^{239} as a function of operational days is given

Table 1
Key characteristics of the reference TRISO fuel used in the analysis [2].

Kernel diameter (μm)	200
Kernel density (g/cm^3)	10.0
Buffer layer thickness (μm)	120
Buffer layer density (g/cm^3)	1.05
Initial actinide composition (at.%)	
^{238}Pu	3%
^{239}Pu	52%
^{240}Pu	24%
^{241}Pu	9%
^{242}Pu	5%
^{237}Np	7%

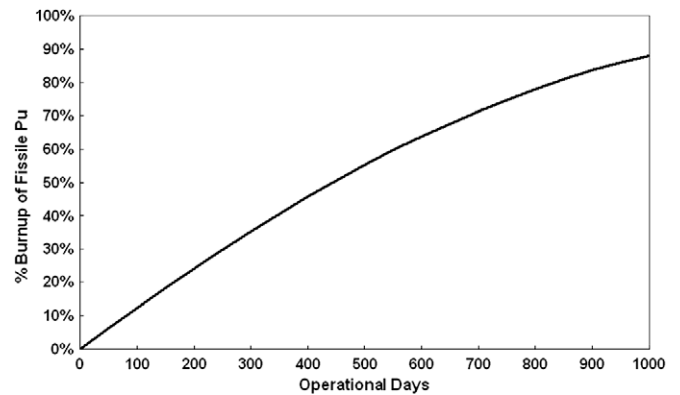


Fig. 1. ORIGEN calculation of burnup of fissile plutonium (Pu^{239}) as a function of operational days for the fuel described in Table 1.

in Fig. 1. Phases expected to be present are those typical for high burnup fuels, although some disagreement exists regarding the formation of some phases, most notably those containing cesium and/or molybdenum [13–18].

The FactSage thermochemical software package [19] was used to perform the thermodynamic equilibrium calculations. The thermochemical data are contained in the 1996 Scientific Group Thermodynamic Europe (SGTE) Pure Substance Database [20] that accompanies FactSage. The exceptions to the use of the SGTE database included the thermochemical model and values for the Pu–O system which were taken from [21] under the FUELBASE program of the Commissariat à l’Énergie Atomique. Subregular solution models for the transition metal phases that contain the five fission product metals molybdenum, technetium, ruthenium, rhodium, and palladium were used based on the work of Kaye et al. [22]. Simplifying assumptions included that initial neptunium and all bred transuranics are inherent in the PuO_{2-x} phase and similarly that all the lanthanides were summed and represented by La_2O_3 . All other metal and oxide phases present within the fuel particle were determined from the existing SGTE database used with the FactSage calculations.

3. Results

The thermochemical models and values of Gueneau et al. [21] were used to compute the partial Pu–O phase diagram shown in Fig. 2. Note that at lower temperatures and O/Pu values the PuO_{2-x}

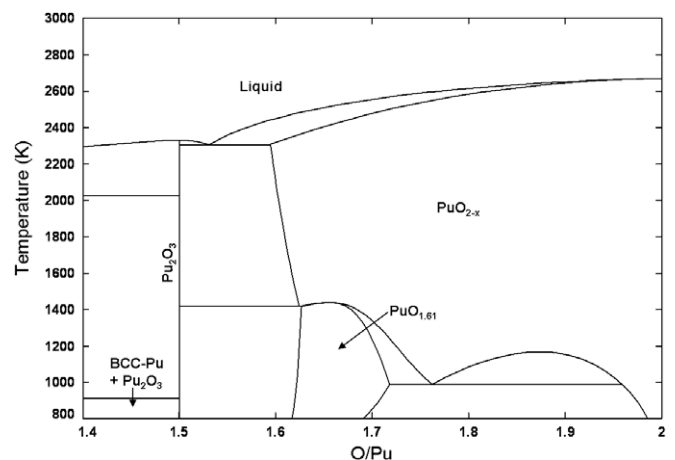


Fig. 2. Computed phase diagram for the Pu–O system utilizing the data and models of Gueneau et al. [21].

phase will transform to the $\text{PuO}_{1.61}$ phase and that below ~ 1160 K a miscibility gap exists for PuO_{2-x} .

Equilibrium calculations were used with the models for the plutonia system to determine oxygen potential as a function of temperature. In Fig. 3 are plotted the log of the oxygen pressure versus reciprocal absolute temperature for a set of plutonia stoichiometries which are straight lines for two-phase equilibria, and near straight lines for single phase systems. As would be expected, the oxygen pressure decreases with decreasing O/Pu. Also plotted are the equilibria between SiC and ZrC and their respective condensed phase oxides. These lie at low oxygen pressures, with the SiC–SiO₂ line almost co-linear with that for $\text{PuO}_{1.70}$, and that for ZrC–ZrO₂ substantially lower aligning approximately with $\text{PuO}_{1.60}$.

Equilibrium calculations utilizing the results from ORIGEN calculations for compositions out to 88% FIMA for Pu^{239} , equivalent to 1000 operational days, were performed to determine the effect of burnup on the oxygen-to-metal ratio (O/M, where M is to the total transuranic content including bred isotopes) of the fluorite-structure fuel phase. Fig. 4 illustrates the effect of temperature and burnup, as represented by operational days, on O/M with and without SiC or ZrC gettering. In the absence of gettering phases the O/M values increase with burnup, consistent with Lindemer and Pearson [3] and Lindemer [16] who tabulated the oxygen fraction which combines with fission products to form the oxides. In this case, the 1.7 original oxygen atoms per actinide in the fuel are released from a fission event. A minor difference with Lindemer and Pearson [3] and Lindemer [16] lies in the assumption here that a portion of the fission product cesium will form an oxide phase as indicated by the thermochemical equilibrium calculations whereas they show it as remaining uncombined. At higher temperatures there is an ultimate decrease in O/M which is steeper at higher burnup, driven by the equilibrium with carbon and the generation of CO.

The presence of SiC or ZrC in the fuel kernel or available perhaps in a separate layer in communication with the kernel will equilibrate with the oxygen in the fuel and fix the oxygen potential. In order for sufficient SiC or ZrC to be available to react with all the released oxygen not taken up by fission products and thus maintain a low oxygen potential at 88% FIMA the kernel requires a concentration of ~ 32 mol% of the carbides. From Fig. 4 it is apparent that the O/M ratio in the fluorite-structure fuel phase varies only slightly for the gettered systems, remaining at very low values. Below ~ 1450 K the fluorite-structure phase is actually not in equilibrium with SiC or ZrC, and instead is present with, respectively, $\text{PuO}_{1.61}$ or Pu_2O_3 .

The O/M in the fuel and the resultant oxygen potential yield a significant CO pressure which can be an equal or greater contributor to total pressure in the particle than the noble fission gases. The

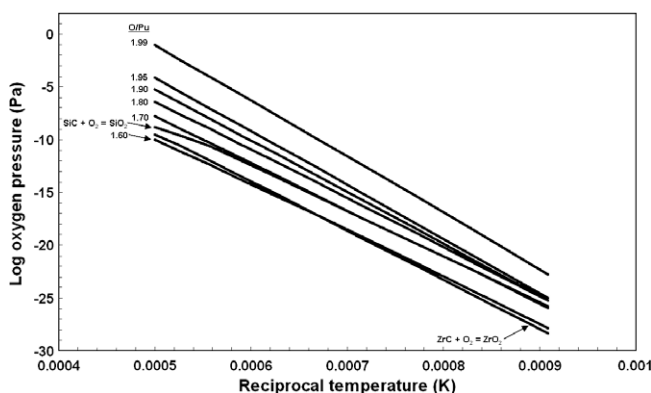


Fig. 3. Plot of the log of oxygen pressure versus reciprocal temperature for plutonia at various O/Pu ratios and for the oxide-carbide equilibria for silicon and zirconium.

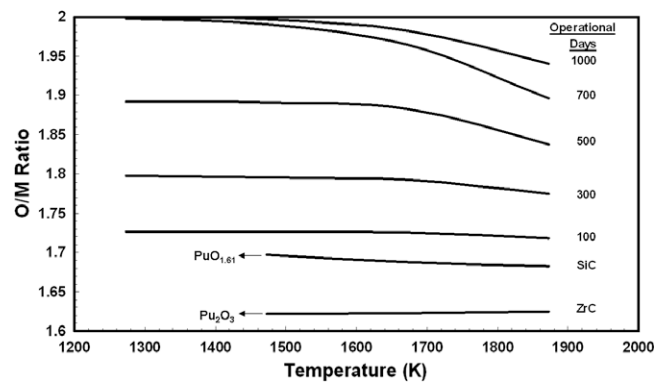


Fig. 4. Plot of O/M versus temperature in the fuel fluorite phase without a getter for various burnups to 1000 days and the constant values in the presence of SiC or ZrC getter. Note that at lower temperatures in the presence of a getter phase the lower oxides can be stable.

CO pressure at equilibrium was therefore calculated as a function of temperature for a series of burnups (operational days) and can be seen in Fig. 5.

The effect of burnup and gettering phases on the CO, CO₂, and total pressures are plotted in Fig. 6 as a function of burnup at 1673 K, near the likely peak operating temperature. The pressure is computed assuming that all noble fission gases are fully released from the kernel and that the available void space is solely that within the buffer layer, equivalent to 0.0212 mm³, or a free/kernel volume ratio of 5. The total pressure includes the noble gases and contributions from CO and CO₂, with a difference of almost a factor of two at high burnup between the systems with and without a carbide gettering phase. The resultant difference in the CO pressure between ungettered and gettered systems at high burnups (1000 operational days or 88% FIMA of Pu^{239}) is over 2.5 orders of magnitude, with the CO₂ pressure difference being ~ 3.5 orders of magnitude lower for the gettered system.

4. Discussion

The oxygen pressure as a function of O/Pu in Fig. 3 and the resultant CO pressures indicated in Figs. 5 and 6 differ to an extent from the results from the earlier model of Besmann and Lindemer [23]. The calculations at high O/Pu yield CO pressures are approximately an order of magnitude lower than those of Besmann and Lindemer [23] and Lindemer [16], although they more closely agree at lower O/Pu values. This is a result of differences in computed oxygen potentials from the differing models of PuO_{2-x} used by Besmann and Lindemer [23] and in this work which adopted

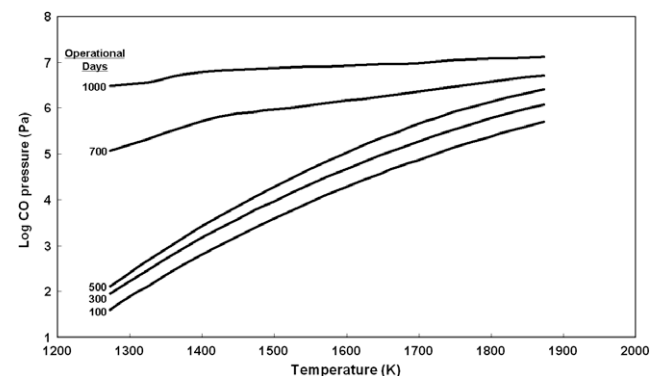


Fig. 5. Plot of CO pressures in the ungettered fuel as a function of temperature for a series of operational days up to 1000 (88% FIMA of Pu^{239}).

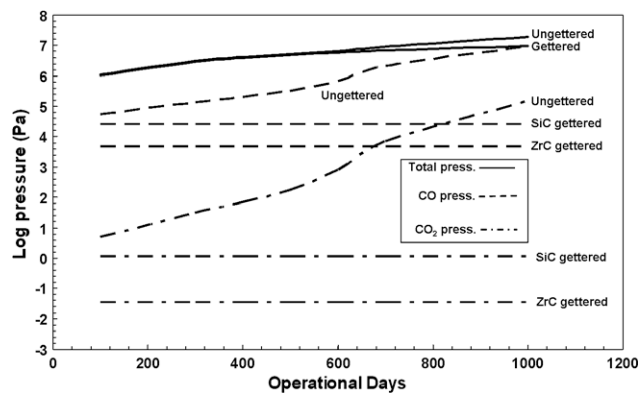


Fig. 6. Plot of the log of the total, CO and CO₂ pressure at 1673 K versus operational days for the ungettered and gettered fuel system.

that of Gueneau et al. [21]. There is no clear distinction as to which model yields overall more accurate oxygen potential behavior, however that of Gueneau et al. [21] is a more appropriate physical representation of the system marking an advance in representing these complex oxides.

As pointed out by Barrachin et al. [24] assuming simple global equilibrium fails to include potentially important effects of kinetics such as fission product migration to oxide fuel grain boundaries and even out of the kernel. Their discussion of the issues, and results from their calculations that include some kinetic effects for UO_{2+x} kernel TRISO particles differ to an extent from the conclusions of this work. Cesium plutonates are not considered here given the absence of any information on such phases yet the uranates are central to their analysis. In contrast to Barrachin et al. [24] Cs₂MoO₄ is not stable under conditions of extensive plutonia burnup, whereas Cs₂ZrO₃ is calculated to be present, possibly due to the lack of competing plutonate phases. Regardless, given that the oxygen balance, that is the O/M in the fluorite fuel phase that results from distribution of fission-liberated oxygen, is not directly affected by these differences they will not impact the conclusions of the analysis. Minato et al. [17], however, in their analysis of urania particulate fuel consider Cs_nC as an intercalated phase given the diffuse appearance of cesium in the buffer and inner pyrolytic carbon layers. Should significant cesium be present as a Cs_nC phase it would alter the oxygen distribution between the fluorite fuel phase and oxide fission products, although the general conclusions of this analysis would not be significantly changed.

Given sufficient burnup the O/M of ungettered fuel can approach 2 (Fig. 4), the maximum value as PuO₂ is the highest oxide of plutonium. The presence of gettering phases such as SiC or ZrC have a substantial impact on oxygen pressure/potential in the Deep Burn fuel as seen in Fig. 3. Both carbides fix oxygen pressures at equivalent O/Pu values near 1.7 or 1.6, and can even shift the equilibria to lower oxygen phases such as PuO_{1.61} or Pu₂O₃.

Experimental observations of the effect of initial O/Pu and thus oxygen potential on kernel migration have been striking. Substantial kernel migration is observed for particles with initial O/Pu of 1.81 and 1.84 in the FTE-13 experiment irradiated for 450 days

to 71% FIMA in the Peach Bottom reactor [25]. Lindemer and Pearson [3] note that because of the relatively high initial O/Pu during burnup the stoichiometry will relatively quickly reach an O/M of 2 and thus a high oxygen potential promoting kernel migration. Within this same irradiation, kernels with an initial O/Pu of 1.69 did not exhibit observable kernel migration, although the O/M should have reached a value of 2 during the irradiation. An explanation for the lack of kernel migration for this experiment remains elusive.

Wagner-Loeffler [7] in their analysis concluded that there is less certainty that solid state transport is the sole controlling mechanism and that vapor-phase transport may play a role. That is also noted by Gulol et al. [5] and Choi and Lee [6] who propose combined solid state and vapor transport mechanisms. This was briefly explored here by employing the analytical model for carbon transport in TRISO fuel developed by Choi and Lee [6] of CO diffusion through the buffer layer. They used a cylindrical geometry for ease of calculation converting kernel and buffer layer dimensions from particle radii to cylindrical lengths. The model treats the system as rate-limited by CO diffusion through the buffer layer down the concentration gradient. The values in Table 1 were used to obtain a function, α , defined by Choi and Lee [6] as

$$\alpha = \frac{\pi m_{\text{CO}} \omega^4 d^2 (1 - a^2)}{160 \eta (1 - \omega)^2} \left[\frac{1 - a^4}{1 - a^2} + \frac{1 - a^2}{\ln a} \right] \quad (1)$$

where m_{CO} is the molecular weight of CO, ω is the density of the buffer layer relative to graphite, $a = L_k/L_b$ with L_k and L_b the cylindrical representations of the kernel and buffer layer radii, respectively, d is the nominal pore size in the buffer layer (5 nm), and η is the viscosity of CO gas. The term for α is used in the expression for the kernel displacement rate x ($\mu\text{m/s}$) such that

$$x = \frac{\alpha m_c \bar{P} \Delta P}{\pi m_{\text{CO}} a^2 L_k \bar{T} \rho_c} \quad (2)$$

where m_c is the molecular weight of carbon, \bar{P} is the average CO pressure in the kernel (sum of the hot and cold side pressure divided by 2), ΔP is the difference in CO pressure between the cold and hot side of the kernel, \bar{T} is the average particle absolute temperature, and ρ_c is the density of the buffer layer carbon.

Migration data and thermal gradient information for the FTE-13 experiment, which would be closest to the Deep Burn configuration is not readily available. Thus to obtain an estimate of kernel migration based on the model of Choi and Lee [6] the measured kernel displacement for UO₂ particles at average temperatures of 1457 K and 1600 K and 763 days in-reactor from Table 2 of Wagner-Loeffler's paper were used. Although the dimensions of the particle were larger, being typical UO₂ TRISO particles, the exposure time was representatively long and the temperatures near those expected maximum for the Deep Burn particles. From Wagner-Loeffler [7] the temperature gradient is reported as 11 K/mm, and thus the gradient across a 200 μm diameter kernel would be 2.2 K.

The results of predicted displacement for the ungettered particle at 1600 K and 1457 K are 10.71 μm and 3.989 μm , respectively, after 700 days (Table 2). These are reasonably close to the

Table 2
Computed equilibrium CO pressures at 1600 K and 1457 K average temperatures and at the hot and cold sides of the 2.2 K gradient across the kernel (the gradient was identical for both temperatures from Wagner-Loeffler [7], with resultant displacement rate, x , and the displacement distance after 700 days for a Deep Burn TRISO particle.

	Average temperature (K)	\bar{P} (MPa)	P_{cold} (MPa)	P_{hot} (MPa)	x ($\mu\text{m/s}$)	Displacement after 700 days (μm)
Ungettered	1600	1.448	1.441	1.4558	1.772×10^{-7}	10.71
Gettered with SiC	1600	0.009714	0.009566	0.009863	2.389×10^{-11}	1.440×10^{-03}
Gettered with ZrC	1600	0.001544	0.001517	0.001570	6.645×10^{-13}	4.019×10^{-05}
Ungettered	1457	0.7540	0.7492	0.7588	6.597×10^{-8}	3.989

experimental average values in Wagner-Loeffler [7] of 30.7 and 13.7 μm , respectively, given the differences in the particles and the broad assumptions of the Choi and Lee [6] model. They are also in relative agreement as the experimental values differ from the computed values by a factor of 2.2 and 2.7 at 1457 and 1600 K, respectively. Table 2 also indicates that the effect of gettering on the calculated displacements is dramatic, with SiC gettering decreasing the displacement by ~ 4 orders of magnitude and ZrC over 5 orders of magnitude.

5. Conclusions

The thermochemical evolution of a Deep Burn TRISO fuel particle designed for consuming plutonia and other transuranics was represented using a compound energy formalism model for PuO_{2-x} , a subregular solution model for the five-metal fission product phase, and thermodynamic database values for other fission product phases. These were used in thermochemical calculations to obtain the equilibrium states and gas species pressures as the fuel underwent burnup. As would be expected, the results show that O/M , and therefore oxygen pressure/potential increase with burnup due to released oxygen that is not fully taken up forming fission product oxides. The presence of gettering phases such as SiC or ZrC greatly reduces the O/M and oxygen pressure. The resultant total pressure in the particle at high burnup is also substantially reduced as the CO contribution to the pressure becomes negligible.

It has been experimentally demonstrated that kernel migration is mitigated in TRISO fuel with SiC or ZrC getter phases, possibly through a reduction in the driving force for oxygen transport from reduced oxygen potentials. The computed effect of the gettering phases on CO pressures is also dramatic, being reduced by several orders of magnitude and the O/M of the fuel restricted to low values despite extensive burnup.

The rate of displacement of a fuel kernel was computed using a model dependent on CO gas species transport through the buffer layer. While the preponderance of evidence indicates that kernel migration is rate-limited by solid state transport, the potential CO gas-phase transport of carbon was considered and is significant in ungettered particles and is restricted by low oxygen potentials in gettered systems. A fundamental conclusion is that regardless of the rate-limiting processes, kernel migration is not an issue in gettered particulate oxide fuel systems.

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