ELSEVIER

Journal of Nuclear Materials 270 (1999) 187–193

journal of nuclear materials

Section 3. Nuclear materials chemistry

Thermodynamics of urania volatization in steam

D.R. Olander *

Department of Nuclear Engineering, University of California, Berkeley, CA 94720-1730, USA

Received 23 February 1998; accepted 22 July 1998

Abstract

The thermochemistry of urania volatilization as UO_3 obtained from several literature sources was reviewed. The predicted equilibrium pressures of UO_3 over pure UO_{2+x} in a gas of known oxygen potential and temperature fall into one of two groups that differ by one order of magnitude at 2000 K and two orders of magnitude at 1500 K. The recent theoretical and experimental work from the Lawrence Livermore National Laboratory is judged to be the most comprehensive and reliable. This work supports the high-UO_3 pressure estimates and gives values about 30% lower than those obtained from the earlier work of Ackermann and Chang. The effects of other factors on urania volatility were investigated: H_2 in steam produces a large reduction in the UO_3 pressure; the other volatile uranium species, $UO_2(OH)_2$, is important only at low temperatures and high steam pressures; fuel burnup has a small effect on the UO_2 pressure. As part of the UO_3 assessment, the oxygen potential models for UO_{2+x} of Blackburn and Lindemer/Besmann were compared. © 1999 Elsevier Science B.V. All rights reserved.

1. Introduction

Fuel volatilization is important in severe accident analyses for several reasons. First, as the oxide matrix evaporates, the fission products formerly associated with the volatilized portion are released. Once freed from fuel, these fission products can either evaporate along with the fuel (the 'matrix-stripping' process) or collect on the receding fuel surface. Which of these occurs depends on the volatility of the fission product relative to the fuel. Second, the evaporated fuel vapor may condense and form aerosol particles as it is swept to cooler zones. Alexander and Ogden have observed this phenomenon in UO_2 vaporization in H_2 and air [1]. Third, the vaporized urania may condense on cooler fuel or structures above the elevation from which it originated; volatilization provides a mechanism for upward fuel relocation that acts in the opposite sense as the more common downward relocation of liquefied fuel.

The kinetics of fuel vaporization depend on the partial pressures of the volatile uranium-bearing species at the fuel surface exposed to steam and steam–hydrogen mixtures. There is general agreement that the principal vapor species is $UO_3(g)$. The oxyhydroxide $UO_2(OH)_2(g)$ is also volatile, although its thermochemistry is less well established than that of $UO_3(g)$. In laboratory experiments in pure steam or steam-inert gas mixtures, volatilization is observed at temperatures as low as 1500 K and increases rapidly with increasing temperature [2,3]. The dependence of the rate of evaporation on steam pressure has not been systematically studied. In fact, with the exception of Ref. [1], the fuel evaporation experiments are normally a byproduct of fuel oxidation or fission-product release investigations. As a result, volatilization has been treated mainly as an experimental artifact to be considered in evaluating the principal oxidation or release data.

The rudimentary understanding of the kinetics of fuel volatilization in steam has not deterred modelers from incorporating this process in severe accident codes [4–6]. Ref. [4] assumes that the rate is given by the Hertz–Langmuir formula for evaporation into a vacuum, which produces a gross overestimate of the kinetics when applied in atmospheric-pressure gases. In the latter environment, conventional boundary-layer mass transfer is a more appropriate description, and this is the

---

* Tel.: +1-510 642 7055; fax: +1-510 643 9685; e-mail: fuel@socrates.berkeley.edu.

PII: S 0 0 2 2 - 3 1 1 5 (9 8) 0 0 7 5 8 - 2

approach adopted in Refs. [5,6]. The observation of Alexander and Ogden [1] that the rate of vaporization of UO_2 in 1 atm air at 1800 K is dependent on the gas flow rate supports (but does not prove) this notion. However, based on comparison of observed urania volatilization rates in air with estimates of the mass-transfer-limited rates, Cox et al. have suggested that surface chemical reaction rather than gas-phase mass transfer is the rate-controlling process [7]. Whether similar surface-reaction limitations influence UO_2 volatilization in steam is not known.

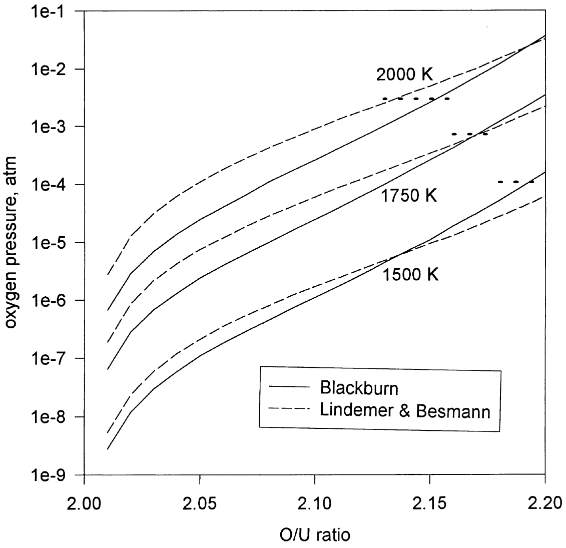

Fig. 1. Oxygen pressures in equilibrium with UO_{2+x}.

2. O_2 and $UO_3(g)$ equilibrium pressures for urania

Whatever the mechanism of the volatilization process, the kinetics depend on the equilibrium pressure of the volatile uranium-containing species in the gas contacting the solid surface. Since $UO_3(g)$ is produced by oxidizing $UO_{2+x}(s)$, its partial pressure depends on the prevailing O_2 pressure. If consideration of the volatilization reaction is restricted to solid oxide that is in oxygen equilibrium with the gas, the oxygen potentials ($RT \ln p_{O_2}$) of the gas and the oxide are equal.

Refs. [5,6] use oxygen potential models of UO_{2+x} to relate p_{O_2} to fuel stoichiometry and temperature. In this approach, the UO_3 pressure calculation is directly dependent on the accuracy of the model used to calculate p_{O_2}. Ref. [6] utilizes the Blackburn formula [8]

$$\ln p_{O_2} = 2\ \ln\left[\frac{x(2+x)}{1-x}\right] + 108x^2 - 32\ \frac{700}{T} + 9.92 \qquad (1)$$

and Ref. [5] utilizes the formula of Lindemer and Besmann [9]: $p_{O_2} = \min(p_1, p_2)$, where

$$\ln p_1 = 2\ \ln\left[\frac{x(1-2x)^2}{(1-3x)^3}\right] - \frac{37\,621}{T} + 15.15, \qquad (2a)$$

$$\ln p_2 = 4\ \ln\left[\frac{2x(1-2x)}{(1-4x)^2}\right] - \frac{43\,298}{T} + 25.74. \qquad (2b)$$

Fig. 1 compares the Blackburn and Lindemer/Besmann oxygen potential models. The agreement is fair at the lowest temperature but deviations approaching one order of magnitude appear at temperatures > 2000 K and O/U ratios < 2.10. Since the UO_3 pressure varies approximately as $\sqrt{p_{O_2}}$, the choice of the Lindemer/Besmann oxygen potential model produces, at least from this source, a factor of three difference in p_{UO_3} compared to the value obtained using the Blackburn oxygen potential model.

The horizontal dashed lines in Fig. 1 represent the oxygen pressures generated by the reaction $H_2 + 1/2O_2 = H_2O$ in pure steam. For a steam pressure p_{H_2O}, the oxygen pressure is [10]

$$p_{O_2} = (p_{H_2O}/2K_W)^{2/3}, \qquad (3)$$

where K_W is the equilibrium constant

$$\ln K_W = 30\,000/T - 6.95. \qquad (4)$$

The stoichiometry of the oxide is determined by equating the oxygen pressure of the gas (Eq. (3)) to that of the solid (Eq. (1) or Eqs. (2a) and (2b)). Fig. 1 shows that in pure steam at 1 atm, the difference between the Blackburn and Lindemer/Besmann stoichiometry predictions is +0.02 O/U units at 1500 K and −0.02 units at 2000 K. The discrepancy is larger in less oxidizing gases (e.g., H_2O/H_2 mixtures) than in pure steam, with the Blackburn model yielding higher oxide stoichiometries than the Lindemer/Besmann model at a specified oxygen pressure.

The other feature of the solid-state thermochemistry of UO_{2+x} is the activity of the uranium-containing component. Following Blackburn [8], UO_{2+x} is represented as a binary solution of stoichiometric UO_2 and neutral atomic oxygen. The activity of the UO_2 component is determined from the oxygen activity (which is proportional to $\sqrt{p_{O_2}}$) by the Gibbs–Duhem equation. In its integrated form this gives

$$\ln a_{UO_2} = -\frac{1}{2}\int_0^x x' \frac{d\ \ln p_{O_2}(x', T)}{dx'}\ dx'. \qquad (5)$$

Using Eq. (1) for the oxygen pressure, Eq. (5) yields

$$\ln a_{UO_2} = -x + 2\ \ln(1 + x/2) + \ln(1-x) - 36x^3. \qquad (6)$$

Ozrin et al. [5] integrated Eq. (5) using Lindemer and Besmann's formula for the oxygen pressure (Eqs. (2a) and (2b)). For O/U > 2.01, their result is closely approximated by

$$\ln a_{UO_2} = 0.99\frac{1-3x}{1-2x}. \tag{7}$$

The plots of Eqs. (6) and (7) in Fig. 2 show three notable features. First, the UO_2 activities from the two oxygen potential models are independent of temperature. This is a consequence of the separated functions of x and T on the right-hand sides of Eqs. (1), (2a) and (2b). Second, the predictions of the two models are in good agreement over the entire O/U range; the maximum deviation is less than 10%. Third, the activity of UO_2 is not greatly different from unity.

The equilibrium that forms UO_3(g) from UO_{2+x}(s) is that of the reaction

$$UO_2(\text{in } UO_{2+x}) + 1/2\,O_2 = UO_3(g) \tag{8}$$

for which the law of mass action yields

$$p_{UO_3} = K_3 a_{UO_2}\sqrt{p_{O_2}}, \tag{9}$$

where K_3 is the equilibrium constant for the formation of UO_3(g) from stoichiometric UO_2(s). It can be expressed in terms of the standard free energies of formation of UO_3(g) and UO_2(s)

$$K_3 = \exp\left[-\frac{\Delta G_f^0(UO_3(g)) - \Delta G_f^0(UO_2(s))}{RT}\right]. \tag{10}$$

Fig. 3 shows UO_3 pressures calculated from four literature sources. The solid curves are Blackburn's original model predictions calculated using Eqs. (9) and (10) with the free energy of formation of UO_3(g) taken from Ref. [11].

The lowest curves at each temperature in Fig. 3 are from the calculation of Ozrin et al. [5]. They also use Eq. (9) but with p_{O_2} and a_{UO_2} taken from Lindemer and Besmann's model (Eqs. (2a), (2b) and (7)). The free energies of formation of UO_3(g) and UO_2(s) for computing K_3 by Eq. (10) were obtained from the table of standard free energies compiled for the VICTORIA code. In Ref. [4], the origin of these data is cited as an inaccessible AEA (UK) document. The curves labeled 'AECL' in Fig. 3 are based on the formulation presented in Ref. [6], which is equivalent to Eqs. (9) and (10). This can be shown by integrating Eq. (5) by parts. In Ref. [6], p_{O_2} and a_{UO_2} were determined from Blackburn's equations (Eqs. (1) and (6)) and the free energy of formation of UO_3(g) was obtained from the work of Ackermann and Chang [12].

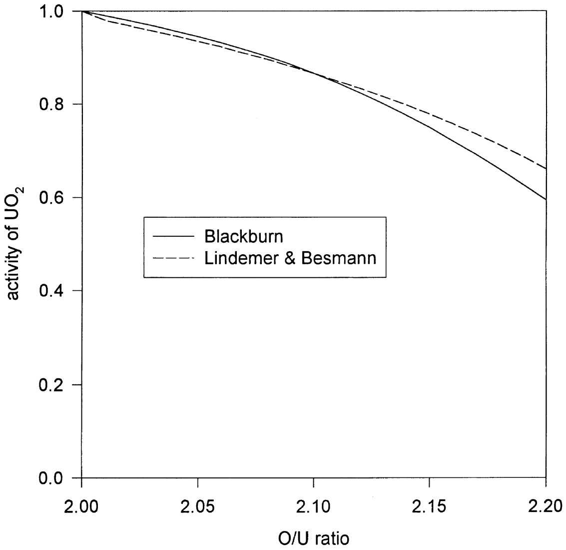

Fig. 2. Activity of UO_2 in UO_{2+x} for 1500 K < T < 2000 K.

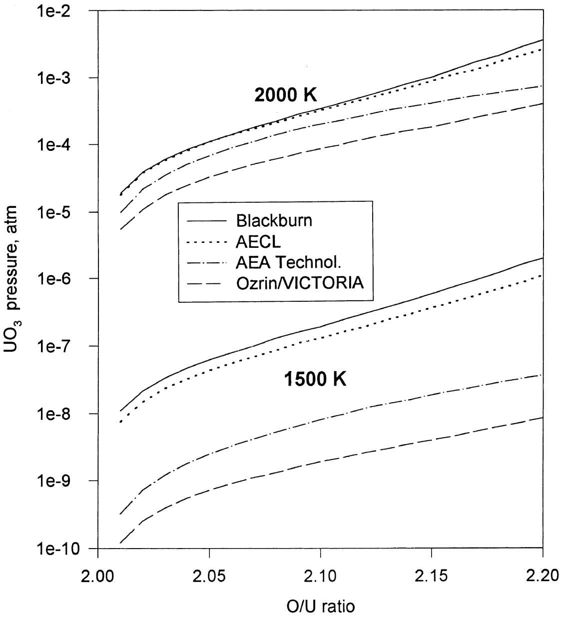

Fig. 3. UO_3 pressures from various literature sources.

The curves labeled 'AEA Technol.' in Fig. 3 are based on the formula provided in Ref. [13]:

$$p_{UO_3} = x\,\exp\left[-\frac{56\,453 + 5754(1-x)^3}{T} + 24.10\right]. \tag{11}$$

The discrepancies among the curves in Fig. 3 are two orders of magnitude at 1500 K and one order of magnitude at 2000 K. These differences are due in part to the use of different oxygen potential models for p_{O_2} in Eq. (9). The UO_3 pressures calculated utilizing Blackburn's p_{O_2} model (Blackburn and AECL in Fig. 3) give larger UO_3 pressures than the calculation presented by Ozrin et al. [5], which utilizes the Lindemer/Besmann model. However, the principal cause of the disparity in the curves in Fig. 3 is the free energy of formation of UO_3(g), which is different in the cases examined.

The equilibrium constant of Eq. (10) from Blackburn [8], Ozrin (i.e., VICTORIA) [5] and AECL (i.e., Ackermann and Chang) [6] are compared to values of K_3 from two critical evaluations of uranium thermochemistry, namely those of Cordfunke and Konings [14] and LLNL [15,16]. Rather than clarify the discrepancies in the $UO_3(g)$ formation free energy, the two critical evaluations serve to solidify the division into two camps. As shown in Fig. 4, the uncertainty in the thermochemistry of UO_3 over pure urania in a gas of known oxygen pressure and temperature exceeds one order of magnitude at 2000 K and two orders of magnitude at 1500 K. Of these, the line based on the recent LLNL work (which is both experimental and theoretical) is judged to be the most accurate. This work consisted in part of classical transpiration experiments with U_3O_8 powder contacted with flowing steam. Care was taken to vary the gas flow rate to insure operation on the flow-rate-independent plateau that is characteristic of equilibrium conditions. UO_3 and $UO_2(OH)_2$ volatilities were independently measured by varying the O_2/H_2O ratio in the gas.

Krikorian et al. [15] also reanalyzed the data of Ackermann et al. [17] and Dharwadkar et al. [18] using the third-law method and found good agreement among the three sets of experiments. The free energy function given by Ebbinghaus [16] is

$$\left(\frac{G^0 - H^0_{298}}{T}\right)_{UO_3(g)} = -288.9 - 0.0651T + 7.93 \times 10^{-6}T^2, \quad \frac{J}{mol - K}, \quad 1500\ K \leqslant T \leqslant 2000\ K. \tag{12}$$

From Ref. [19], $\Delta H^0_{f,298}(UO_3(g)) = -796.7 \pm 3.5$ kJ/mol. The corresponding properties for $UO_2(s)$ and $O_2(g)$ are given in Ref. [14]. The recommended value of K_3 derived from these values is

$$K_3 = \exp\left(-\frac{30\,800}{T} + 11.26\right). \tag{13}$$

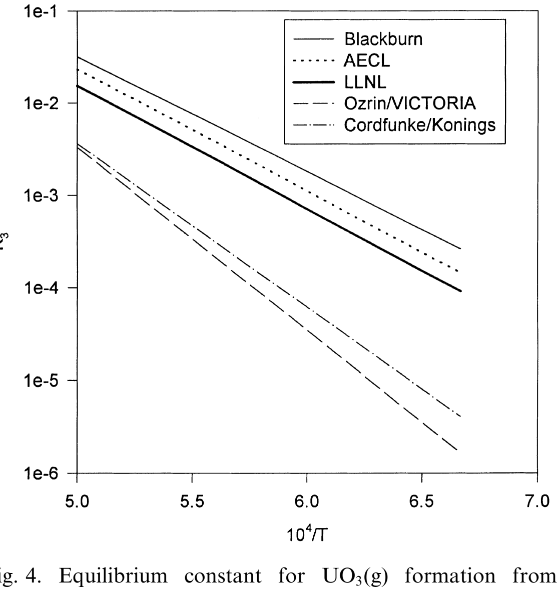

Fig. 4. Equilibrium constant for $UO_3(g)$ formation from $UO_2(s)$.

In applying Eq. (9) to calculate the UO_3 pressure, inaccuracies resulting from lack of accord in the relation between the fuel oxygen potential and fuel stoichiometry can be removed by recognizing that the oxygen pressure is established by the gas phase, not by the fuel. Although these O_2 pressures are equal at equilibrium, the former is better established than the latter. Additionally, in a severe accident, the oxygen pressure characteristic of the flowing steam–hydrogen mixture is imposed on the fuel, not vice versa. The uncertainties in the fuel oxygen potential model affect the calculation of the UO_2 activity in Eq. (9). However, Fig. 2 shows that significant errors in the O/U ratio incurred by processing the gas-phase O_2 pressure through an oxygen potential model do not greatly affect the UO_2 activity, which in any case is not too different from unity.

3. Effect of H_2 in steam

In pure steam, the oxygen pressure due to dissociation of H_2O is given by Eq. (3). In steam containing a controlled amount of hydrogen, the oxygen pressure is

$$p_{O_2} = \left(\frac{p_{H_2O}/p_{H_2}}{K_W}\right)^2. \tag{14}$$

In Fig. 5, a relatively modest addition of H_2 to the steam is seen to produce reductions in the UO_3 pressure of an order of magnitude at 2000 K to 2–3 orders of magnitude at 1500 K. Because hydrogen is generated in a severe fuel damage accident by the steam–Zircaloy reaction, the volatility of the fuel may be significantly suppressed compared to the steam-only laboratory experiments.

4. Effect of uranium oxyhydroxide

In addition to UO_3, the other volatile uranium species likely to be produced in a severe accident is $UO(OH)_2$. Ref. [19] provides the standard enthalpy of formation of the oxyhydroxide: $\Delta H^0_{f,298}(UO_2(OH)_2(g)) = -1200 \pm 10$ kJ/mol. Its free-energy function is given in Ref. [16]

$$\left(\frac{G^0 - H^0_{298}}{T}\right)_{UO_2(OH)_2(g)} = -337.8 - 0.1033T + 1.20 \times 10^{-5}T^2, \quad \frac{J}{mol - K}, \quad 1500\ K \leqslant T \leqslant 2000\ K. \tag{15}$$

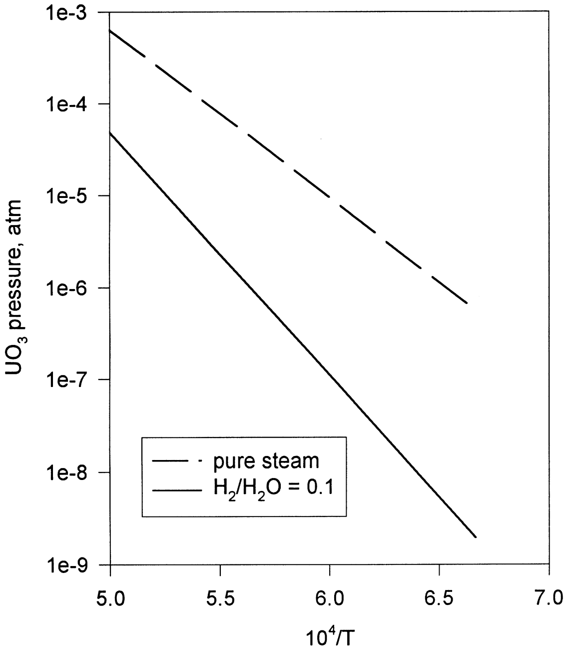

Fig. 5. Effect of hydrogen in steam on UO_3 pressure. K_3 from Eq. (13); urania stoichiometry and UO_2 activity from Blackburn.

This information and the corresponding properties for UO_3(g) and H_2O(g) permit calculation of the oxyhydroxide pressure by the equilibrium reaction

$$UO_3(g) + H_2O(g) = UO_2(OH)_2(g). \tag{16}$$

The total pressure of the uranium-bearing vapors is the sum of the pressures of UO_3 and $UO_2(OH)_2$

$$p_{U,tot} = p_{UO_3} + p_{UO_2(OH)_2} = p_{UO_3}\left(1 + K_{oxy}p_{H_2O}\right), \tag{17}$$

where K_{oxy} is the equilibrium constant of Eq. (16). Calculations based on the thermochemical properties given above show that K_{oxy} ranges from 0.19 at 1500 K to 0.010 at 2000 K. Thus, oxyhydroxide formation is important only at low temperatures and for accident scenarios involving high steam pressures.

5. Effect of burnup

The reaction governing the UO_3 pressure over fuel containing dissolved fission products is

$$UO_2(\text{in } MO_y) + \frac{1}{2}O_2 = UO_3(g), \tag{18}$$

where the UO_2 component is dissolved in a mixed oxide MO_y which consists, in addition to UO_2, of oxides such as MoO_2, ZrO_2, $REO_{1.5}$, and (Ba,Sr)O. In order to apply Eq. (9) to determine the UO_3 partial pressure, the activity of UO_2 in the mixed oxide must be estimated. The UO_2 activity is reduced from the value in pure UO_{2+x} because some uranium has been removed by fission and replaced by fission product cations of generally lower valence.

After a fractional burnup β, 1 mol of uranium is converted to $\beta\sum Y_i$ moles of soluble cations, where Y_i is the nuclear yield of the stable isotopes of the ith element (or group of elements). The summation includes only fission products that dissolve in the fluorite lattice. Only stable isotopes are included because their concentrations significantly exceed that of the radioactive species at typical burnups. The remaining heavy metal (uranium plus ingrown plutonium and lesser amounts of neptunium and americium) is $N_{HM} = 1 - \beta$, so the total moles of metal is

$$N_M = 1 + \left(\sum Y_i - 1\right)\beta. \tag{19}$$

In order to determine the equilibrium O/M ratio in a gas of specified oxygen potential, the theory of Rand and Markin developed for $(U,Pu)O_{2+x}$ is utilized [20]. In this model, the dopant cations are assigned fixed oxidation states characterized by valence V_i and the average uranium valence V_U is permitted to exceed 4. The basic assumption of the mean-valence model is that V_U depends only on the temperature and the oxygen pressure of the ambient gas, but does not depend on whether the oxide is pure UO_{2+x} or the mixed oxide representing irradiated fuel. The theory appears to work satisfactorily in synthetic high-burnup fuel [21].

With the uranium valence fixed, the O/M ratio of the oxide is determined by the condition of electrical neutrality, which leads to

$$\begin{aligned}\frac{O}{M} &= \frac{V_U}{2}\,\frac{1 + (\sum(V_i/V_U)Y_i - 1)\beta}{1 + (\sum Y_i - 1)\beta} \\ &\cong \frac{V_U}{2}\left[1 - \beta\sum\left(1 - \frac{V_i}{V_U}\right)Y_i\right].\end{aligned} \tag{20}$$

With V_U fixed by the temperature and the oxygen pressure of the gas, the O/M ratio is seen to be less than the corresponding O/U ratio of the fresh fuel (O/U = V_U/2). This is because the oxidation states of the

Table 1
Soluble fission products

Fission product	Yield	Valence
Zr	0.30	4
Mo [a]	0.24	4
RE [b]	0.53	3
Ba,Sr	0.15	2

[a] The noble metals form a separate phase and do not dissolve in the oxide. Molybdenum, which can partition between fuel and the noble metal fission product precipitates, has been assumed to be completely oxidized and soluble in the fuel.
[b] Includes La, Ce, Pr, Nd, Pm, Sm, Eu, Gd and Y.

soluble fission product cations are less than the valence of uranium in the fuel. Table 1 shows the yields (for ^{235}U) and oxidation states of the principal fuel-soluble fission products. Solubility limits to incorporation of non-volatile fission products in the oxide lattice have been ignored.

As an example, for $T = 1750$ K in pure steam at 1 atm pressure, $p_{O_2} = 7.2 \times 10^{-4}$ atm and at zero burnup, O/U = 2.17 from either oxygen potential model. The uranium valence is $2 \times 2.17 = 4.34$ and the UO_2 activity in the fresh fuel is 0.69 (Blackburn curve in Fig. 2). Preserving this valence in fuel with burnup $\beta = 0.05$, Eq. (18) gives O/M = 2.14, or 0.03 units less than the fresh fuel in the same environment.

In order to evaluate the UO_2 activity in the mixed oxide, a model of multicomponent nonideal solutions would be needed. Lacking such a general theory, two approximations that are a level higher than ideal solution theory are applied.

5.1. Method 1

The solution is represented as a mixture of atomic oxygen and the mixed stoichiometric oxide MO_y, which contains UO_2 and the oxides of the elements in Table 1. The activity of UO_2 consists of two parts: the first describes the effect of the excess oxygen on the activity of MO_y in the O – O_y pseudo-binary solution. This contribution, a_{MO_y}, is assumed to follow the behavior shown in Fig. 2 for the O – UO_2 binary. The second part, $a^*_{UO_2}$, is the activity of UO_2 in the pseudo-binary UO_2–MO_y,. These two components are combined to yield

$$a_{UO_2} = a^*_{UO_2}\, a_{MO_y} = \gamma^*_{UO_2}\, z_U\, a_{MO_y}, \tag{21}$$

where $z_U = N_U/N_M$ is the cation fraction of uranium in the oxide and $\gamma^*_{UO_2}$ is the activity coefficient of UO_2 in the stoichiometric oxide MO_y. It is assumed to follow regular solution theory, for which

$$RT\ \ln \gamma^*_{UO_2} = \Omega(1 - z_U)^2, \tag{22}$$

where R is the gas constant and Ω is the interaction parameter. In the UO_2 – $NdO_{3/2}$ binary oxide, $\Omega = -27$ kJ/mol [22], and this value is assumed to apply to the mixed oxide containing the oxides of the elements in Table 1. Using the numerical values in the previous example, $a_{MO_y} = 0.78$, $z_U = 0.94$, and $\gamma^*_{UO_2} = 0.99$. Eq. (21) gives $a_{UO_2} = 0.72$ by this method.

5.2. Method 2

In this approximate method, the activity coefficient of UO_2 in UO_{2+x} is assumed to be the same as in the mixed oxide representing spent fuel. This is equivalent to assuming that the gas phase controls the uranium valence, which fixes the UO_2 activity coefficient just as it does the oxygen pressure. The activity of UO_2 is written as

$$a_{UO_2} = \gamma_{UO_2}\, z_{UO_2}. \tag{23}$$

In pure UO_{2+x}, the activity of UO_2 is given by the plots in Fig. 2. In the O – UO_2 binary solution, the mol fraction of UO_2 is $z_{UO_2} = (1+x)^{-1}$. In the previous example using $UO_{2.17}$, the activity of UO_2 is 0.69 and the mol fraction of UO_2 is 0.86. Therefore the activity coefficient is $\gamma_{UO_2} = 0.69/0.86 = 0.80$. In the mixed oxide, the mol fraction of UO_2 is

$$z_{UO_2} = \frac{1-\beta}{(\mathrm{O/M} - 1)[1 + (\sum Y_i - 1)\beta]}. \tag{24}$$

For a fractional burnup of 5%, the value of O/M = 2.14 calculated in the preceding example, and the yields from Table 1, Eq. (24) gives $z_{UO_2} = 0.82$. Assuming that the activity coefficient is the same as in the fresh fuel with the same uranium valence, the UO_2 activity by this method is $a_{UO_2} = 0.80 \times 0.82 = 0.66$.

Comparison of the UO_2 activities in high burnup fuel by these two methods shows that the first method gives a value greater than that in fresh UO_{2+x} while the second method predicts a slight reduction. However, the values of a_{UO_2} for the fresh fuel and for the irradiated fuel (by either method) are within 10% of each other, and neither are very far from unity.

6. Conclusions

Disparities in existing literature values of the standard free energy of formation of UO_3(g) have been reviewed. The recommended choice is based on recent work by LLNL, which gives values of the equilibrium constant for formation of UO_3(g) from UO_2(s) that are ~30% lower than the earlier data of Ackermann and Chang.

Disagreement in predicted O_2 pressures over UO_{2+x} between the two most widely used models can be as large as one order of magnitude. This conflict cannot be resolved with the present database. The third quantity needed to calculate UO_3 pressures is the activity of UO_2 in the oxide. This property is better established than the oxygen pressure of the oxide.

The recommended method for calculating the UO_3 pressure is to apply Eq. (9) with

- K_3 from Eq. (13).
- p_{O_2} from Eq. (3) in pure steam or Eq. (14) in H_2O/H_2 mixtures.
- Oxide stoichiometry from Eq. (1) or Eqs. (2a) and (2b) (or Fig. 1).
- a_{UO_2} from Eq. (6) or Eq. (7) (or Fig. 2).

Minor additions of H_2 to steam drastically reduces the gas oxygen potential and hence the UO_3 pressure. The oxyhydroxide of uranium, $UO_2(OH)_2$(g), is a minor contributor to the total pressure of uranium-bearing vapors at high temperatures and in atmospheric pressure steam. The effect of the burnup on the UO_3 pressure is not significant compared to other uncertainties involved in fixing this property.

Acknowledgements

The useful comments of Dr Wei-E Wang are gratefully acknowledged.

References

[1] C.A. Alexander, J.S. Ogden, High Temp. – High Press. 22 (1990) 149.
[2] J. Abrefah et al., J. Nucl. Mater. 208 (1994) 98.
[3] B.J. Lewis et al., J. Nucl. Mater. 227 (1995) 83.
[4] T.J. Heames et al., VICTORIA: A mechanistic model of radionuclide behavior under severe accident conditions, NUREG/CR-5545, 1992.
[5] V. Ozrin et al., Models for release of low-volatile fission products, Nuclear Safety Institute of the Russian Academy of Sciences NSI-SARR-43-96, 1996.
[6] B.J. Lewis et al., J. Nucl. Mater. 252 (1998) 235.
[7] D.S. Cox et al., A model for the release of low-volatility fission products in oxidizing conditions, AECL-10440, 1991.
[8] P.E. Blackburn, J. Nucl. Mater. 46 (1973) 244.
[9] T.B. Lindemer, T.M. Besmann, J. Nucl. Mater. 130 (1985) 472.
[10] D.R. Olander, Nucl. Technol. 74 (1986) 215.
[11] C.A. Alexander, PhD thesis, Ohio State University, 1961.
[12] R.J. Ackermann, A.T. Chang, J. Chem. Thermo. 5 (1973) 873.
[13] J.S. Punni and P.K. Mason, UO_2 oxidation and volatilisation, AEAT-1277, 1997.
[14] E.H.P. Cordfunke, R.J.M. Konings (Eds.), Thermochemical Data for Reactor Materials and Fission Products, North-Holland, 1990.
[15] O.H. Krikorian et al., Experimental studies and thermodynamic modeling of volatilities of uranium, plutonium, and americium from their oxides, Lawrence Livermore National Laboratory Report UCRL-ID-114774, 1993.
[16] B.B. Ebbinghaus, Calculated thermodynamic functions for gas-phase uranium, neptunium, plutonium, and americium oxides, oxyhydroxides, oxychlorides, and oxyfluorides, Lawrence Livermore National Laboratory Report UCRL-JC-122278, 1995.
[17] R.J. Ackermann et al., J. Phys. Chem. 64 (1960) 350.
[18] S.R. Dharwadkar et al., Thermodynamic properties of gaseous uranium hydroxide, in: Proceedings of the Symposium on the Thermodynamics of Nuclear Materials 1974, vol. II, IAEA, Vienna, 1975, p. 455.
[19] B.B. Ebbinghaus, O.H. Krikorian, D.L. Fleming, Thermodynamic study of $UO_3(g)$, $UO_2(OH)_2$, $UO_2Cl_2(g)$ and $UO_2F_2(g)$, unpublished manuscript.
[20] M.H. Rand, T.L. Markin, in: Proceedings of the Symposium on the Thermodynamics of Nuclear Materials – 1967, IAEA, Vienna, 1968, p. 637.
[21] K. Une, M. Oguma, J. Nucl. Sci. Technol. 20 (1983) 844.
[22] W.-E. Wang, D.R. Olander, T.B. Lindemer, J. Nucl. Mater. 211 (1994) 85.