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Phase diagram analysis of (U,Pu)O_{2-x} sub-system

Renu Agarwal *, B.K. Sen, V. Venugopal

Product Development Section, Radiochemistry and Isotope Group, Bhabha Atomic Research Centre, Mumbai 400085, India

ABSTRACT

The high plutonium, hypo-stoichiometric fuel exists as two phase system at low temperatures. The partial phase diagram of (U,Pu)O_{2-x} with two coexisting cubic phases was extensively investigated in this work using theoretical models. The critical temperature of the miscibility gap varies with Pu/M and O/M of the system. Based on the similar miscibility gap behaviour observed in PuO_{2-x} system and the experimental data available on the phase boundaries of (U,Pu)O_{2-x} for various Pu/M, some semi-empirical relationships and solution models were developed. With the help of these relationships, ternary isothermal sections of the miscibility gap, O/M at different temperatures and the critical temperature of the miscibility gap of (U,Pu)$_{2-x}$ for different Pu/M values were calculated. These calculated values were compared with the available literature data.

1. Introduction

The $(U_{(1-p)}Pu_p)O_{2-x}$ system is extensively investigated [1–7]. Due to their relevance in fast breeder reactor programme, most of these studies are limited to Pu/M < 0.3 (where M represents U + Pu). However, with increasing interest in plutonium rich fast breeder reactor fuels, interest in the *MOX* fuels with $p > 0.3$ is also increasing. The hypo-stoichiometric *MOX* with high plutonium content is known to be biphasic at low temperatures [8,9]. The two coexisting phases of (U,Pu)O_{2-x} system, with $p > 0.35$, can be two FCC phases or FCC with cubic or rhombohedral phase [10], depending on the Pu/M, O/M and temperature. Presence of even 3 at.% uranium is known to suppress the appearance of hexagonal, Pu_2O_3 type phase [8]. The (U,Pu)O_{2-x} system is a single phase FCC at all temperatures for Pu/$M \leqslant 0.3$. Markin and Street [5] have reported that the mixed oxide, (U,Pu)O_{2-x}, is stable as single phase for Pu/M = 0.11, 0.15 and 0.3, from room temperature to at least 1400 K. Brett et al. [1,2] have shown that the reduction of (U,Pu)O_{2-x} with Pu/M > 0.4 results in two cubic phases at room temperature, $MO_{2.00}$ and an oxygen deficient MO_{2-x} cubic fluorite phase. Therefore, the transition from single phase to biphasic hypo-stoichiometric system takes place at Pu/M between 0.3 and 0.4 [1,5]. Markin and Street investigated six oxides of $(U_{0.58}Pu_{0.42})O_{2-x}$ with O/M between 1.79 and 2.00, using high temperature camera. They observed that these oxides became single phase from two phase cubic in the temperature range 473–773 K, depending on the O/M ratio. They found that in case of $(U_{0.58}Pu_{0.42})O_{2-x}$, the two phases in equilibrium were MO_2 and $MO_{1.789}$. For Pu/M = 0.58, they studied only one value of O/M corresponding to the composition, $(U_{0.42}Pu_{0.58})O_{1.722}$ and found that in this case the two phase system became single phase at 773 K. By analogy with $(U_{0.58}Pu_{0.42})O_{2-x}$, they deduced that in this case the sub-stoichiometric phase in equilibrium with MO_2 was $MO_{1.710}$. By comparing the temperatures of transition of (U,Pu)O_{2-x} as a function of Pu/M, it was seen that the transition temperature increases as Pu/M increases. However, for the same oxidation state of plutonium, transition temperature can be different. The oxidation state of plutonium is same for $(U_{0.58}Pu_{0.42})O_{1.8}$ and $(U_{0.42}Pu_{0.58})O_{1.72}$, i.e., 3.04, and the temperatures of transition from biphasic to single phase are 648 and 773 K, respectively.

All the experimentally determined thermodynamic data, e.g. oxygen potential values and partial pressures of plutonium bearing species available in literature for (U,Pu)O_{2-x} system are for high temperature, single phase system [11–18]. Many theoretical models, e.g. Chereau [19], Besmann and Lindemer [20], Blackburn and Johnson [21] etc., are available in literature to calculate oxygen potential of the single phase (U,Pu)O_{2-x} system, valid at high temperatures and low Pu/M. At lower temperatures, where the system segregates into two cubic phases, its thermo-physical behaviour is different. Yamanaka et al. [22] have carried out phase diagram assessment and thermodynamic modeling of U–Pu–O system using sub-lattice model. However, due to their assessment of the complete composition range, they did not carry out detailed investigation of the narrow composition region of our interest. To be able to estimate the thermo-physical properties of this biphasic region, the Gibbs energy of the system was defined using the available phase diagram data in this region. Lukas computer program [23,24] was used for the calculations. The phase boundaries of the miscibility gap of the system calculated for different compositions and temperatures were compared with the literature values.

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* Corresponding author. Tel.: +91 22 25590645.
E-mail address: arenu@barc.gov.in (R. Agarwal).

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2. Calculation method

The miscibility gap of $(U,Pu)O_{2-x}$ system at low temperature was calculated by defining the ternary system U–Pu–O as a solution of PuO_2–UO_2–$PuO_{1.5}$. The Gibbs energies of formation of these compounds used for the present calculations are given in Table 1. These values were calculated from the Gibbs energy equations of the compounds, PuO_2 [22], $PuO_{1.5}$ [25], UO_2 [22] and the corresponding elements [26]. The thermodynamic stability of the solution was expressed in terms of Gibbs energies of formation of the constituent compounds and their binary interaction parameters while ignoring the presence of any ternary interactions.

$$G = \Sigma x_i{}^{o}G_i + RT\Sigma x_i \ln x_i + {}^{xs}G \tag{1}$$

where ${}^{o}G_i$ represents the Gibbs energies of the compounds UO_2, PuO_2 and $PuO_{1.5}$, x_i is their mole fractions in the solution and ${}^{xs}G$ is the excess Gibbs energy of the ternary solution due to non-ideal binary interactions of UO_2–PuO_2, PuO_2–$PuO_{1.5}$ and UO_2–$PuO_{1.5}$. The excess Gibbs energies of the binary interactions were expressed as a function of composition by the following Redlich–Kister polynomial [27]:

$${}^{xs}G = x_i(1 - x_i)(a_0 + a_1(1 - 2x_i)), \tag{2}$$

where x_i is the mole fraction of the second component of the binary and a_0, a_1 are temperature dependent coefficients, e.g., a_0 can be expressed as follows:

$$a_0 = A_0 + B_0T. \tag{3}$$

The binary interaction parameters were extrapolated to the ternary solution using Muggianu's model [28].

$$\begin{aligned} {}^{xs}G = {} & x_{PuO_2}x_{UO_2}(a_0 + a_1(x_{PuO_2} - x_{UO_2})) \\ & + x_{PuO_2}x_{PuO_{1.5}}(a'_0 + a'_1(x_{PuO_2} - x_{PuO_{1.5}})) \\ & + x_{UO_2}x_{PuO_{1.5}}(a''_0 + a''_1(x_{UO_2} - x_{PuO_{1.5}})), \end{aligned} \tag{4}$$

where a_0 and a_1, a'_0 and a'_1, and a''_0 and a''_1, are the temperature dependent, binary interaction parameters of PuO_2–UO_2, PuO_2–$PuO_{1.5}$ and UO_2–$PuO_{1.5}$ systems, respectively.

Before carrying out the calculations for the ternary system, the interaction parameters of PuO_2–$PuO_{1.5}$ were calculated so as to reproduce the miscibility gap in FCC region of Pu–O binary system. The interaction parameters of other two binaries, PuO_2–UO_2 and UO_2–$PuO_{1.5}$ were calculated such as to best reproduce the ternary phase diagram data. As the data available in literature on U–Pu–O system was very limited, the number of interaction parameters used for the calculations were kept as low as possible.

3. Results and discussion

3.1. PuO_{2-x} FCC miscibility gap

The pure Pu–O system shows an FCC miscibility gap between PuO_2–$PuO_{1.7}$ [29–36]. The O/M of 1.7 indicates that up to 60 at.% plutonium can be reduced to the oxidation state III in pure Pu–O system while retaining the FCC structure. However, according to the reported experimental data in the FCC miscibility gap region of the U–Pu–O ternary system, for some investigated Pu/M values, almost all the plutonium was found to be in oxidation state III, corresponding to $PuO_{1.5}$ compound. This was especially true for the biphasic ternary system with higher uranium content. Therefore, interaction parameters of PuO_2–$PuO_{1.5}$ system were calculated instead of PuO_2–$PuO_{1.7}$ system. The interaction parameters of PuO_2–$PuO_{1.5}$ binary could reproduce the miscibility gap of PuO_2–$PuO_{1.7}$ region. The system shows a strong asymmetric composition dependence which was taken care by using first-order Redlich–Kister polynomial coefficients in the excess Gibbs energy of the PuO_2–$PuO_{1.5}$ binary system. These interaction parameters are given in Table 2. The miscibility gap in PuO_{2-x} calculated using these coefficients is compared with experimental data in Fig. 1. The critical temperature and composition of the calculated miscibility gap were found to be 929.3 K and $PuO_{1.86}$, respectively. These values are in reasonably good agreement with the experimentally observed values of 930 K and $PuO_{1.875}$ [6]. The small difference in O/Pu can be ignored considering the flat miscibility gap curve near critical temperature. Phase boundaries calculated from the interaction parameters given by Besmann and Lindemer [37] are also

Table 1
Gibbs energies of formation of compounds used for the present calculations.

Compound	Temperature range	Δ_fG^o (J/mol) = $A + BT + CT\ln(T) + DT^2 + E/T + FT^3$					
		A	*B*	*C*	*D*	*E*	*F*
PuO_2	<400 K	−1072930.651	476.372974	−42.7341199	0.02676598	990235	$-1.3237149 \times 10^{-6}$
	400–487.9 K	−1075453.306	433.425205	−33.4439199	0.01088598	990235	$-1.3237149 \times 10^{-6}$
	487.9–593.9 K	−1077384.190	468.349287	−38.8366199	0.01583548	990235	$-1.3237149 \times 10^{-6}$
	593.9–736 K	−1076406.179	429.087820	−32.3818199	0.00975948	990235	$-1.3237149 \times 10^{-6}$
	736–757 K	−1074204.653	383.324276	−25.2999199	0.00435598	990235	$-1.3237149 \times 10^{-6}$
	757–913 K	−1077436.143	399.795399	−27.1399199	0.00435598	990235	$-1.3237149 \times 10^{-6}$
	913–944 K	−1070329.098	332.401053	−18.4412199	0.00570091	410910	$-1.5871579 \times 10^{-6}$
	944–1000 K	−1072472.904	336.226103	−18.6119199	0.00435598	990235	$-1.3237149 \times 10^{-6}$
	>1000 K	−1066297.118	259.900267	−7.2563199	−0.00465020	387695	$-1.3584924 \times 10^{-8}$
$PuO_{1.5}$	<400 K	−828725.656	−189.890386	54.1493800	−0.10744517	−682131.25	3.7998589×10^{-5}
	400–487.9 K	−839456.541	253.440156	−17.3567500	0.00704151	432845	$-9.9278536 \times 10^{-7}$
	487.9–593.9 K	−841387.425	288.364238	−22.7494500	0.01199101	432845	$-9.9278536 \times 10^{-7}$
	593.9–736 K	−840409.414	249.102771	−16.2946500	0.00591501	432845	$-9.9278536 \times 10^{-7}$
	736–757 K	−838207.888	203.339227	−9.2127500	0.00051151	432845	$-9.9278536 \times 10^{-7}$
	757–913 K	−841439.378	219.810350	−11.0527500	0.00051151	432845	$-9.9278536 \times 10^{-7}$
	913–944 K	−834332.333	152.416004	−2.3540500	0.00185644	−146480	$-1.2562284 \times 10^{-6}$
	944–1000 K	−836476.139	156.241054	−2.5247500	0.00051151	432845	$-9.9278536 \times 10^{-7}$
	>1000 K	−831844.300	98.996677	5.9919500	−0.00624313	−19060	$-1.0187863 \times 10^{-8}$
UO_2	<941.5 K	−1096685.816	353.902832	−25.4612000	0.00284317	687172	3.274488×10^{-6}
	941.5–1000 K	−1090766.241	240.689963	−9.4516000	0.00409473	725740	-1.151562×10^{-6}
	1000–1049 K	−1084590.455	164.364127	1.9040000	−0.00491145	123200	1.58568×10^{-7}
	1049–1408 K	−1094219.399	205.845512	−2.7402000	−0.00491145	123200	1.58568×10^{-7}
	1408–1500 K	−1088751.464	126.734954	7.6362000	−0.00491145	123200	1.58568×10^{-7}
	>1500 K	−1684123.044	4062.828047	−522.3914000	0.20646760	126054690	-1.584504×10^{-5}

Table 2
The Redlich–Kister binary interaction parameters used for the calculations.

Binary phases	Composition parameters	^{xs}G (J/mol)	
		A	*B*
Present calculations			
PuO_2–UO_2	$x(1-x)$	0.0	0.0
	$x(1-x)(1-2x)$	15000.0	−14.0
PuO_2–$PuO_{1.5}$	$x(1-x)$	−45000.0	59.0
	$x(1-x)(1-2x)$	100000.0	−100.0
UO_2–$PuO_{1.5}$	$x(1-x)$	−37800.0	60.5
	$x(1-x)(1-2x)$	32800.0	−36.5
Besmann–Lindemer [37]			
PuO_2–UO_2	$x(1-x)$	0.0	0.0
	$x(1-x)(1-2x)$	0.0	0.0
PuO_2–$Pu_{4/3}O_2$	$x(1-x)$	0.0	0.0
	$x(1-x)(1-2x)$	41510.0	−30.2
UO_2–$Pu_{4/3}O_2$	$x(1-x)$	0.0	0.0
	$x(1-x)(1-2x)$	0.0	0.0

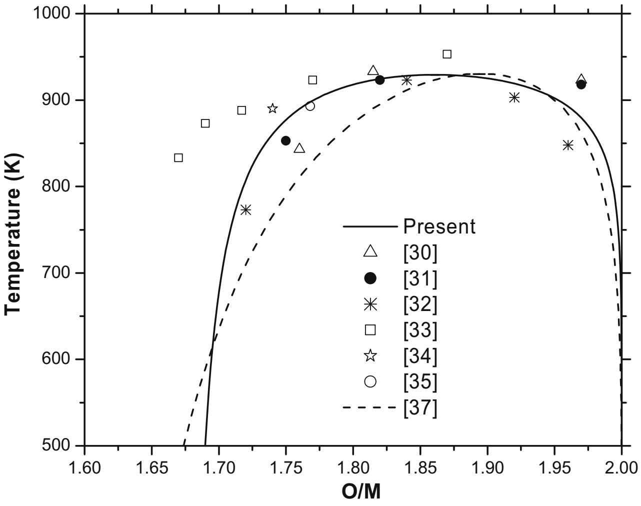

Fig. 1. A comparison of calculated miscibility gap in PuO_{2-x} with literature values.

shown in the figure. The present set of interaction parameters gave better agreement with the experimental data than the ones calculated from the parameters given by Besmann and Lindemer. The miscibility gap calculated from the interaction parameters given by the later was not as wide and flat near critical temperature as was found experimentally. However, the present interaction parameters could closely reproduce this flat immiscibility region. This is mainly because Besmann and Lindemer had ignored zeroth-order Redlich–Kister polynomial coefficient that results in symmetric change in the Gibbs energy of the solution with change in composition. The present coefficient values indicate that with increase in temperature, especially near critical temperature, contribution due to the first-order Redlich–Kister coefficients decreases and contribution due to the zeroth-order coefficients increases. Therefore, near critical temperature of the miscibility gap, zeroth-order Redlich–Kister polynomial plays an important role. The asymmetric composition dependence of first-order Redlich–Kister polynomial results in high fraction of x_{PuO_2} in near-stoichiometric phase in equilibrium with a hypo-stoichiometric phase with smaller fraction of $x_{PuO_{1.5}}$. The symmetric composition dependence of the zeroth-order Redlich–Kister polynomial tends to bring equality in these two fractions of the equilibrium phases. Therefore, increase in contribution of the zeroth-order coefficient with increase in temperature results in a near constant value of $x_{PuO_{1.5}}$ in hypo-stoichiometric phase, causing a very flat and wide miscibility gap near critical temperature.

Gueneau et al. [25] have recently reported thermodynamic modeling of Pu–O system based on extensive investigation of its thermodynamic and phase diagram properties. However, as our interest was limited to the narrow miscibility gap of PuO_{2-x} phase, we calculated excess Gibbs energy of solution of PuO_2–$PuO_{1.5}$ system, which was used for the calculation of Gibbs energy values of PuO_2–UO_2–$PuO_{1.5}$ system.

3.2. $(U,Pu)O_{2-x}$ FCC miscibility gap

During the present calculations, the FCC solution of U–Pu–O system was assumed to be a solution of three cubic phases, UO_2, PuO_2 and $PuO_{1.5}$. The ternary U–Pu–O system should have at least 40 at.% of the cationic lattice sites in IV oxidation state to retain cubic structure. The system with 30 at.% Pu/*M* is known to exist as single phase FCC even when all the plutonium is reduced to III oxidation state [5]. However, the system with very high plutonium content has a fraction of the plutonium reduced to oxidation state III. The variation in the oxidation states with change in Pu/*M* of the system was accommodated by changing the fractions of PuO_2 and $PuO_{1.5}$. The Redlich–Kister polynomial interaction parameters of the other two binaries, PuO_2–UO_2 and UO_2–$PuO_{1.5}$, used for the present calculations of the ternary system are given in Table 2. The ternary isothermal miscibility gaps of the PuO_2–UO_2–$PuO_{1.5}$ system at different temperatures, calculated using these coefficients, are plotted in Fig. 2. In this figure, a constant value of x_{UO_2} represents constant Pu/*M* and a constant value of $x_{PuO_{1.5}}$ means constant O/*M*. The tie-lines shown in the figure indicate that the difference in Pu/*M* of the equilibrium phases is more pronounced at lower temperatures and lower O/*M* fuels. According to the degree of freedom principle, the composition of the coexisting phases changes even in isothermal condition by changing Pu/*M* or O/*M*. Thus a plutonium rich $(U,Pu)O_{2-x}$ system at low temperatures has two coexisting phases with similar Pu/*M* but very different O/*M*. Whereas, a uranium rich system has one sub-stoichiometric phase with very high Pu/*M* in equilibrium with a uranium rich near-stoichiometric phase.

Besmann and Lindemer [37] have also carried out calculations of the miscibility gap in hypo-stoichiometric, FCC region of U–Pu–O system. They have considered this region to be a solution

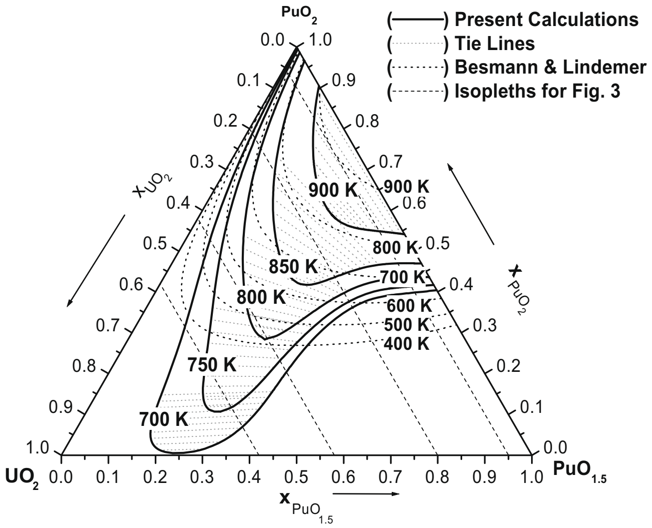

Fig. 2. A comparison of isothermal miscibility gaps in $(U,Pu)O_{2-x}$ obtained from the present calculations with that of Besmann and Lindemer.

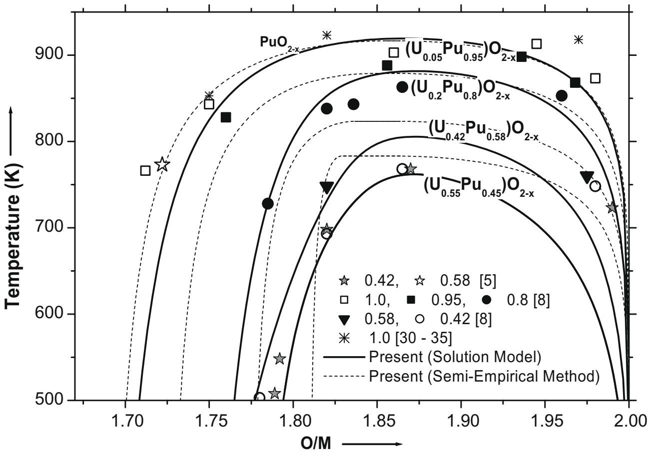

Fig. 3. A Comparison of calculated isopleth phase boundaries with experimental data (Pu/*M* values of experimental data given with symbols).

of UO_2–PuO_2–$Pu_{4/3}O_2$–$U_2O_{4.5}$. In their earlier work [20] they had calculated oxygen potential of the single phase (U,Pu)O_{2-x} system. In that work also they had assumed that the system was a solution of these four compounds. Our recalculations of the system using the method suggested by them, based on Newton–Raphson iteration technique, showed that at high temperatures the fraction of $U_2O_{4.5}$ compound in the solution is reasonable to affect the oxygen potential of the system. However, at low temperatures (<1000 K) the fraction of $U_2O_{4.5}$ is practically zero. That can also be understood from the instability of U(VI) in highly reducing atmosphere, especially at low temperatures when the defect structures are relatively unstable. The ternary isothermal miscibility gaps calculated using the interaction parameters given by Besmann and Lindemer are also plotted in Fig. 2. The scale of the mole fraction of the third component was adjusted for $PuO_{1.5}$ instead of $Pu_{4/3}O_2$ used by Besmann and Lindemer [37]. The miscibility curves obtained from the parameters given by Besmann and Lindemer indicated that a solution with Pu/*M* = 0.42 should not show a miscibility gap even at temperature as low as 400 K. This is in contradiction to the experimental observation.

The temperature vs O/*M* relations of the miscibility gaps were calculated from the equilibrium compositions along the isopleths by fixing the UO_2 mole fraction. The isopleths used for these calculations are given in Fig. 2. The temperature vs O/*M* plots for different Pu/*M* values are given in Fig. 3. These isopleths were drawn by calculating the compositions of the major phase in equilibrium with a minor phase of negligible amount. The x_{UO_2} of the major phase was kept constant and equilibrium temperatures were calculated for different values of $x_{PuO_{1.5}}$. The O/*M* values calculated from the compositions of the major phase were used for the plot. The Pu/*M* and O/*M* of the minor phase were completely different from those of the major phase, hence they could not be shown in this two dimensional plot. Therefore, drawing the tie-lines in this figure was not possible. A reasonably good agreement of the calculated values with experimental data can be seen in this figure, indicating the validity of this model. However, Besmann–Lindemer values show very poor agreement with the experimental data, particularly the ones with lower Pu/*M* values. The critical temperatures calculated using Besmann–Lindemer parameters for the systems with low Pu/*M* values were found to be very low compared to the experimental data.

Interestingly it was observed that the temperature vs O/*M* relationship of the miscibility gap for different Pu/*M* of (U,Pu)O_{2-x} system were related to the miscibility gap of PuO_{2-x} system by simple semi-empirical relations. The O/*M* of the hypo-stoichiometric phase was calculated by using the following assumptions:

(i) (U,Pu)O_{2-x} with up to 35 at.% Pu/*M* remains single phase and on complete reduction of $(U_{0.65}Pu_{0.35})O_{2-x}$, all the plutonium is in oxidation state III, corresponding to an O/*M* = 1.825.
(ii) $PuO_{1.7}$ is the sub-stoichiometric phase of Pu–O miscibility gap with only 60 % plutonium in oxidation state III. So the fraction of Pu(III) reduces from 1.0 to 0.6 with increase in Pu/*M*.

A linear interpolation between $(U_{0.65}Pu_{0.35})O_{1.825}$ and PuO_{2-x} gave the following relation:

$$(\mathrm{O}/\boldsymbol{M})_p = \frac{((\mathrm{O}/\boldsymbol{M})_{\mathrm{PuO_2}} - 1.825)}{(1-0.35)}(p-0.35) + 1.825, \tag{5}$$

where $(\mathrm{O}/M)_p$ and $(\mathrm{O}/M)_{\mathrm{PuO_2}}$ represent the O/*M* of the sub-stoichiometric phases of $(U_{(1-p)}Pu_p)O_{2-x}$ and PuO_{2-x}, respectively, and '*p*' is Pu/*M*. For the calculated O/*M* values of $(U_{(1-p)}Pu_p)O_{2-x}$, the temperatures of the miscibility limits were calculated by using another empirical relation:

$$(T)_p = (T)_{\mathrm{PuO_2}} - 252(1-p), \tag{6}$$

where $(T)_p$ and $(T)_{\mathrm{PuO_2}}$ are the temperatures of the miscibility limits corresponding to $(\mathrm{O}/M)_p$ and $(\mathrm{O}/M)_{\mathrm{PuO_2}}$, respectively. A linear interpolation of critical temperatures of miscibility gap for various Pu/*M*, reported in literature, gave a slope of 252. The O/*M* values for the near-stoichiometric phase were assumed to be same as that of oxygen rich phase of Pu–O miscibility gap but the temperatures of miscibility limit for these compositions were calculated using Eq. (6). These values are also plotted in Fig. 3 and show reasonable agreement with the experimental data.

The Gibbs energy of the ternary solution with miscibility gap can be further used for the calculation of oxygen potentials and partial pressures of various gaseous species, e.g., PuO_2, PuO, UO_2 etc. There is no experimental data available for these important thermodynamic parameters of this system in the region of miscibility gap. Defining Gibbs energy values of the system using the phase diagram data is helpful in calculating these thermodynamic parameters. Though full discussion of these thermodynamic parameters is out of scope of this paper and will be discussed in a later publication, it was found that the calculated oxygen potential and partial pressure values show composition and temperature dependence trends similar to the ones observed in the high temperature, single phase $(U_{(1-p)}Pu_p)O_{2-x}$ system. However, in biphasic region, oxygen potential does not change as sharply with change in O/*M*, as observed in single phase region.

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