

Chemical thermodynamic representation of (U, Pu, Am)O_{2-x}

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Abstract

The oxygen potential isotherms of (U, Pu, Am)O_{2-x} were represented by a chemical thermodynamic model proposed by Lindemer et al. It was assumed in the present model that (U, Pu, Am)O_{2-x} consisted of the chemical species [UO₂], [PuO₂], [Pu_{4/3}O₂], [AmO₂] and [Am_{5/4}O₂] in a pseudo-quaternary system by treating the reduction rates of Pu and Am as identical; furthermore an interaction between [Am_{5/4}O₂] and [UO₂] was introduced. The agreement between analytical and experimental isotherms was good, but the analytical values slightly overestimated the experimental values especially in the case of lower Am content. Adding an interaction between [Am_{5/4}O₂] and [PuO₂] to the model resulted in a better representation.

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1. Introduction

Americium (²⁴¹Am) is not only generated in nuclear fuel during irradiation, but it also accumulates in the mixed oxide of U and Pu (MOX) fuel by beta-decay of ²⁴¹Pu (half-life, 14.4 y) during storage. ²⁴¹Am is of note among minor actinides, since it has a large cross-section for neutron absorption and it emits strong gamma-rays. In addition, it merits attention from the view point of reducing the environmental burden in the disposal of high level radioactive waste, because ²⁴¹Am decays to the long-lived and highly radiotoxic

²³⁷Np (half-life, 2.1 × 10⁶ y) by the alpha-decay at the half life of 432 y. Thus, recovery and recycling of Am from spent fuel are, therefore, promising ways to reduce the environmental burden and save natural resources by transmutation in the reactor core [1].

Am is loaded into the reactor core in the form of a homogeneous fuel or a so-called heterogeneous target for transmutation in both existing light water reactors (LWRs) and fast reactors (FRs). For the latter transmutation, MOX fuel with several percent of Am, as (U, Pu, Am)O_{2-x}, is one promising candidate [2]. Using (U, Pu, Am)O_{2-x} is advantageous because it has only a small impact on the core characteristics [3] and existing technology already developed for the MOX fuel can be used.

The R&D program to fabricate and irradiate (U, Pu, Am)O_{2-x} fuel in the FR is now underway in Japan Nuclear Cycle Development Institute [2]. In this program, characterizations of (U, Pu, Am)O_{2-x} fuel are

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also important as well as establishment of its fabrication technology. Oxygen potential is an important property because of its significant effect on the sintering properties and irradiation performance.

The oxygen potentials of $(U, Pu, Am)O_{2-x}$ have, thus, been measured as a function of oxygen to metal (O/M) ratio in the temperature range between 1123 K and 1423 K, and it was found that the oxygen potentials were much higher in $(U, Pu, Am)O_{2-x}$ than in the MOX fuel with the same content of Pu [4]. It is important to estimate the oxygen potential reliably at high temperature from data obtained in a limited temperature range, since the fuel experiences high temperature during irradiation.

The purpose of this study is to give a chemical thermodynamic representation of $(U, Pu, Am)O_{2-x}$ for estimation of the oxygen potentials at various temperatures, especially high temperatures, and for several Am contents. The representation results are discussed in terms of thermodynamic aspects.

2. Measurement data of oxygen potentials for $(U, Pu, Am)O_{2-x}$

Fig. 1 shows the oxygen potential isotherms of $(U_{0.685}Pu_{0.270}Am_{0.045})O_{2-x}$ and $(U_{0.684}Pu_{0.300}Am_{0.016})O_{2-x}$ at 1273 K [4]. The isotherms of MOX [5–8], AmO_{2-x} [9] and $(Am_{0.5}U_{0.5})O_{2-x}$ [10] around 1273 K are also shown there. The oxygen potential measurements for $(U, Pu, Am)O_{2-x}$ were carried out by thermogravimetric analysis with H_2O/H_2 and CO_2/H_2 gas

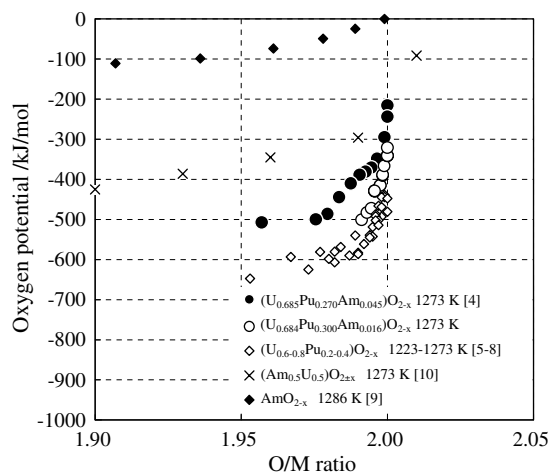


Fig. 1. Oxygen potentials of $(U, Pu, Am)O_{2-x}$ as a function of O/M ratio.

equilibria. Experimental details have been given elsewhere [4]. It is seen that the oxygen potentials of $(U_{0.685}Pu_{0.270}Am_{0.045})O_{2-x}$ are much higher than those of MOX, in other words, the addition of an Am amount as low as 4.5% increases the oxygen potentials. The oxygen potentials of $(U_{0.685}Pu_{0.270}Am_{0.045})O_{2-x}$ are intermediate between those of $(Am_{0.5}U_{0.5})O_{2-x}$ and $(U_{0.7}Pu_{0.3})O_{2-x}$.

3. The representation of $(U, Pu, Am)O_{2-x}$

In this study, the chemical thermodynamic model proposed by Lindemer and Besmann [11] was adopted for the representation of oxygen potentials of $(U, Pu, Am)O_{2-x}$. This model is based on the solute-solvent model, and it assumes the compounds consist of several chemical species which have known thermodynamic properties determined by analyses of experimental data. That is, the actual values of Gibbs energy of formation and interaction energy between the species have been determined by least square analyses of the experimental data. The oxygen potential isotherms of binary and ternary oxides, namely CeO_{2-x} [12], $UO_{2\pm x}$ [11], PuO_{2-x} [13], $(U, Pu)O_{2\pm x}$ [13], $(U, RE)O_{2\pm x}$ [14,15] ($RE =$ rare earth elements) and AmO_{2-x} [16] have been represented well by this model. For example, the two chemical species $Pu_{4/3}O_2$ and PuO_2 and the interaction between them were considered in the representation of PuO_{2-x} [13].

For the representation of oxygen potential isotherms of $(U, Pu, Am)O_{2-x}$ by this model, unfortunately, no attempt has been made to adapt it to the quaternary system. In particular, difficulties are found in the modeling of reduction of Pu and Am in $(U, Pu, Am)O_{2-x}$, which forms both dioxides having only hypostoichiometry. As shown in Ref. [17], the oxygen potential is much higher in AmO_{2-x} than in PuO_{2-x} at the same O/M ratio. This means that Am in AmO_{2-x} is reduced much easier than Pu in PuO_{2-x} when the oxygen potential decreases. Actually, at the oxygen potential of about -140 kJ/mol, O/M ratio of AmO_{2-x} is about 1.85, while that of PuO_{2-x} is 2.00, which means no reduction of Pu. If the same reduction behaviors of Am and Pu are assumed also in $(U, Pu, Am)O_{2-x}$, all AmO_{2-x} are reduced to a lower oxide such as $AmO_{1.62}$ at $O/M = 1.9775$ or less in $(U_{0.685}Pu_{0.270}Am_{0.045})O_{2-x}$, which is considered to be the boundary composition between fcc single-phase and two-phase regions in the Am–O phase diagram [18]. If this priority of AmO_{2-x} reduction in $(U, Pu, Am)O_{2-x}$ is true, some form of discontinuity in the oxygen potential isotherm curve around this O/M ratio is expected to appear, regardless of the temperature. For example, the oxygen potential changes steeply at the boundary of the two phases, and they become constant beyond the boundary. Such

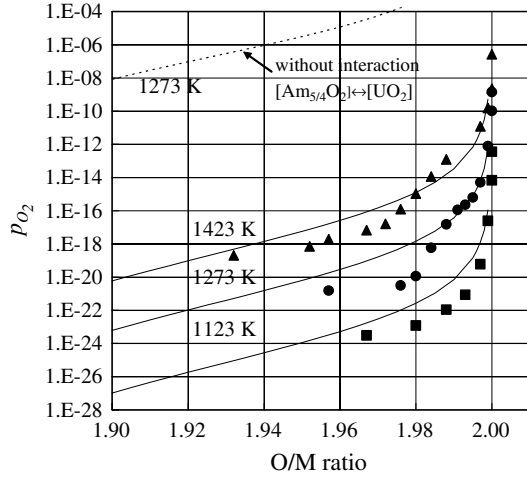


Fig. 2. Analytical oxygen potential isotherms obtained by a chemical thermodynamic model for $(U_{0.685}Pu_{0.270}Am_{0.045})O_{2-x}$.

discontinuities are seen in the isotherms of $(U_{0.685}Pu_{0.270}Am_{0.045})O_{2-x}$ at 1423 K and 1273 K between $O/M = 1.97$ and 1.99 , as shown in Fig. 2. However, these discontinuities, namely sharp changes of slopes of $\log(p_{O_2})$ versus $\log(x)$, should not result from the complete reduction of Am to its trivalent state, since no discontinuities are seen at 1123 K. The slopes give information about defect structures of non-stoichiometric compounds and the changes of slopes observed in $(U, Pu, Am)O_{2-x}$ are those inherent in $(U, Pu, Am)O_{2-x}$ as discussed in Ref. [4]. In brief, the slopes in the relevant O/M regions at 1423 K and 1273 K are more than 10, which should not be due to a disordered defect or defect cluster but to an ordered intermediate phase, since such high slopes ($n > 10$) cannot be seen by the disordered defect or defect cluster [19]. Besides, these changes of slopes appear around $O/M = 1.985$ [4]. This O/M value is more than $O/M = 1.9775$, which is the upper limit that all Am are reduced to trivalent states. From these facts, the priority of AmO_{2-x} reduction in $(U, Pu, Am)O_{2-x}$ does not seem to occur. The same reduction rate of AmO_{2-x} and PuO_{2-x} in $(U, Pu, Am)O_{2-x}$ was, therefore, assumed in this study. This hypothesis seems to be reasonable to some degree as shown in Woodley's study [6] on $(U_{0.750}Pu_{0.225}Ce_{0.025})O_{2-x}$, which showed smooth isotherm curves and had somewhat higher oxygen potentials than those of $(U_{0.75}Pu_{0.25})O_{2-x}$.

For the representation of oxygen potential of $(U, Pu, Am)O_{2-x}$, an equation for Gibbs energy of formation of $(U_{0.685}Pu_{0.270}Am_{0.045})O_{2-x}$ with a constant energy of interaction is given below. The chemical species considered in the present model are $[UO_2]$, $[PuO_2]$, $[Pu_{4/3}O_2]$, $[AmO_2]$ and $[Am_{5/4}O_2]$.

$$\begin{aligned} \Delta G_f^0 \langle (U_{1-z-y}Pu_yAm_z)O_w \rangle &= \frac{m_{UO_2}}{m} \Delta G_f^0 \langle UO_2 \rangle + \frac{m_{PuO_2}}{m} \Delta G_f^0 \langle PuO_2 \rangle \\ &+ \frac{m_{Pu_{4/3}O_2}}{m} \Delta G_f^0 \langle Pu_{4/3}O_2 \rangle + \frac{m_{Am_{5/4}O_2}}{m} \Delta G_f^0 \langle Am_{5/4}O_2 \rangle \\ &+ \frac{m_{AmO_2}}{m} \Delta G_f^0 \langle AmO_2 \rangle + \frac{m_{UO_2}}{m} RT \ln \left[\frac{m_{UO_2}}{m} \right] \\ &+ \frac{m_{PuO_2}}{m} RT \ln \left[\frac{m_{PuO_2}}{m} \right] + \frac{m_{Pu_{4/3}O_2}}{m} RT \ln \left[\frac{m_{Pu_{4/3}O_2}}{m} \right] \\ &+ \frac{m_{AmO_2}}{m} RT \ln \left[\frac{m_{AmO_2}}{m} \right] + \frac{m_{Am_{5/4}O_2}}{m} RT \ln \left[\frac{m_{Am_{5/4}O_2}}{m} \right] \\ &+ \frac{m_{AmO_2} \cdot m_{Am_{5/4}O_2}}{m} E_a + \frac{m_{PuO_2} \cdot m_{Pu_{4/3}O_2}}{m^2} E_b, \quad (1) \end{aligned}$$

where $\Delta G_f^0 \langle i \rangle^1$ is standard Gibbs energy of formation for species i , m_i is mole of species i , m is the sum of the moles of all the species, R is the ideal gas constant, T is absolute temperature. E_a and E_b are the interaction energies for $[AmO_2] \leftrightarrow [Am_{5/4}O_2]$ and $[PuO_2] \leftrightarrow [Pu_{4/3}O_2]$, respectively, and expressed as the form $\Delta H_E - T\Delta S_E$. These interaction energies have already been obtained by the analyses of isotherms of PuO_{2-x} [13] and AmO_{2-x} [16]. Interactions between U-oxides and Pu-oxides are not considered because the oxygen potential isotherms of MOX could be represented well without these interactions [13], which mean that these interactions are small or negligible. From the mass balance of each species, we obtain the following.

$$m_{AmO_2} = z - \frac{5}{4}m_{Am_{5/4}O_2}, \quad (2)$$

$$m_{PuO_2} = y - \frac{4}{3}m_{Pu_{4/3}O_2}, \quad (3)$$

$$m_{UO_2} = 1 - y - z, \quad (4)$$

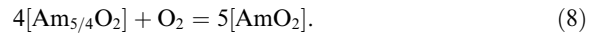
$$m_{Pu_{4/3}O_2} = 3 - \frac{3}{4}m_{Am_{5/4}O_2} - \frac{3}{2}w. \quad (5)$$

The partial molar Gibbs energy for each species is obtained by differentiating $m \cdot \Delta G_f^0 \langle (U_{1-z-y}Pu_yAm_z)O_w \rangle$ by m_i . For the Am-oxide species

$$\begin{aligned} \Delta \bar{G}[AmO_2] &= \Delta G_f^0 \langle AmO_2 \rangle + RT \ln \left[\frac{m_{AmO_2}}{m} \right] \\ &+ \frac{m_{Am_{5/4}O_2}(m - m_{AmO_2})}{m^2} E_a - \frac{m_{Pu_{4/3}O_2} \cdot m_{PuO_2}}{m^2} E_b \quad (6) \end{aligned}$$

$$\begin{aligned} \Delta \bar{G}[Am_{5/4}O_2] &= \Delta G_f^0 \langle Am_{5/4}O_2 \rangle + RT \ln \left[\frac{m_{Am_{5/4}O_2}}{m} \right] \\ &+ \frac{m_{AmO_2}(m - m_{Am_{5/4}O_2})}{m^2} E_a - \frac{m_{Pu_{4/3}O_2} \cdot m_{PuO_2}}{m^2} E_b. \quad (7) \end{aligned}$$

In the Am–O system, the following reaction has been assumed [16].



The equilibrium partial molar Gibbs energy relation for reaction (8) can be written as

¹ The notation $\langle \rangle$ solid phase; $[\]$ solid solution component is used throughout.

$$RT \ln(p_{O_2}) = 5\Delta\bar{G}[\text{AmO}_2] - 4\Delta\bar{G}[\text{Am}_{5/4}\text{O}_2], \quad (9)$$

where p_{O_2} is the value of oxygen partial pressure, which is derived from the ratio of oxygen partial pressure in atmosphere to its standard state, 0.101 MPa. Introducing Eqs. (6) and (7) into (9), we obtain the following equation.

$$\begin{aligned} RT \ln(p_{O_2}) &= 5\Delta G_f^0 \langle \text{AmO}_2 \rangle - 4\Delta G_f^0 \langle \text{Am}_{5/4}\text{O}_2 \rangle \\ &+ RT \ln \left[\frac{m_{\text{AmO}_2}^5}{m_{\text{Am}_{5/4}\text{O}_2}^4 \cdot m} \right] + \left\{ m(5 \cdot m_{\text{Am}_{5/4}\text{O}_2} - 4 \cdot m_{\text{AmO}_2}) \right. \\ &\left. - m_{\text{Am}_{5/4}\text{O}_2} \cdot m_{\text{AmO}_2} \right\} \frac{E_a}{m^2} - m_{\text{Pu}_{4/3}\text{O}_2} \cdot m_{\text{PuO}_2} \frac{E_b}{m^2}. \quad (10) \end{aligned}$$

The oxygen potential isotherms for $(\text{U}_{0.685}\text{Pu}_{0.270}\text{Am}_{0.045})\text{O}_{2-x}$ are expressed by solving the non-linear Eq. (10) with a numerical analysis technique such as the Newton–Raphson method. Fig. 2 compares analytical isotherms with those of experimental for $(\text{U}_{0.685}\text{Pu}_{0.270}\text{Am}_{0.045})\text{O}_{2-x}$. As shown in the figure, the analytical isotherm without interactions (indicated as ‘without interaction $[\text{Am}_{5/4}\text{O}_2] \leftrightarrow [\text{UO}_2]$ ’ in the figure) is much higher than that of experimental one, although the interactions $[\text{AmO}_2] \leftrightarrow [\text{Am}_{5/4}\text{O}_2]$ and $[\text{PuO}_2] \leftrightarrow [\text{Pu}_{4/3}\text{O}_2]$ were considered. This suggests that the interaction between Am-oxide and U-oxide or Pu-oxide is important. In this study, an interaction between Am-oxide and U-oxide is assumed to be the main source to reduce the oxygen potential of AmO_{2-x} when AmO_{2-x} is solved in $(\text{U}, \text{Pu})\text{O}_{2-x}$, since the oxygen potential of AmO_{2-x} is greatly reduced by forming a solid solution with UO_2 , as shown in Fig. 1. Thus, the interaction between $[\text{Am}_{5/4}\text{O}_2]$ and $[\text{UO}_2]$ is newly introduced in this study and it was estimated by the following procedure.

First, a representation of oxygen potentials for $(\text{Am}_{0.5}\text{U}_{0.5})\text{O}_{2-x}$ solid solution was carried out by using the present model considering chemical species of $[\text{Am}_{5/4}\text{O}_2]$, $[\text{AmO}_2]$ and $[\text{UO}_2]$ and the interactions of $[\text{AmO}_2] \leftrightarrow [\text{Am}_{5/4}\text{O}_2]$ and $[\text{Am}_{5/4}\text{O}_2] \leftrightarrow [\text{UO}_2]$. The interaction $[\text{Am}_{5/4}\text{O}_2] \leftrightarrow [\text{UO}_2]$ with energy E_c was obtained by a least square fitting of analytical isotherms to the experimental ones at five different temperatures.

Table 1 summarizes interaction energy values considered in this study. After the introduction of the interac-

Table 1

Interaction energies obtained by analyses of experimental oxygen potentials

	ΔH_E (J mol ⁻¹)	ΔS_E (J K ⁻¹ mol ⁻¹)
E_a : $[\text{AmO}_2] \leftrightarrow [\text{Am}_{5/4}\text{O}_2]$ [16]	87,573 ± 2596	59.3 ± 0.2
E_b : $[\text{PuO}_2] \leftrightarrow [\text{Pu}_{4/3}\text{O}_2]$ [13]	63,470 ± 2760	49.36 ± 2.15
E_c : $[\text{Am}_{5/4}\text{O}_2] \leftrightarrow [\text{UO}_2]$	172,958 ± 8526	36.1 ± 1.8
E_d : $[\text{Am}_{5/4}\text{O}_2] \leftrightarrow [\text{PuO}_2]$	86,887 ± 18,324	54.4 ± 13.4

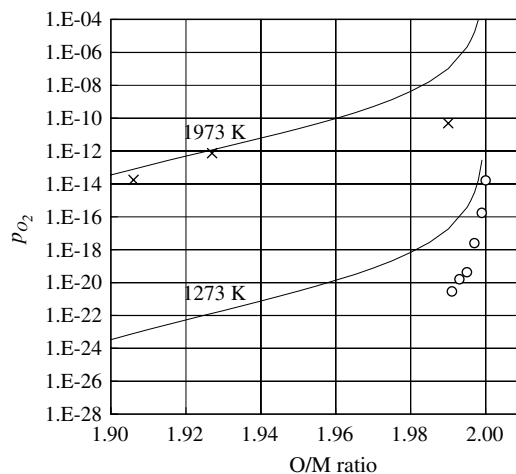


Fig. 3. Analytical oxygen potential isotherms obtained by a chemical thermodynamic model for $(\text{U}_{0.684}\text{Pu}_{0.300}\text{Am}_{0.016})\text{O}_{2-x}$.

tion energy E_c , the analytical isotherm of oxygen potentials for $(\text{U}_{0.685}\text{Pu}_{0.270}\text{Am}_{0.045})\text{O}_{2-x}$ expresses the experimental one very well, as shown in Fig. 2. It implies that the oxygen potentials of AmO_{2-x} are reduced significantly by solving it into UO_2 solvent and this interaction is predominant, compared to those between Am and Pu oxides. This result is also consistent with the assumption mentioned above. As for the case of $(\text{U}_{0.684}\text{Pu}_{0.300}\text{Am}_{0.016})\text{O}_{2-x}$, which contains low Am content, the agreement between analytical and experimental isotherms is good at 1973 K, while the analytical isotherm overestimate the experimental one at 1273 K, as shown in Fig. 3.

4. Discussion

As already noted in Figs. 2 and 3, the analytical isotherms showed good agreement with the experimental ones, but slight overestimation could be seen especially at the lower temperature. Two reasons for this deviation were considered. One was that the reduction of AmO_{2-x} and PuO_{2-x} in $(\text{U}, \text{Pu}, \text{Am})\text{O}_{2-x}$ does not occur congruently as assumed in the present model, and the other was that there may be an interaction between Am-oxide and Pu-oxide, which was not considered in the present model. No information has been reported on the reduction aspect of Am and Pu oxide in $(\text{U}, \text{Pu}, \text{Am})\text{O}_{2-x}$, and thus, an interaction between $[\text{Am}_{5/4}\text{O}_2] \leftrightarrow [\text{PuO}_2]$ with energy E_d was considered. Eq. (1) was modified by adding the corresponding term $m_{\text{Am}_{5/4}\text{O}_2} \cdot m_{\text{PuO}_2} \cdot E_d/m$ and then a modified version of Eq. (10) was obtained. E_d was then determined by a least square fitting to the experimental data and shown in Table 1. Fig. 4 compares analytical oxygen potential isotherms with those of experimental

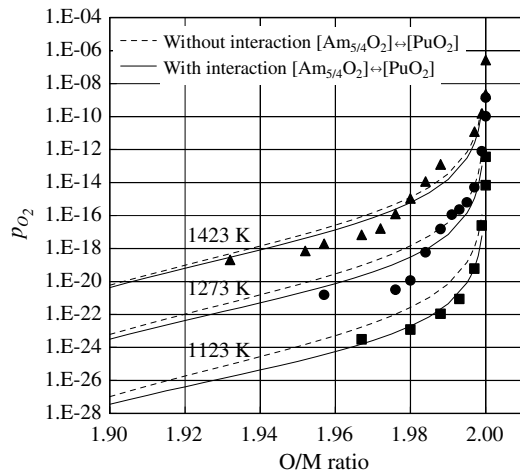


Fig. 4. Effect of an interaction between $\text{Am}_{5/4}\text{O}_2$ and PuO_2 on the analytical oxygen potential isotherms for $(\text{U}_{0.685}\text{Pu}_{0.270}\text{Am}_{0.045})\text{O}_{2-x}$.

one. The isotherm at 1423 K changed slightly with introduction of the $[\text{Am}_{5/4}\text{O}_2] \leftrightarrow [\text{PuO}_2]$ interaction, while those at 1273 K and 1123 K were clearly lowered; good agreement between analytical and experimental isotherms can be seen.

The oxygen potential isotherms were expressed well by the present model that uses a pseudo-quaternary system, assuming the same reduction rate of Am and Pu oxide in $(\text{U}, \text{Pu}, \text{Am})\text{O}_{2-x}$ and the interaction between $[\text{Am}_{5/4}\text{O}_2] \leftrightarrow [\text{UO}_2]$. Further experimental investigations will be done to confirm appropriateness of the present model.

5. Conclusion

The oxygen potential isotherms of $(\text{U}, \text{Pu}, \text{Am})\text{O}_{2-x}$ were represented by a chemical thermodynamic model. It is assumed in the present model that $(\text{U}, \text{Pu}, \text{Am})\text{O}_{2-x}$ consisted of the chemical species $[\text{UO}_2]$, $[\text{PuO}_2]$, $[\text{Pu}_{4/3}\text{O}_2]$, $[\text{AmO}_2]$ and $[\text{Am}_{5/4}\text{O}_2]$ in a pseudo-quaternary system by treating the reduction rates of Pu and Am as identi-

cal. An interaction between $[\text{Am}_{5/4}\text{O}_2]$ and $[\text{UO}_2]$ was also introduced. The agreement between analytical and experimental isotherms was good.

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