

Letter to the Editor

Some Important Fuel Chemistry Aspects Related to the Use of MOX as a Fast Reactor Fuel

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Abstract—*With interest in sodium-cooled fast reactors showing a clear sign of renewal, we seek to address and discuss in this letter some important thermochemical aspects pertinent to mixed oxide (MOX) fuel: phase separation, vaporization, oxygen and plutonium redistribution, chemical interaction between fuel and clad, and chemical interaction between fuel and coolant.*

I. INTRODUCTION

We were motivated to prepare this letter for the following reasons: the choice of a mixed oxide fuel (MOX) ($M = U + Pu$) for the Prototype Fast Breeder Reactor (PFBR) in India,¹ the renewed interest in sodium-cooled fast reactors^{2,3} (SFRs), the reappearance of papers related to thermodynamic modeling for $(U,Pu)O_2$ oxide fuels⁴ as well as irradiated UO_2 fuels^{5,6} or to experimental determination of phase separation in high-plutonium MOX fuels,⁷ and the use of high-plutonium-content fuels with or without uranium to incinerate Pu and minor actinides^{8,9} or even as a possible nominal core of the Fast Breeder Test Reactor¹⁰ (FBTR) (India). The objective is to present in a condensed form some salient thermochemical information that became available due to intense research that was being conducted on MOX fuels prior to and during the 1980s. It is known that during reactor operation, initially homogeneous single-phase, MOX, $U_{(1-y)}Pu_yO_z$, could undergo many changes due to various factors such as temperature gradient across the fuel pellet, temperature recycling, reactions between fuel and fission products and also among the fission products, fission product–assisted chemical interactions with the cladding material, and chemical interaction between fuel and the coolant, too, if a breach were to occur in the clad. Two parameters, O/M (that is, z) and Pu/M (that is, y), control many aspects of fuel chemistry, some of which figure in this letter: MOX phase separation, vaporization behavior, O/M evolution and actinide distribution, and fuel-clad and fuel-coolant chemical interactions. Information from the proceedings of three major conferences^{11–13} and from many studies reported in different journals are presented and/or discussed briefly.

II. MOX PHASE SEPARATION

If any fracturing of the sintered oxide MOX occurs (due to a temperature cycling, for instance), the fuel performance could

be affected since it could perturb the redox processes.¹⁴ The conditions under which the MOX phase separation can occur were studied long ago by Markin and Street¹⁵ and Sari et al.,¹⁶ and recently by Truphémus et al.⁷ The essential findings of these studies are the following: MOX will remain single phase up to a value of $y \leq 0.2$, if $z \geq 1.90$ (Ref. 16); disproportionation of hypostoichiometric MOX (with $y = 0.42$) could occur upon cooling, resulting in generation of $MO_{2.00}$ and a hypostoichiometric phase with $z = 1790 \pm 0.005$ (Ref. 15); phase transformation temperature and the O/M difference between the two phases increase with increase in the value of y (Ref. 7). In brief, two factors are of relevance to two-phase formation: one, the relative compositions of the two phases, and the other, the volume expansion. If the two phases were to be in equilibrium, their compositions ($z = O/M$ and $y = Pu/M$) must be such that the oxygen potentials are the same in both phases. Consequently, since no hypostoichiometric MOX phase can have an oxygen potential equivalent to a stoichiometric MOX phase, both phases might be hypostoichiometric, and the more oxygen-deficient phase should be richer in plutonium. The lattice parameter decreases with decrease in temperature within the single-phase region until the two-phase region is reached. In the two-phase region, the lattice parameter of the more-oxygen-deficient phase is higher than that of single phase, but the lattice parameter of the less-oxygen-deficient phase is lower than that of the single phase. Thus, the relative amounts of each phase may determine whether there will be a volume expansion of disproportion.

III. VAPOR PRESSURE AND VAPORIZATION BEHAVIOR

The trivariant nature and presence of different $(U+O)$ and $(Pu+O)$ species in the vapor phase of MOX make the vaporization chemistry of the ternary system $U_{(1-y)}Pu_yO_z$ complex and interesting, as revealed by mass spectrometric measurements by Battles et al.¹⁷ and Ohse and Olson¹⁸ and by calculations by Rand and Markin,¹⁹ Tetenbaum²⁰ and by us.²¹ Table I gives information such as total vapor pressures (sum of partial pressures of all vapor species) and O/M and Pu/M ratios of the vapor phase for different values of y and z in the condensed MOX. Given that the primary input for the results in Table I is the oxygen potential, $\Delta\mu(O_2) = [R \cdot T \cdot \ln p(O_2)]$, it is useful to mention that the values of $\Delta\mu(O_2)$ (Ref. 22) used by us in these calculations²¹ deviate by no more than 12% from those deducible with the most recent model proposed by Guéneau et al.⁴ Even this deviation was seen only at the extreme O/M and/or extreme Pu/M.

From Table I, one infers that the effect of O/M is more prominent than that of Pu/M on vapor pressures. The vapor pressure minimum (VPM) is observed for each y at $\approx 2 \times 10^{-7}$ atm. The VPM, QCVC1 (quasi-congruently vaporizing composition with respect to O/M), and QCVC2 (quasi-congruently vaporizing composition with respect to Pu/M) all occur at progressively decreasing O/M values with increasing y . The O/M corresponding to these three stationary points decrease in this order: QCVC2 > VPM > QCVC1. At O/M < QCVC1, the vapor phase will have more Pu than MOX, and consequently, MOX will get enriched in U.

At O/M > QCVC2, the vapor phase will have more U than MOX, and consequently, MOX will get enriched in Pu. Therefore, the optimum value of O/M for MOX will be between QCVC1 and QCVC2, if the criterion set is to minimize vapor transport assisted actinide redistribution.

IV. OXYGEN REDISTRIBUTION

The large temperature gradient that will develop across the radius of a MOX fuel pin during irradiation would

TABLE I
Information Pertinent to Vaporization Behavior of MOX

z	$y = 0.15$	$y = 0.20$	$y = 0.25$	$y = 0.30$	$y = 0.35$	$y = 0.40$	$y = 0.45$	$y = 0.50$
Total Vapor Pressures for $U_{(1-y)}Pu_yO_z$ (in atm)								
1.91		$9.8 \cdot 10^{-7}$	$3.5 \cdot 10^{-7}$	$2.5 \cdot 10^{-7}$	$2.3 \cdot 10^{-7}$	$2.4 \cdot 10^{-7}$	$2.5 \cdot 10^{-7}$	$2.8 \cdot 10^{-7}$
1.92		$4.8 \cdot 10^{-7}$	$2.7 \cdot 10^{-7}$	$2.3 \cdot 10^{-7}$	$2.3 \cdot 10^{-7}$	$2.5 \cdot 10^{-7}$	$2.7 \cdot 10^{-7}$	$3.0 \cdot 10^{-7}$
1.93	$1.3 \cdot 10^{-6}$	$3.1 \cdot 10^{-7}$	$2.3 \cdot 10^{-7}$	$2.2 \cdot 10^{-7}$	$2.4 \cdot 10^{-7}$	$2.7 \cdot 10^{-7}$	$3.0 \cdot 10^{-7}$	$3.4 \cdot 10^{-7}$
1.94	$4.4 \cdot 10^{-7}$	$2.4 \cdot 10^{-7}$	$2.1 \cdot 10^{-7}$	$2.3 \cdot 10^{-7}$	$2.6 \cdot 10^{-7}$	$3.0 \cdot 10^{-7}$	$3.5 \cdot 10^{-7}$	$3.9 \cdot 10^{-7}$
1.95	$2.6 \cdot 10^{-7}$	$2.0 \cdot 10^{-7}$	$2.2 \cdot 10^{-7}$	$2.6 \cdot 10^{-7}$	$3.1 \cdot 10^{-7}$	$3.6 \cdot 10^{-7}$	$4.2 \cdot 10^{-7}$	$4.7 \cdot 10^{-7}$
1.96	$2.0 \cdot 10^{-7}$	$2.1 \cdot 10^{-7}$	$2.5 \cdot 10^{-7}$	$3.1 \cdot 10^{-7}$	$3.7 \cdot 10^{-7}$	$4.5 \cdot 10^{-7}$	$5.2 \cdot 10^{-7}$	$5.9 \cdot 10^{-7}$
1.97	$1.9 \cdot 10^{-7}$	$2.4 \cdot 10^{-7}$	$3.1 \cdot 10^{-7}$	$4.0 \cdot 10^{-7}$	$5.0 \cdot 10^{-7}$	$6.0 \cdot 10^{-7}$	$7.0 \cdot 10^{-7}$	$7.9 \cdot 10^{-7}$
1.98	$2.3 \cdot 10^{-7}$	$3.3 \cdot 10^{-7}$	$4.5 \cdot 10^{-7}$	$5.9 \cdot 10^{-7}$	$7.4 \cdot 10^{-7}$	$8.9 \cdot 10^{-7}$	$1.0 \cdot 10^{-6}$	$1.2 \cdot 10^{-6}$
1.99	$4.0 \cdot 10^{-7}$	$6.1 \cdot 10^{-7}$	$8.7 \cdot 10^{-7}$	$1.1 \cdot 10^{-6}$	$1.5 \cdot 10^{-6}$	$1.8 \cdot 10^{-6}$	$2.1 \cdot 10^{-6}$	$2.4 \cdot 10^{-6}$
2.00	$2.1 \cdot 10^{-6}$	$3.1 \cdot 10^{-6}$	$4.3 \cdot 10^{-6}$	$5.6 \cdot 10^{-6}$	$7.2 \cdot 10^{-6}$	$9.2 \cdot 10^{-6}$	$1.2 \cdot 10^{-5}$	$1.5 \cdot 10^{-5}$
VPM ^a	1.9669	1.9543	1.9415	1.9287	1.9160	1.9034	1.8912	1.8795
Vapor Phase O/M Values for $U_{(1-y)}Pu_yO_z$								
1.91		1.10	1.37	1.65	1.89	2.08	2.23	2.33
1.92		1.24	1.54	1.82	2.05	2.22	2.34	2.43
1.93	1.06	1.42	1.74	2.00	2.20	2.34	2.45	2.52
1.94	1.26	1.64	1.95	2.18	2.34	2.46	2.55	2.61
1.95	1.49	1.88	2.15	2.34	2.48	2.57	2.64	2.69
1.96	1.78	2.12	2.34	2.50	2.60	2.68	2.73	2.77
1.97	2.07	2.34	2.52	2.64	2.72	2.77	2.81	2.84
1.98	2.36	2.57	2.70	2.78	2.83	2.87	2.89	2.91
1.99	2.67	2.79	2.86	2.91	2.94	2.96	2.98	2.99
2.00	2.97	3.00	3.02	3.04	3.06	3.09	3.13	3.18
QCVC1 ^b	1.9665	1.9532	1.9395	1.9255	1.9113	1.8970	1.8825	1.8679
Vapor Phase Pu/M Values for $U_{(1-y)}Pu_yO_z$								
1.91		0.90	0.72	0.58	0.48	0.42	0.38	0.36
1.92		0.79	0.61	0.49	0.41	0.36	0.33	0.31
1.93	0.92	0.67	0.50	0.39	0.33	0.30	0.28	0.27
1.94	0.77	0.52	0.39	0.31	0.27	0.25	0.24	0.23
1.95	0.59	0.38	0.29	0.25	0.22	0.20	0.19	0.19
1.96	0.40	0.27	0.22	0.19	0.17	0.16	0.16	0.15
1.97	0.24	0.18	0.16	0.14	0.13	0.12	0.12	0.12
1.98	0.15	0.12	0.10	0.094	0.087	0.083	0.082	0.082
1.99	0.076	0.063	0.056	0.052	0.049	0.047	0.046	0.045
2.00	0.016	0.014	0.013	0.012	0.011	0.0099	0.0089	0.0080
QCVC2 ^c	1.9798	1.9679	1.9552	1.9416	1.9227	1.9134	1.8987	1.8835

^aValue of z where the VPM is observed for each y .

^bValue of z where the O/M of the vapor phase will be the same as that in MOX.

^cValue of z where the Pu/M of the vapor phase will be the same as that in MOX.

cause redistribution of oxygen resulting in O/M and $\Delta\mu(\text{O}_2)$ gradients.

IV.A. O/M Evolution

The essential findings from the calculations performed by Rand and Markin¹⁹ for six values of z (the results shown as a figure by Rand and Markin and also by Johnson et al.²³) are that there would be negligible O/M evolution if the initial O/M ≈ 2 . For other values of z , the center would become relatively more hypostoichiometric if the initial O/M < 2 and relatively more hyperstoichiometric if the initial O/M > 2 . The surface, however, would tend to be at an OM of ~ 2 , if the initial O/M is from 1.98 to 2.02 and at ~ 1.99 if the initial O/M is 1.96.

In their classic paper, Johnson et al.²³ also gave the results of calculation of the rate of change in O/M with burnup. The following inference emerges: When $y = 0.25$ and $z = 1.95$ to 2.00, the rate of increase in mean O/M initially would be about 0.006 O/M unit per at. % burnup, and after 4 to 6 at. % burnup, the increase rate drops to 0.001 O/M unit per at. % burnup. The measurements by Davies and Ewart²⁴ indicate that after ~ 10 at. % burnup, the O/M increased by $\sim 0.05 \pm 0.01$ unit. Measurements by Ewart et al.²⁵ on two pins with differing initial O/M values show that while the O/M for one pin increased from 1.948 to 1.992 (i.e., an increase of $+0.044$ O/M unit), the O/M of the other pin increased from 1.976 to only 1.997 against an expected value of 2.005 on the basis of the results of the first pin. The smaller increase in the case of the latter was attributed to the formation of some unknown oxide phase acting as buffer. Also, only the second pin showed some evidence of clad attack. Conte and Marcon²⁶ summarized the Phénix experience with 18% and 25% PuO₂ fuel pins such that there was no tendency to increase O/M to 2.00, but instead to a global value of 1.985, even when the initial O/M was 1.995. Conte and Marcon opined that different pins will reach a similar O/M after different burnup values, and thus, only until this period will the initial O/M influence the clad attack.

IV.B. Oxygen Potential Gradient

The O/M evolution will result in $\Delta\mu(\text{O}_2)$ gradient as well. With increasing burnup, however, since the O/M itself would increase, $\Delta\mu(\text{O}_2)$ of an irradiated fuel will be increasing at every location. Additionally, the dissolution of fission products (as oxides in the MOX fluorite lattice) would increase the mean valence of Pu and U and thus effectively cause further increase in $\Delta\mu(\text{O}_2)$. Johnson et al.²³ gave results of their calculations in the form of a figure, $\Delta\mu(\text{O}_2)$ versus O/M, at 2000 K for MOX with 20% PuO₂ at 0, 2, 4, 6, 8, and 10 at. % burnup, with all fissions assumed to occur in ²³⁹Pu. The increase in $\Delta\mu(\text{O}_2)$ is from about $-460 \text{ kJ}\cdot\text{mol}^{-1}$ (O/M = 1.95 at zero at. % burnup) to $-250 \text{ kJ}\cdot\text{mol}^{-1}$ (after 10 at. % burnup). The latter value would correspond to O/M > 2.00 for an unirradiated fuel.

V. PLUTONIUM REDISTRIBUTION

The O/M evolution and associated changes in vaporization behavior would result in actinide redistribution as well.

Bober et al.²⁷ investigated the effects of redistribution with cylindrical MOX specimens having 15% PuO₂ and differing O/M values. Practically no redistribution was noticed in the case of the specimen with initial O/M between 1.96 and 1.97. This O/M window is only slightly on the lower side compared to the window set by the values of QCVC1 and QCVC2 given in Table I (~ 1.97 and ~ 1.98 , respectively). A subsequent paper by Bober et al.²⁸ gave results of autoradiographs of cross sections of five different fuel pins, subjected to fast flux in the RAPSODIE reactor, which indicated preferential evaporation of uranium at slightly hypostoichiometric or stoichiometric compositions and preferential evaporation of plutonium oxides at highly hypostoichiometric compositions. The calculations shown in Ref. 28 (for MOX with 20% PuO₂ and O/M = 2.00) revealed that (a) the composition at the fuel center can become 28 mol % PuO₂ (at the rating of 500 W/cm) and 36 mol % PuO₂ (at 600 W/cm) and (b) the center temperature can increase by 100°C (500 W/cm) and 250°C (600 W/cm). Bober et al. predicted that if local power ratings were to go above 600 W/cm, then melting could occur, and consequently, plutonium-rich molten fuel (highly enriched fissile material) could collect at the bottom of the central cavity. Based on these inferences, their choice of O/M for 20 mol % PuO₂ fuel is between 1.96 and 1.97, if the redistribution effects were to be minimized. The QCVCs for 20 mol % PuO₂ as given in Table I at $T = 2000 \text{ K}$ (QCVC1 = 1.953 and QCVC2 = 1.968) are in accord with Bober et al.'s recommendation of using hypostoichiometric fuels with plutonium valencies of 3.6 to 3.7.

Lackey et al.²⁹ showed the result from a low-burnup MOX pin (0.7% burnup; 15% Pu; O/M = 2.00), which indicated clear Pu enrichment at the central void to 21%, attributed to preferential vaporization of uranium. Ishii and Asaga³⁰ performed calculations of radial Pu distribution and showed the electron probe results on two MOX fuels to validate that dependence of Pu migration on initial O/M: The percentage of Pu at the center increased to (a) 38% in the case of a fuel pin with initial O/M = 1.99, initial Pu = 27 wt%, burnup = 106 GWd/ton; (b) 34% in the case of a fuel pin with initial O/M = 1.955, initial Pu = 30 wt%, burnup = 110 GWd/ton.

There were however incidences of Pu redistribution that are not consistent with the above observations or predictions. Lawrence et al.,³¹ for instance, reported plutonium concentration on the outer low-temperature region of the fuel after a peak burnup of 12.5 at. % in the case of 25% Pu and O/M = 1.96. Wegen⁹ observed in the case of a failed fuel pin (initial Pu = 45% and initial O/M = 1.96) an increasing Pu content from the center to the periphery, with the central hole having substantially below the original 45% Pu, contrary to the expected Pu concentration. A possible explanation we can think of in the latter case is that operation under failed condition would have changed the O/M profile, which would have facilitated preferential evaporation of Pu from the center.

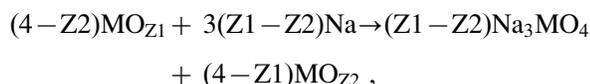
VI. FUEL-CLADDING CHEMICAL INTERACTION

It is not only oxidation of the clad that one needs to deal with during reactor operation but also the possibility of chemical attack of the fuel-clad by reactive fission products such as cesium, tellurium, and iodine. In our recent paper,³² we have discussed the role of the four key elements in fuel-cladding

chemical interaction (FCCI) and also the various clad wastage correlations available in literature. Reference 32 also describes the following in great detail: (a) selection of one of the models³³ for MOX with initial O/M < 1.98, (b) proposal of our own model for O/M > 1.98, and (c) proposal of multiplication factors for use (in conjunction with both these models) for high-plutonium-content fuels. Accordingly, we would summarize the relevant equations in Table II and merely state that from the point of view of clad corrosion, a large number of studies^{26,34-48} provide evidence in support of initial O/M < 2, while the results from a few studies^{49,50} seem to favor or reconcile with O/M = 2. The effects of high Pu on FCCI are reported in a few references^{40,51} while information on cladding component chemical transport into the fuel matrix can be found in a few studies.^{36,52,53}

VII. FUEL-COOLANT CHEMICAL INTERACTION

Occurrence of a breach in the fuel cladding will make the fuel chemistry more complex than it is already. The following reaction occurs when hot sodium comes into contact with MOX:



where Z1 is the initial O/M of the fuel and Z2 is the O/M of the fuel when the reaction is complete or stopped. The reaction written in this way implies that the U/Pu ratio is assumed to be the same in MO_{Z1}, Na₃MO₄, and MO_{Z2}. This assumption gains support from the results of Blackburn et al.,⁵⁴ who performed electron microprobe analysis of the product of the reaction between U_{0.8}Pu_{0.2}O₂ powder with Na₂O (sometimes with excess Na) in sealed Ni capsules for up to 3 days at different temperatures from 1123 to 1273 K. Blackburn et al.⁵⁴

determined Z2, the threshold O/M value for fuel-sodium reaction, from the weight increase for the pellets, allowed to react with sodium containing only 4 ppm of oxygen. The value of Z2 was 1.95 when Z1 = 1.995. Blackburn et al. have also estimated Z2 taking it as the value, the oxygen potential corresponding to which equals that over (Na + MO₂ + Na₃MO₄). The lowest value reported yet for Z2 was 1.92 (Ref. 54) in the case of U_{0.8}Pu_{0.2}O₂, the value recalculated by Blackburn et al. from the data of thermal gradient experiments of Aitken and Evans (Ref. 2 in Ref. 54). The value of Z2 will depend on the Pu content of MOX and the temperature (heat rating): The higher the Pu content and the higher the temperature are, the lower will be Z2. It is clear that to prevent fuel-sodium reaction, the O/M of the fuel surface should be below 1.95. To achieve this, the mean O/M of the fuel should be 1.91. Thus, if one takes into account that with burnup, the mean O/M of the fuel will increase, it is just not practicable that one can prevent fuel-sodium reaction if a breach occurs. Adamson and Aitken⁵⁵ reported a threshold O/M of 1.96 for the reaction of sodium with exposed fuel to occur. Caputi et al.⁵⁶ have stated that when $x < 0.04$ in U_{0.75}Pu_{0.25}O_{2-x}, reaction between sodium and fuel will not occur. To have O/M < 1.96 at the surface, the interior fuel stoichiometry should be far lower, that is, ~1.91.

The fuel-sodium reaction product, Na₃MO₄, is a gray phase with a theoretical density of ~5.6 g/cm³, nearly half that of MOX. It has low thermal conductivity, too. These two factors have many implications, most important being the stress that will be imparted on the clad due to formation of the voluminous reaction product, and a steeper temperature gradient that will develop across the fuel radius. Many studies exist in literature that deal with kinetics or extent of swelling under defected conditions, some purposely effected.⁵⁷⁻⁶⁷

One other consequence of fuel-coolant chemical interaction when the fuel is operated after fuel failure is⁵⁹ that the O/M profile will change to an average O/M of 1.90 (at which

TABLE II

Clad Wastage Correlations Depicting the Dependence of O/M and PuO₂%*

Equation Number	Equation	Condition
(1)	$d/\mu\text{m} = (0.507 \cdot [B/(\text{at. \% fission})] \cdot (T/K - 705) \cdot [(O/M)_i - 1.935]) + 20.5$	For O/M ≤ 1.98
(2)	$d/\mu\text{m} = [B/(\text{at. \% fission})] \cdot [(T/K) - 800]^{0.5} \cdot [(O/M)_f - 1.94] \cdot [P/(W \cdot \text{cm}^{-1})]^{0.5}$	For O/M ≥ 1.98
(3)	Multiplication factor y for use along with either of the above equations. $d[\text{Eq. (3)}] = d[\text{Eq. (1) or (2)}] \cdot y$. $x = \text{PuO}_2$ fraction = from 0.25 to 0.70.	
(3a)	$y = 17.420 \cdot x^3 - 30.354 \cdot x^2 + 18.082 \cdot x - 1.8873$; $R^2 = 0.9981$	O/M = 1.97
(3b)	$y = 15.664 \cdot x^3 - 27.231 \cdot x^2 + 16.185 \cdot x - 1.5824$; $R^2 = 0.9987$	O/M = 1.98
(3c)	$y = 13.427 \cdot x^3 - 23.587 \cdot x^2 + 14.177 \cdot x - 1.2744$; $R^2 = 0.9985$	O/M = 1.99
(3d)	$y = 13.489 \cdot x^3 - 23.282 \cdot x^2 + 13.883 \cdot x - 1.2192$; $R^2 = 0.9985$	O/M = 2.00

*Equations (1) and (2) are taken from Ref. 32. Equations (3a) through (3d) are deduced from the values of y given in Ref. 32.

fuel-sodium reaction will stop) and that the center of the fuel could lose plutonium preferentially by evaporation.

VIII. CONCLUSION

A few important fuel chemistry aspects were considered in this letter such that it could serve as a kind of a minicompilation, refresh the understanding of MOX behavior and its performance as fast reactor fuel, and be a good starting point for new researchers.

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