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Pu, U, REDISTRIBUTION IN (U,Pu)O₂ FUELS BY TEMPERATURE GRADIENTS

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Pu, U REDISTRIBUTION IN (U,Pu)O₂ FUELS BY TEMPERATURE GRADIENTS[†]

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ABSTRACT

A predictive model of the time-dependent evolution of radial Pu/U + Pu gradients in operating nonmolten mixed oxide fuels is being developed. This model is based on preferential evaporation-condensation and vapor transport of metal-bearing species either along cracks and porosity channels within the fuel or inside closed pores migrating up the temperature gradient; equilibrium thermodynamics are applied to calculate the continuously changing vapor composition over the hot fuel. Predictions of the model are used in combination with recent in-pile experimental data (from nonmolten mixed oxide fuel pins irradiated in EBR II from 0.2 to about 11 at. % burnup at varying powers) to delineate the key parameters in radial actinide redistribution. A correlation between fuel stoichiometry (O to Pu ratio) and the degree of redistribution (described by an enrichment factor γ_{Pu}) has been identified; however, the degree of restructuring of the fuel — as determined by the peak linear power rating, time at this power, and the initial fuel density — also has a strong influence on the final value of γ_{Pu} . Certain kinetic features of the process and intended modifications to the present model are discussed, and evidence for axial actinide vapor transport in an irradiated annular fuel is also presented.

1 INTRODUCTION

Last year we critically reviewed the principal mechanisms (and variants) that have been proposed to account for radial actinide element redistribution in irradiated mixed oxide fuels and also outlined a possible analytical description of this phenomenon based on a preferential evaporation — condensation mechanism (i.e., vapor transport) [1]. This paper brings up to date our theoretical studies in this area and also incorporates some recent GE irradiated fuel pin data in a comparison between theoretical predictions and measured end-of-life radial Pu/U+Pu profiles at a variety of fuel power ratings, burnups, initial fuel densities and O/M ratios. In this way some of the key parameters characterizing radial and axial actinide redistribution have been delineated.

Our earlier conclusion that vapor transport would be the most important mechanism for actinide redistribution in fuels which do not undergo center melting [1] has since received convincing confirmation both from out-of-pile and in-pile experiments. Bober and co-workers at Karlsruhe [2] in some carefully designed

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out of pile axial temperature gradient tests essentially confirmed Rand and Markins' initial calculated cross over O/M value at 2400°C (i.e. the O/M value at which the vapor changes from U rich to Pu rich as O/M decreases 1.96 to 1.97). These workers also showed that the overall vapor transport rate in a temperature gradient of 2400° to 1900°C falls off as the overall fuel O/M drops from 2.00 to 1.96. Lackey et al. [3] found evidence in irradiated SPHEREPAC mixed oxide fuel pins for vapor deposited U rich deposits in cooler regions of the fuel (on the hot side of intact microspheres) and more recently Argonne workers [4] found evidence of radial vapor phase preferential evaporation - condensation effects along the interface between originally dish ended mixed oxide fuel pellets.

2 THEORETICAL (VAPOR TRANSPORT) MODEL

Our approach to developing an authentic model for actinide element redistribution in noncenter melted irradiated mixed oxide fuels has been to start with a simple yet realistic description of the basic phenomenon (i.e. U/Pu redistribution by preferential evaporation/condensation) and then to take account step by step of modifying factors such as solid state back diffusion and fuel restructuring (plus concomitant effects). In the basic fuel pin model, a transverse fuel section is divided into 30 concentric equal width annuli (radial segments) and each is assigned starting values of O/Pu ratio, temperature, mass of fuel and open pore volume. For initial calculations the radial temperature gradient and O/Pu gradients (see Figure 1) are assumed to be time independent (the heat of transport used to calculate the O/Pu gradients is ~ 10 kcal/mole [1]). Recent work at this laboratory indicates that use of this overall heat of transport represents an oversimplification; however, at this stage of development of the model errors incurred by its use are trivial. The initial equilibrium vapor pressures of the metal bearing species PuO_2 , PuO , Pu , UO_3 , UO_2 , UO and U above each fuel position are calculated using VAPRES, a FORTRAN IV computer program which has been written as a subroutine. From these equilibrium partial pressures the radial partial pressure gradient of each gas species is computed at time zero. At subsequent times new equilibrium partial pressures are calculated by application of Raoult's Law (partial pressure of species i = mole fraction of i in solid). The metal bearing species are allowed to flow (by molecular diffusion) between the fuel segments for a time interval δt (i.e. down the temperature and respective partial pressure gradients - the area available for vapor transport between consecutive fuel segments is assumed to be a fixed fraction of the open porosity assigned to those particular segments). The amount of each vapor species lost or gained by each radial segment in the time interval δt is determined by solving the appropriate diffusion equation, - which is Fick's Second Law expressed in cylindrical coordinates

$$D_i (\partial^2 C_i / \partial r^2 + 1/r \partial C_i / \partial r - \partial C_i / \partial t) = 0 \quad (1)$$

where C_i is the concentration of gaseous species i , r is the fuel radius, t is time and D_i is the simple kinetic theory diffusion coefficient for species i ($= 3/8 \sigma^2 n \sqrt{RT/(M_1 + M_2)}/2\pi M_1 M_2$ where the symbols have their usual meanings).

Equation (1) is solved numerically using the appropriate difference equations for $C(r + \delta t)$, etc. The total mass of each segment and the absolute amounts of U and Pu contained in the solid part of each segment are adjusted after each gas phase transfer interval (δt) in preparation for the next cycle of calculations; these values are either stored in the computer or printed out as they are calculated. One assumption implicit in this representation of vapor phase mass transport is that the rate of vapor transport of a particular species at a particular radial location is slower than its rates of evaporation or condensation - in other words at a particular location fuel and vapor are always assumed to be in thermodynamic equilibrium.

A flow diagram of the calculation sequence is shown in Figure 2. Initial input contains such information as fuel dimensions, initial O/M and Pu/U+Pu ratios, fuel density, linear power level, fuel surface temperature, etc. In its present form the computer program generates radial Pu/U+Pu profiles with the same general shape as those measured in irradiated fuels. Some of these calculated profiles are illustrated in Figure 3 except for one unique value of O/M the calculated degree of plutonium enrichment (or depletion - depending on overall O/M) at the fuel center increases continually with time until just before the mass of the innermost fuel segment(s) reaches zero, the fuel composition approaches pure plutonia (or uranium). Clearly this is unrealistic since available data from fuel pin irradiations (see below) show that (i) radial Pu/U+Pu gradients are established early in life (concomitant with restructuring), (ii) the (maximum) degree of U enrichment at the fuel center rarely exceeds $\sim 50\%$ and (iii) once established the radial Pu/U+Pu gradients are relatively insensitive to further irradiation. This

is taken as evidence that another process or combination of processes is operating which causes the system to attain or at least approach a steady state condition. Such processes are believed to be either back diffusion of uranium and plutonium ions through the solid phase (i.e. down the concentration gradients created by vapor transport) or a combination of ionic back diffusion and decreased vapor transfer rates as a result of fuel restructuring (i.e. both porosity and temperature decrease with time at a particular radial position). The next step in the development of our calculational model will be to include solid state (back) diffusion of U and Pu to see whether this process alone can bring about a steady state condition; an attempt will then be made to simulate fuel restructuring by allowing the open porosity in each radial segment to decrease as the segment gains mass through vapor transport. Inclusion of this last process should make preferential evaporation - condensation in migrating closed pores and vapor transport through open porosity equivalent. In this context it may be noted Lackey et al. have related actinide redistribution to restructuring kinetics by tying vapor phase fractionation of U and Pu to radial pore migration rates, [5] in this way a significant slowing down of the rate of actinide redistribution with time was obtained.

3 PREDICTIONS OF THE VAPOR TRANSPORT MODEL AND COMPARISONS WITH FUEL PIN DATA

3.1 EFFECT OF STOICHIOMETRY

A prediction of the present vapor transport model is that the degree of plutonium enrichment (or depletion) at the center of fuels operated at similar power levels (and hence with similar radial temperature gradients) will be a function of fuel stoichiometry. [1] According to equilibrium thermodynamic partial pressure calculations there will be no tendency for evaporative Pu enrichment or depletion at the hot center of fuel with $(\text{O/Pu})_{\text{center}} \approx 3.45$ and $T_c = 2500^\circ\text{C}$, in a radial temperature gradient of $2500 - 920^\circ\text{C}$ this corresponds to an overall fuel composition $\text{U}_{0.80}\text{Pu}_{0.20}\text{O}_{1.973}$ (assuming $\Delta H_v = 10$ kcal/mole). The calculated Pu/U+Pu profiles shown in Figure 3 illustrate that Pu enrichment is expected when the overall O/M > 1.973 and that Pu depletion should occur when O/M < 1.973 . In Figure 4 an attempt has been made to correlate the degree of Pu enrichment (or depletion) at the center of a number of high rated fuels ($T_c \approx 1500^\circ\text{C}$) with overall plutonium valency, using the calculated quasi-congruently evaporating composition as an anchor point. The degree of actinide redistribution has been expressed as a 'Pu enrichment factor', γ_{Pu} , which is simply the ratio of the Pu mole fraction measured at the inner surface (or center) of the fuel to that measured at the outer surface of the fuel at end of life. The U and Pu mole fractions were determined, via chemical concentrations, from measurements of the intensity of the uranium M_α and plutonium M_β characteristic x rays in a MAC-450 shielded electron microprobe analyzer (~ 1 micrometer diameter electron beam). Data were collected across the radii of transverse fuel cross sections (from at least twenty different fuel matrix locations) and reduced by MAGIC IV, a computer program for microprobe data reduction, [6] the values of the Pu mole fraction at the inner and outer surfaces of the fuel were determined by linear extrapolation of the data from contiguous fuel positions. The error bars drawn in Figure 4 represent the total uncertainty in the calculated Pu enrichment factor γ_{Pu} . All the fuels plotted on this diagram underwent appreciable restructuring during irradiation in EBR 2, their vital statistics [7] are summarized in Table 1. In determining the final overall Pu valency of the fuels the O/M change with burnup was accounted for by using $\Delta(\text{O/M}) \approx 0.002$ per atom percent burnup, which refers to fully U-235 enriched mixed oxide fuel in a fast neutron flux at O/M ratios less than 2.00. It appears there is a reasonably good linear correlation between γ_{Pu} and V_{Pu} for fuels with O/M (20% Pu) > 1.98 however, thus far, data for low O/M fuels are lacking. A number of fuel pins recently examined at ANL [4] also show Pu enrichments which can be fitted to the correlation shown in Figure 4; these pins (Numelec series) either finished or started near the stoichiometric composition ($V_{\text{Pu}} \approx 4.00$) and were sufficiently high rated that appreciable restructuring and central void formation occurred.

The evaporation-condensation mechanism for radial actinide redistribution predicts that low O/M fuels (< 1.97) should show Pu depletion at the center provided that center melting does not occur. It has been suggested [1,3] that this may never take place because, under such conditions, the proposed solid state Soret effect, which would be expected to move Pu preferentially up the thermal gradient in significantly hypostoichiometric fuel, could become important. Further, the much reduced vapor pressures of metal-bearing species over fuel at low O/M ratios may reduce the rate of U-Pu partitioning via the evaporation-condensation process to such an extent that solid state back diffusion of U and Pu ions may prevent the development of significant Pu/U+Pu gradients. The combined effect of these various factors may be to cause a lower degree of Pu depletion than predicted by extrapolation of the linear γ_{Pu} O/M correlation shown in Figure 4 to low O/M

values. Uncertainties in some of the high temperature thermodynamic data used (e.g., the vapor pressure calculations and in the high temperature/low O/M value of O_2) may also influence the actual value of the cross-over O/M corresponding to a particular linear power rating. We are hopeful that some of these uncertainties will be resolved as we continue to refine our calculational model and evaluate new irradiated fuel pin data. In concluding this action it is worth noting that the first report of Pu depletion at the center of an irradiated mixed oxide fuel was made recently by F. Schmitz and coworkers from CEA Euratom [8]; this effect was observed in a fuel with starting O/M = 1.90 which underwent appreciable restructuring during irradiation. Further results of this type are awaited with interest.

3.2 EFFECT OF BURNUP ON RADIAL ACTINIDE REDISTRIBUTION

Accumulation of data from post irradiation examinations of a number of adequately characterized GE mixed oxide fuel pins has reached the point where we can begin to search for correlations between γ_{Pu} and various suspected primary parameters. The effect of fuel stoichiometry has already been described; however, it is evident that interacting with this are the degree of restructuring of the fuel and its maximum temperature (which to a large extent are determined by the initial fuel density) and burnup or irradiation time. Table 2 summarizes data from four fuel sections which were selected to illustrate the effect of burnup at approximately constant stoichiometry. Further data on these fuel pins are given in Table 1.

Samples F2G G and F5Q H1 which started at the same O/M, attained markedly different burnups at similar (high) power levels. Appreciable restructuring occurred in each case. The difference in γ_{Pu} for these two pins is small and the direction of this change may simply reflect the difference in the final O/M ratios, i.e., γ_{Pu} is higher for the higher O/M fuel. Although they showed appreciable restructuring, the two intermediate burnup samples operated with significantly lower calculated center temperatures which may account for the somewhat low γ_{Pu} values obtained. At any event it appears from these preliminary observations that the effect of burnup on actinide redistribution is small and most probably derives from the O/M change with burnup.

3.3 EFFECT OF FUEL DENSITY

Table 3 summarizes the results of two short term irradiations performed with low and high density $U_{0.80}Pu_{0.20}O_{1.98}$ fuel pellets. It is judged significant that the high density fuel, which had the highest linear power rating, suffered less Pu enrichment than the relatively low density fuel. The photomicrographs of these fuel sections showed considerably less restructuring in the high density fuel than in the low density fuel, thus clearly the degree of actinide redistribution which can occur in a given fuel is predicated by the extent to which that fuel can restructure. Since fuel restructuring occurs primarily via evaporation/condensation across pores and bubbles migrating up the temperature gradient, the connection between actinide redistribution and restructuring becomes obvious.

In Table 4 a comparison is made between three fuel samples, all with high densities but each having a different starting O/M. The irradiations were short term (low burn up) and each fuel operated at the same (high) power rating. From the results an O/M effect whose direction agrees with the prediction of the vapor transport model is apparent—however the degree of Pu enrichment is substantially less than obtained from analogous low density fuels (see Figure 4). These results provide additional support for the correlation between degree of actinide redistribution and available volume for restructuring (and hence vapor transport) mentioned above.

3.4 AN EXAMPLE OF AXIAL ACTINIDE MIGRATION

To date our theoretical studies of actinide redistribution have been focussed on radial effects, as it was considered important to understand this basic fuel pin phenomenon before any attempt was made to model three dimensional actinide redistribution. However, during examinations of the radial Pu/Pu+U profiles in transverse sections from different axial locations of fuel pin F3B 3 (which began irradiation with a 40 mil diameter central void) [9] it became apparent that some axial transport of fuel had occurred and that this had produced a marked effect on the "radial" results. Here an attempt is made to explain the observed results in terms of the vapor transport "model."

Figure 5 shows a photomicrograph of specimen F3B 3M and corresponding radial distributions of Pu/U+Pu and oxygen. Figure 6 shows specimen F3B 3G. Sections 3M and 3G were taken from positions 12.9 inches and 6.8 inches from the bottom of the fuel column respectively; the 6.8 inch position corresponds to the reactor core mid plane and it ran 300 400°C hotter at the center line than the upper section. The simultaneous observations of a decrease in the central void diameter of the upper (cooler) fuel section and a particularly high value of γ_{Pu} together with an increase in the central void diameter of the central section, initially suggested that vapor transport of Pu rich material up the central void had occurred (i.e., evaporation from the central (hot) section and that preferential evaporation of Pu could not occur (unless gross reduction of the fuel occurred during irradiation) and mass balance calculations performed on these two fuel sections essentially eliminated this possibility. Qualitative radial oxygen profiles for these two fuel sections, taken with the shielded electron microprobe, indicated that the fuel in the upper (cool) section contained more oxygen (O/M = 2.00) than the central section (O/M = 2.00). Partial pressure calculations for the various metal bearing species (of the type originally performed by Rand and Markin [10]) indicate that uranium should move preferentially (as UO_3) to cooler regions inside the central void. This migration of UO_3 would also account for the observed axial relocation of oxygen. The resulting increase in O/M of the upper fuel section would then, because of the O/M - γ_{Pu} correlation discussed previously, tend to bring about a greater degree of Pu enrichment due to radial actinide redistribution than at the central fuel section. Although it is probably over simplified this explanation is fully consistent with the predictions of our current vapor transport models for both oxygen and actinide redistribution.

SUMMARY AND CONCLUSIONS DELINEATION OF KEY PARAMETERS IN RADIAL ACTINIDE REDISTRIBUTION

Three important parameters that have been identified as contributing to the occurrence of this phenomenon are fuel O/M (over all value), maximum (center line) temperature and time of operation at this temperature and initial fuel density. Radial actinide redistribution is intimately associated with fuel restructuring and thus, when over all O/M is favorable, the degree to which Pu depletion or enrichment takes place is directly dependent on fuel linear power rating and the consequent degree of fuel restructuring. The latter is a function of initial fuel density, above 96% theoretical density fuel, even at very high power ratings, shows very little actinide redistribution when compared with fuel of ~90% theoretical density. In terms of the vapor transport model this is consistent with a drastic reduction in the available volume for vapor flow as fuel density increases. Fuel which undergoes appreciable restructuring without center melting shows Pu enrichment at the center line when its over all O/M (20% Pu) > 1.97, the degree of enrichment appears to increase linearly with O/M up to ~2.00. Theoretically a fuel should not show any actinide redistribution when its over all O/M has reached ~1.97; however this remains to be confirmed. Because the center line temperature falls as restructuring proceeds (at constant power rating) it is not possible to give an exact figure for a "redistribution" temperature threshold. Such a determination is feasible however from the out of pile radial temperature gradient tests currently underway at GE. The uncertainties presently inherent in calculating center line temperatures also tend to make an in-pile determination a rather futile exercise. However, based on vapor pressure calculations out of pile experiments it is considered unlikely that preferential evaporation/condensation is an effective mechanism for actinide redistribution at temperatures below 2000°C.

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TABLE 1 Irradiated Fuel Pin Data

Simpl. No.	Inlet Composition O/M	MPu	Section Height (inches from bottom)	Density (% T.D.) Sintered Pellet	Annulus diam. (in mils)	Peak Linear Power (kw/ft)		T ₀ (calcd.) °C		Burnup (at. %)	
						SOL	EOL	SOL	EOL	Anal	Calc
F3B-3M	2.00	25	12.9	—	41	16.36	15.85	2163*	—	4.9	—
F3B-3G	2.00	25	6.8	—	41	16.36	15.85	2563*	—	5.3	—
F2G-G	1.975	20	6.2	92.9	—	14.86	17.81	2570	2319	10.8	12.7
F50-H1	1.974	25	9.8	90.85	—	13.92	13.38	2519	—	4.9	5.7
E1H-30F	1.98	25	7.5	89.9	—	13.1	11.6	2175	1975	7.7	9.7
E1H-23K	1.974	25	11.0	—	41	~13	—	1800	1800	7.0	~9
F1E	1.98	20	6.5	86.6	—	14.8	—	~2540	—	0.17	0.20
F1D	1.98	20	6.5	92.8	—	16.2	—	—	—	0.17	0.20
F1B	2.00	20	8.5	93.9	—	16.4	—	—	—	0.7	0.20
F1C	2.03	20	8.5	93.8	—	16.8	—	—	—	0.16	0.21

*All fuel made from coprecipitated mixed oxide.

*With high power driver c.

TABLE 2 Effect of Burnup

Sample	O/M	Average Power (kW/ft)	Burnup (at %)	γ Pu
F2G G	1 975 (1 992)	14 2	10 8	1 23
E1H 30F	1 98 (1 992)	12 4	7 7	~1 1
E1H 23K	1 974 (1 985)	13 0	7 0	~1 1
F5OH1	1 974 (1 982)	13 6	4 9	1 18

TABLE 3 Effect of Density

Pin No	O/M	Pellet Density (% th)	Average Power kW/ft	γ Pu
F1E	1 98	89 2	14 3	~1 1
F1D	1 98	95 3	16 2	1 0

(Burnup = 0 17 at %)

TABLE 4 Effect of O/M at High Density

Pin No	O/M	Pellet Density (% th)	Average Power kW/ft	γ Pu
F1D	1 98	95 3	16 2	1 00
F1B	2 00	96 2	16 4	1 07
F1C	2 03	96 6	16 8	1 18

(Burnup = 0 17 at %)

FIGURE 1 CALCULATED RADIAL OXYGEN (O/M) GRADIENTS FOR SEVERAL FUEL COMPOSITIONS ($Q_0 = 10$ kcal/mole)

FIGURE 2 FLOW DIAGRAM FOR PUGRAD

FIGURE 3 CALCULATED Pu PROFILES FOR $MO_{1.968}$, $MO_{1.988}$ AND $MO_{2.000}$
(J indicates number of iterations)

FIGURE 4. CORRELATION BETWEEN DEGREE OF PLUTONIUM ENRICHMENT (γ_{Pu}) AND STOICHIOMETRY (O/Pu) IN IRRADIATED MIXED OXIDE FUELS HAVING UNDERGONE APPRECIABLE RESTRUCTURING

FIGURE 5. PHOTOMICROGRAPH OF SPECIMEN F3B-3M AND CORRESPONDING RADIAL DISTRIBUTIONS OF $Pu/U + Pu$ AND OXYGEN

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SUMMARY

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FIGURE 6. PHOTOMICROGRAPH OF SPECIMEN F38-3G AND CORRESPONDING RADIAL DISTRIBUTIONS OF Pu/U + Pu AND OXYGEN

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