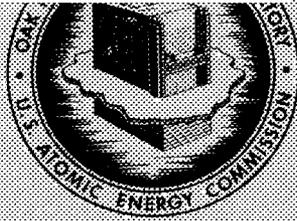


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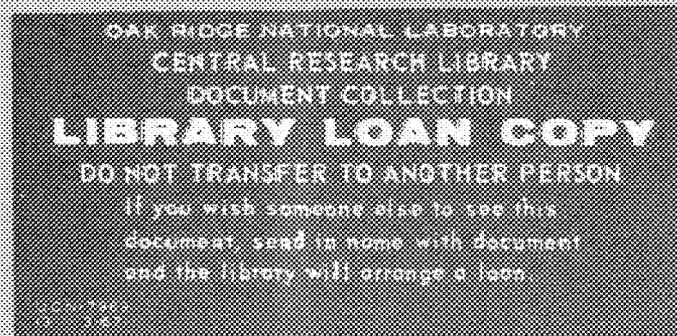


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IN-REACTOR RESTRUCTURING TEMPERATURES AND KINETICS FOR  $(U, Pu)O_2$

A. R. Olsen, R. B. Fitts, and W. J. Lackey



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A. R. Olsen, R. B. Fitts, and W. J. Lackey

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IN-REACTOR RESTRUCTURING TEMPERATURES

AND KINETICS FOR (U,Pu)O<sub>2</sub>

A. R. Olsen    R. B. Pitts    W. J. Lackey

ABSTRACT

Data from a number of thermal flux irradiation tests of stainless-steel-clad (U,Pu)O<sub>2</sub> fuels were analyzed in terms of proposed mechanisms of fuel restructuring and actinide redistribution. The data indicate that columnar grain growth is the result of a vaporization-condensation process in fuels operating above about 1700°C. The columnar structure is not fully developed in fuels with a center temperature of 2000°C even after 28 days of operation, but the rate of restructuring increases with increasing temperature. Sintering and equiaxed grain growth processes are slower and therefore of secondary importance in early-life restructuring. A model is postulated in which the vaporization-condensation process can account for observed actinide concentration variations with either uranium or plutonium depletion in the highest temperature regions, depending on the operating temperatures and probably on the oxygen activity in the gas phase over the fuel.

## INTRODUCTION

In-reactor restructuring of oxide fuels has long been studied, primarily because of the effects on fuel operating temperatures. This is of interest for two reasons: (1) one of the reactor design criteria normally imposed is that there be no molten fuel during steady-state operation, and (2) if the restructuring kinetics are known, the reactor startup procedures can be optimized so that the condition of no fuel melting can be met with new fuel subassemblies or with assemblies that have been moved from a region of low to higher flux. In addition, an understanding of the modes of in-reactor restructuring of (U,Pu)O<sub>2</sub> fuels and the kinetics of the various processes as functions of temperature, temperature gradient, and time are necessary for the analysis of fuel pin performance.

We have examined the data on fuel microstructures, composition gradients, and porosity from a number of irradiation tests, both instrumented<sup>1</sup> and uninstrumented,<sup>2,3</sup> and analyzed these data in terms of proposed processes of fuel restructuring. The data indicate that early-life restructuring above 1700°C occurs by the vaporization-condensation mechanism and suggest that this same process leads to observed variations in actinide distribution.

## OXIDE RESTRUCTURING

Table I is a compilation of various structural phenomena observed in irradiated fuel together with most of the conditions now thought to affect these phenomena. Other facets of fuel or fuel pin performance, such as fission gas release rates, are closely related to restructuring.

Table I. Conditions Affecting (U,Pu)O<sub>2</sub> Fuel Structural Phenomena

Condition	Involvement of Condition in Each Phenomenon		
	Grain Growth		Sintering
	Columnar	Equiaxed	
Temperature	✓	✓	✓
Temperature gradient	✓		
Time	✓	✓	✓
Fuel oxygen-to-metal ratio	✓		✓
Fuel composition	✓		
Fuel density	✓	✓	✓
Hydrostatic stress		✓	✓

Although it is beyond the scope of this paper to discuss each of these items in detail or the effects of neutron flux and fissioning processes on many of them, the tabulation does reveal the most important factors to which our analyses of the experimental data have been directed.

Temperature is a key factor in all these phenomena and also depends on the extent of restructuring, since the thermal conductivity of the fuel increases with its density. The instrumented tests in our program were specifically designed to investigate the effects of temperature on restructuring in both pellet and Sphere-Pac fuels and to evaluate the methods of predicting fuel operating temperatures in non-instrumented tests. The Sphere-Pac process, which uses low-energy vibratory compaction of fuel microspheres, has been described previously.<sup>4</sup> The results of the instrumented tests confirmed the method of temperature prediction as reported previously.<sup>3</sup> Briefly, these tests showed that the predictions agree within  $\pm 8\%$  of the measured temperature and that for equivalent smear densities there is no significant difference in overall fuel pin thermal conductance for pellet and Sphere-Pac fuels. The comparative data are shown in Fig. 1. The instrumented test results are important for two major reasons. First, they provide confidence in the temperature quoted for the uninstrumented tests. Second, postirradiation examination showed that the fuel form does not have a significant effect on restructuring. This second point can be seen in Fig. 2, where we have plotted calculated temperatures on radial cross sections of the two fuel types — pellet and Sphere-Pac — from an instrumented test. Note that above about  $1650^{\circ}\text{C}$  the original fuel form has been completely obliterated by restructuring, and the two fuel forms are very similar.

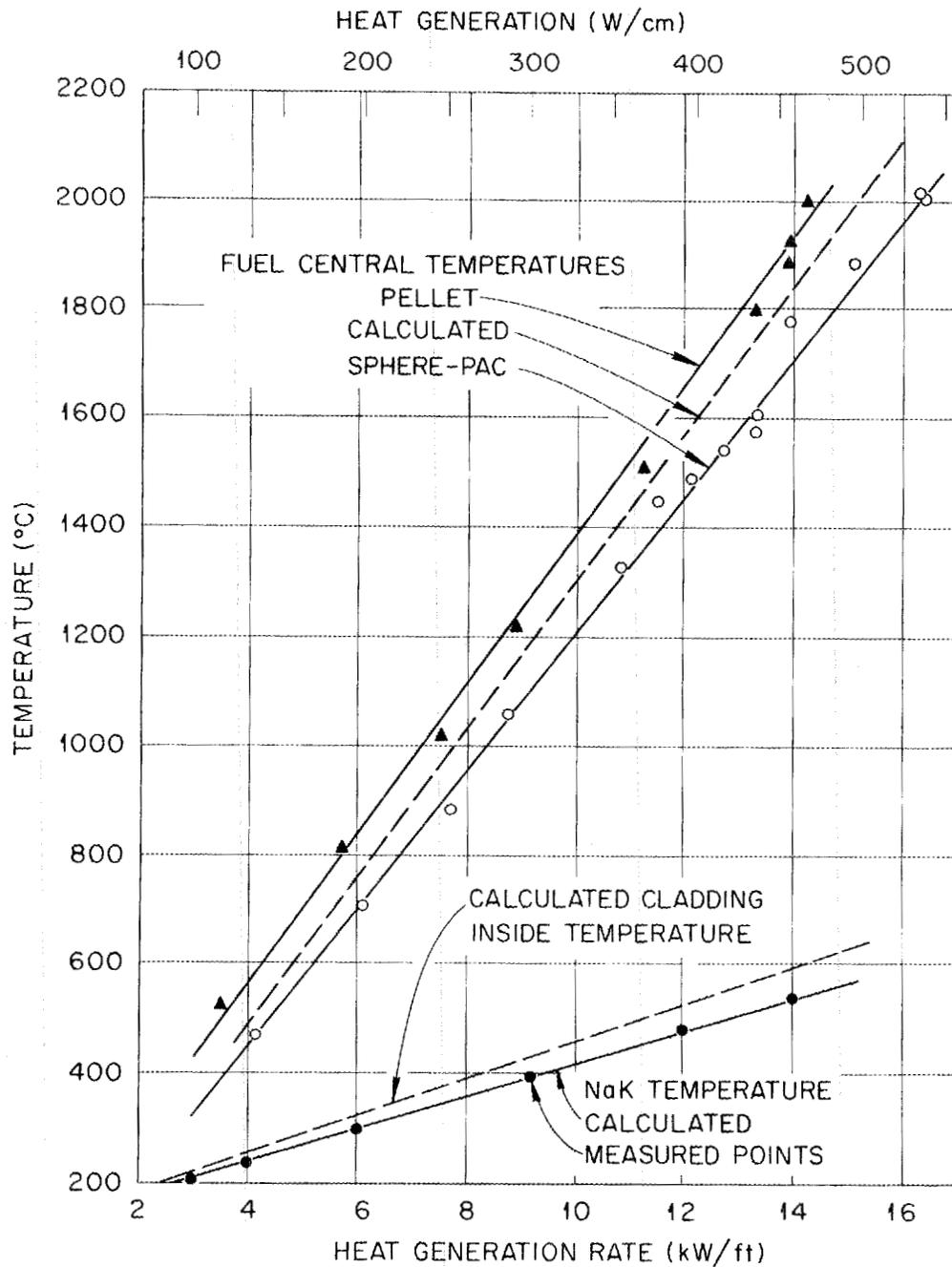


Fig. 1. Variation of Calculated and Measured Fuel and Capsule Temperatures with Heat Generation Rate in Capsule SG-3.

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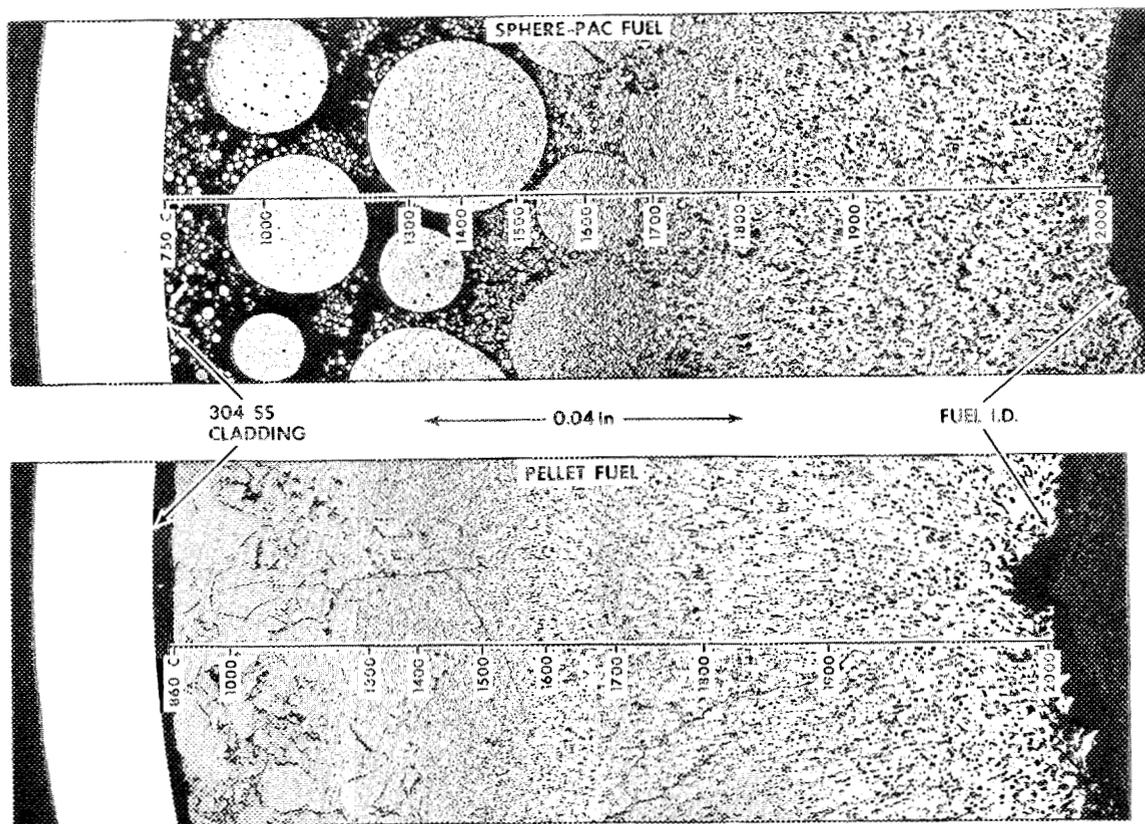


Fig. 2. Temperatures in  $U_{0.8}Pu_{0.2}O_{1.99}$  Fuel During Irradiation.

Slightly elongated grains occur in this high-temperature region for the pellet pin. A second temperature demarcation for these tests is at approximately 1300°C, where the first discernible outlining of grain boundaries (porosity or bubbles in the grain boundaries) is observed in both fuels.

Being assured that the fuel fabrication form, Sphere-Pac or pellet, is not a primary factor in fuel restructuring and that fuel temperatures can be calculated with reasonable reliability, we can confidently utilize the data from noninstrumented Sphere-Pac tests to investigate the effects of temperature, temperature gradient, and time on restructuring.

The effects of temperature and possibly temperature gradient on the restructuring rate are shown in Figs. 3 and 4. Each figure shows cross sections from pins operated in the same capsule but at different flux levels. The  $U_{0.85}Pu_{0.15}O_{2.00}$  Sphere-Pac fuel in all six pins was identical. The calculated temperatures of apparent restructuring limits (radii of observable equiaxed-grain growth) for Fig. 3 are approximately 1300°C, in agreement with the temperature limits defined by the instrumented tests. In Fig. 4 the temperature limits of restructuring are not well defined because of the changes in temperature profile as the heat rate decreases and the power distribution changes with burnup.<sup>5</sup> The change in radial extent of restructuring is difficult to define for  $\int k d\theta$  between 32 and 40 W/cm but is quite apparent when  $\int k d\theta$  rises to a time-averaged value of 46 W/cm.

A surprisingly long time dependence for restructuring in fuels operated with an  $\int k d\theta$  of 31 to 32 W/cm is shown in Fig. 5. The upper two

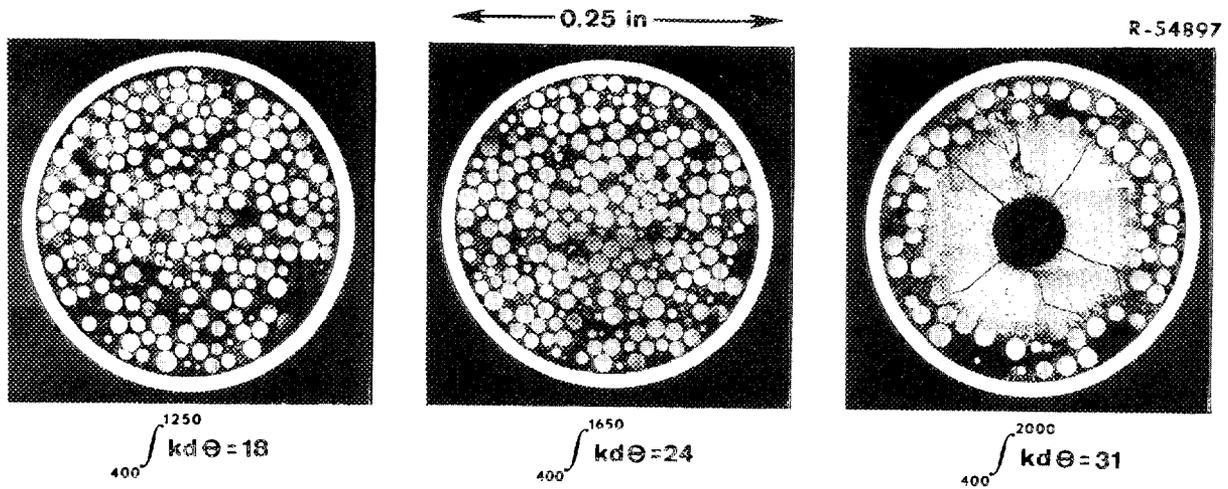


Fig. 3. Effect of Fuel Temperature on Restructuring During 28 Effective Full Power Days.

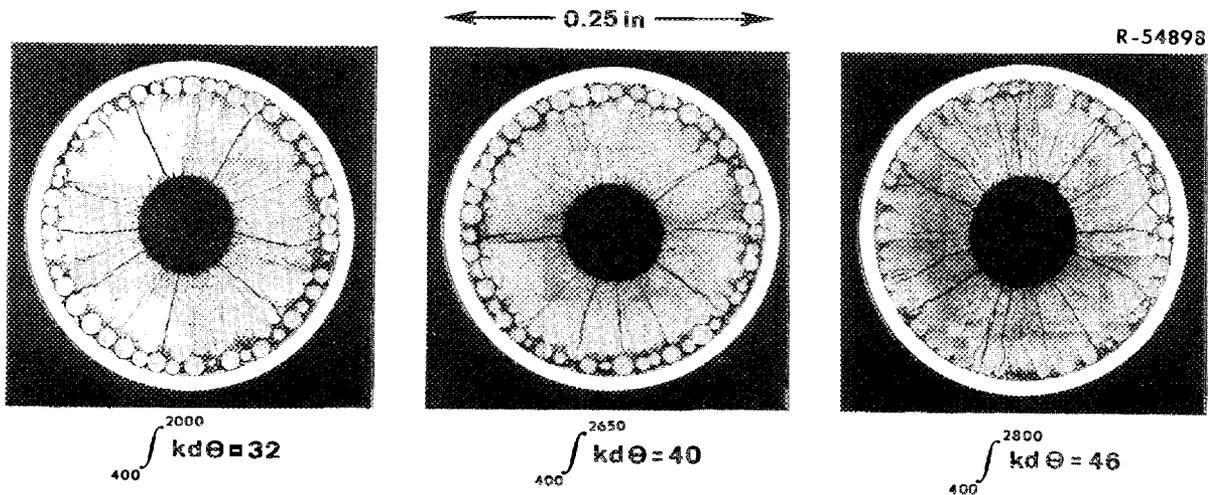


Fig. 4. Effect of Fuel Temperature on Restructuring During 174 Effective Full Power Days of Irradiation.

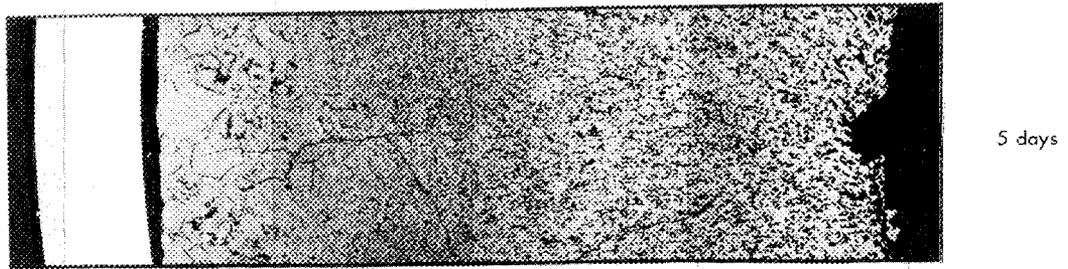
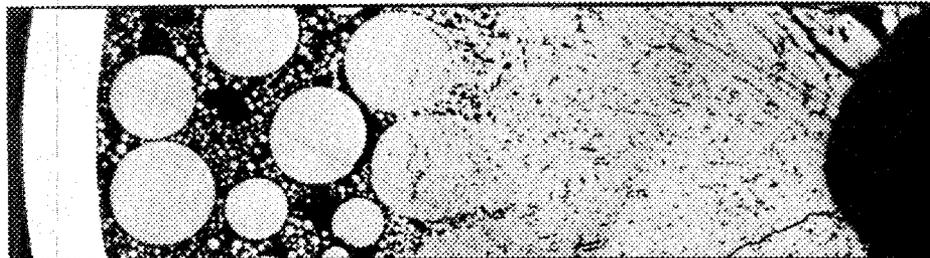
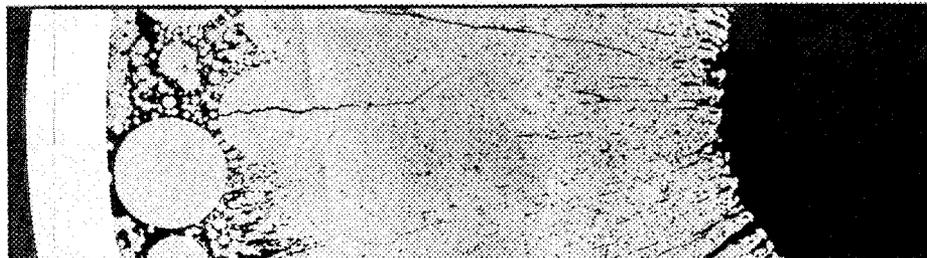
Pellets  $(U_{0.80}Pu_{0.20})O_{1.98}$ Sphere Pac  $(U_{0.80}Pu_{0.20})O_{1.98}$ Sphere Pac  $(U_{0.85}Pu_{0.15})O_{2.00}$ Sphere Pac  $(U_{0.85}Pu_{0.15})O_{2.00}$ 

Fig. 5. Effect of Irradiation Time on Restructuring of  $(U,Pu)O_2$  Fuels with a Center Temperature of Approximately  $2000^\circ\text{C}$ .

structures are from the instrumented tests, and the lower two are from uninstrumented capsule irradiations in the Engineering Test Reactor (ETR). These microstructures show that the columnar grain structure is indistinct after two to five days of operation and not fully developed after 28 days of operation. For example, a large number of lenticular voids are still visible in the columnar grain region. However, after 174 days the structure is free of lenticular voids and has grown radially. We do not have information yet on longer times; however, we believe that in the 174-day structure the columnar grain growth is essentially complete.

The unique characteristics of the Sphere-Pac microstructure indicate that the primary restructuring process in fuels at temperatures above about  $1700^{\circ}\text{C}$  is vaporization-condensation. Figure 6 clearly shows the growth of condensed dendrites at the outer radial boundary of the columnar grain region for the shorter term tests (28 EFPD). This phenomenon has long been proposed as the process by which columnar grains are formed as pores sweep up the temperature gradient.<sup>6,7</sup> In fact, calculations of pore distribution using the pore migration model of Nichols<sup>8</sup> showed reasonable agreement with porosity distribution measurements made on our tests.<sup>9</sup> Our calculations and observations show a slower rate of densification for these thermal flux tests than the predictions of others<sup>10</sup> for fast reactor conditions at similar linear heat rates. This difference is largely the result of a different fuel center temperature, which is some  $300^{\circ}\text{C}$  higher in the fast reactor calculations; the rate of pore migration in the model varies exponentially with temperature. A secondary effect, resulting from flux perturbation effects,

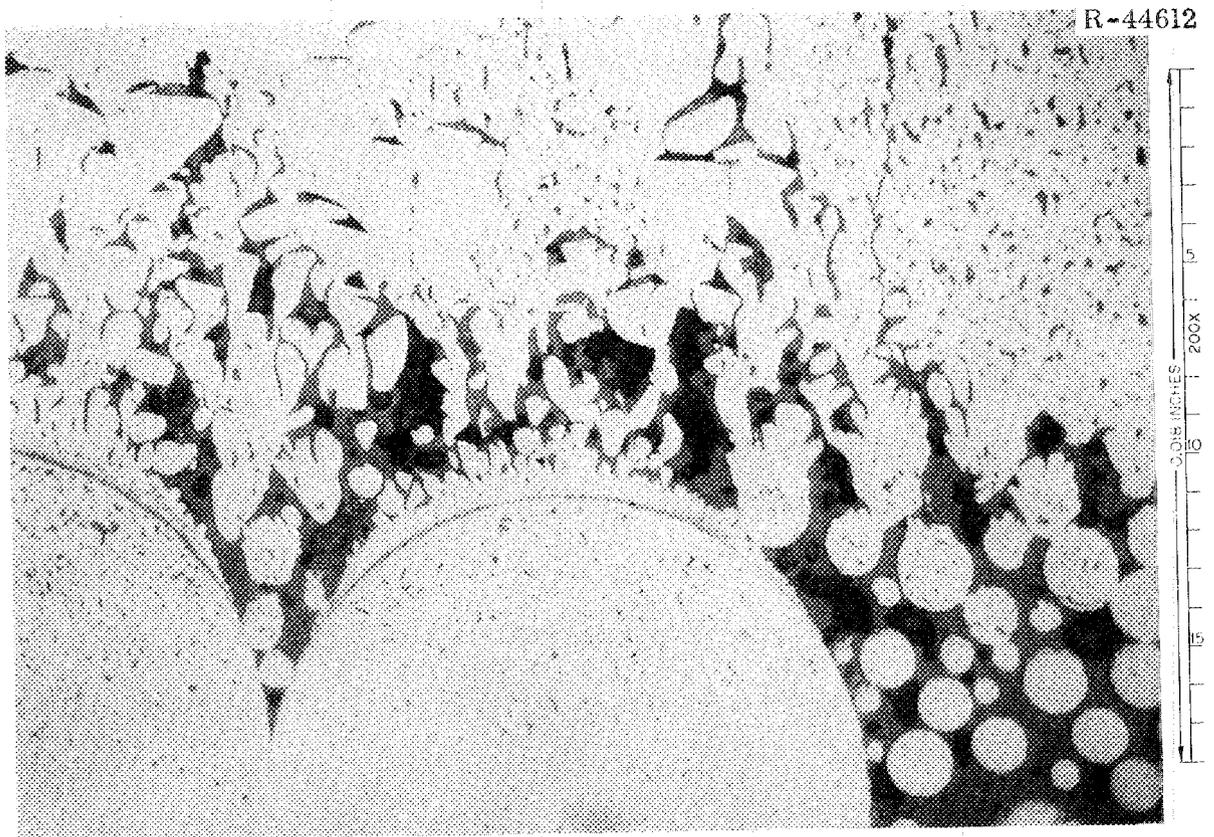


Fig. 6. Fuel Deposits on the Inner Surface of Microspheres Located at the Periphery of the Columnar Grain-Growth Region.

may be the differences in local thermal gradients, particularly in the higher temperature region.

Sintering and grain growth processes are of secondary importance in short-term restructuring, although they do provide a means of agglomerating as-fabricated porosity for the vaporization-condensation process. This agglomeration process is apparent in the electron micrographs shown in Fig. 7. The irradiated structure shown in the figure is from a fuel region with a calculated temperature of 1450°C. At this temperature and higher we occasionally observed the rows of small voids seen in Fig. 7. We believe these to be bubbles of fission gas at grain boundaries. These voids were 0.1 to 0.2  $\mu\text{m}$  in diameter, an order of magnitude smaller than the agglomerated porosity.

Examination of the cooler fuel regions from the higher burnup samples has revealed the structure shown in Fig. 8. Here we see the development of subgrains, indicated by the arrows within the equiaxed grains. Similar structural changes have been reported by Bailey et al.<sup>11</sup> As yet we do not know whether the subgrains are outlined by bubbles or inclusions. The development of such a structure could increase the "surface area" of the fuel and result in increasing fission gas release with burnup for fuel in the 800 to 1400°C range, as the experiments of Findlay et al.<sup>12</sup> have shown. Additional observations will be made on higher burnup tests to see if this subgrain structure develops with burnup. Preliminary measurements of equiaxed, grain-growth kinetics have been reported,<sup>13,14</sup> and additional measurements are in progress. Since the current equations and constants are reported elsewhere<sup>9</sup> at this conference, they will not be discussed here. However, we have seen grain growth at fuel temperatures as low as 900°C.

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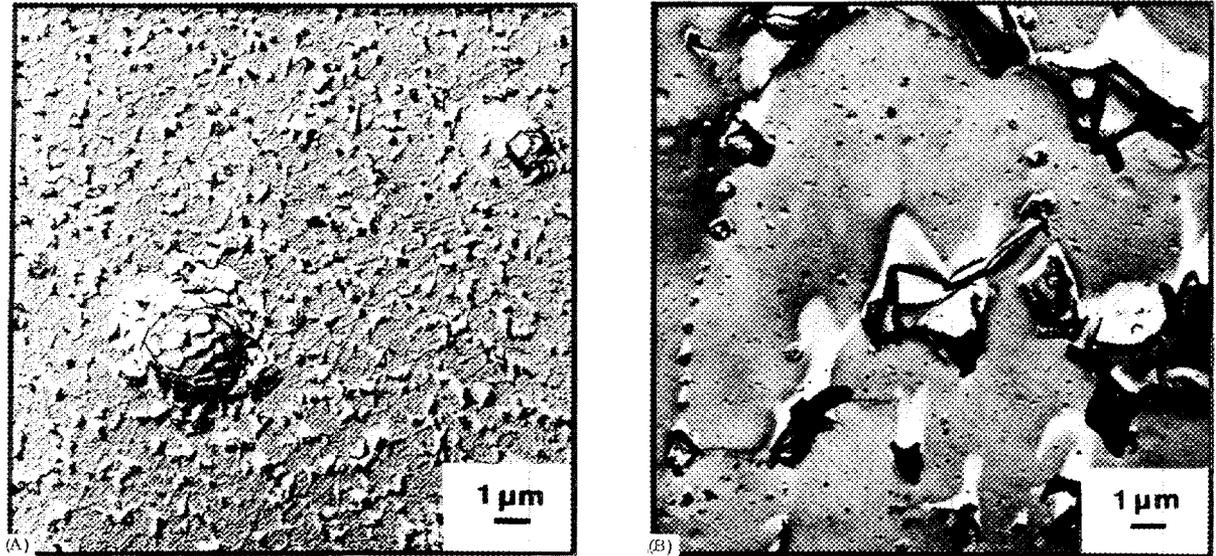


Fig. 7. Electron Micrographs of Pores in Coarse Microspheres.

(A) As fabricated and (B) after irradiation at temperatures below 1450°C for 28 effective full power days.

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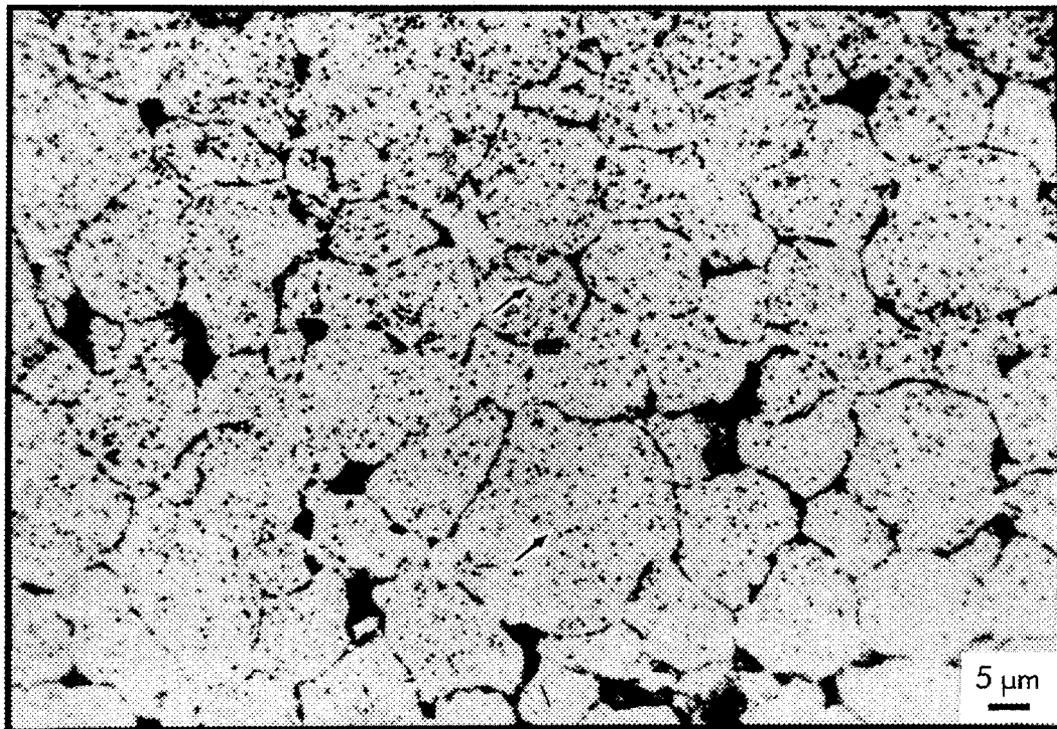


Fig. 8. Subgrain Growth in (U,Pu)O<sub>2</sub> Irradiated for 174 Effective Full Power Days.

## URANIUM-PLUTONIUM REDISTRIBUTION

The vaporization-condensation restructuring process does have a secondary effect on actinide distribution. Although we have not been able to establish a quantitative method of predicting such redistribution, our observations and calculations have allowed us to establish a qualitative mechanism.

The unique structure of Sphere-Pac fuels allows us to see the effects of the vaporization process on plutonium distribution. Figure 9 shows the  $\alpha$  and  $\beta$ - $\gamma$  autoradiographs along with the photomacrographs for two different pins from the same capsule. Figure 9(a) from the pin with the lower heat rate does not show the ring structure in the  $\beta$ - $\gamma$  autoradiograph typically seen where fuel melting has occurred. The interesting features are revealed in the  $\alpha$  autoradiograph. First there is a large increase in plutonium content near the central void. Secondly, there are spherical islands of higher plutonium content in the cooler parts of the regions with the columnar grain-growth structure. We attributed<sup>3,14</sup> these observations to a migration of uranium-rich vapors down the temperature gradient, with condensates filling the voids around the larger microspheres. At intermediate temperatures, diffusion and short-range vaporization-condensation processes provide homogenization. The loss of the uranium-rich vapors from the fuel near the central void leaves a plutonium-enriched solid behind.

Figure 9(b) is from a pin with a higher heat rate where, at least in the early stages of irradiation, a portion of the fuel was molten. The extent of melting is indicated by the inner edge of the high  $\beta$ - $\gamma$  activity ring. The  $\alpha$  autoradiograph shows the plutonium-rich spherical

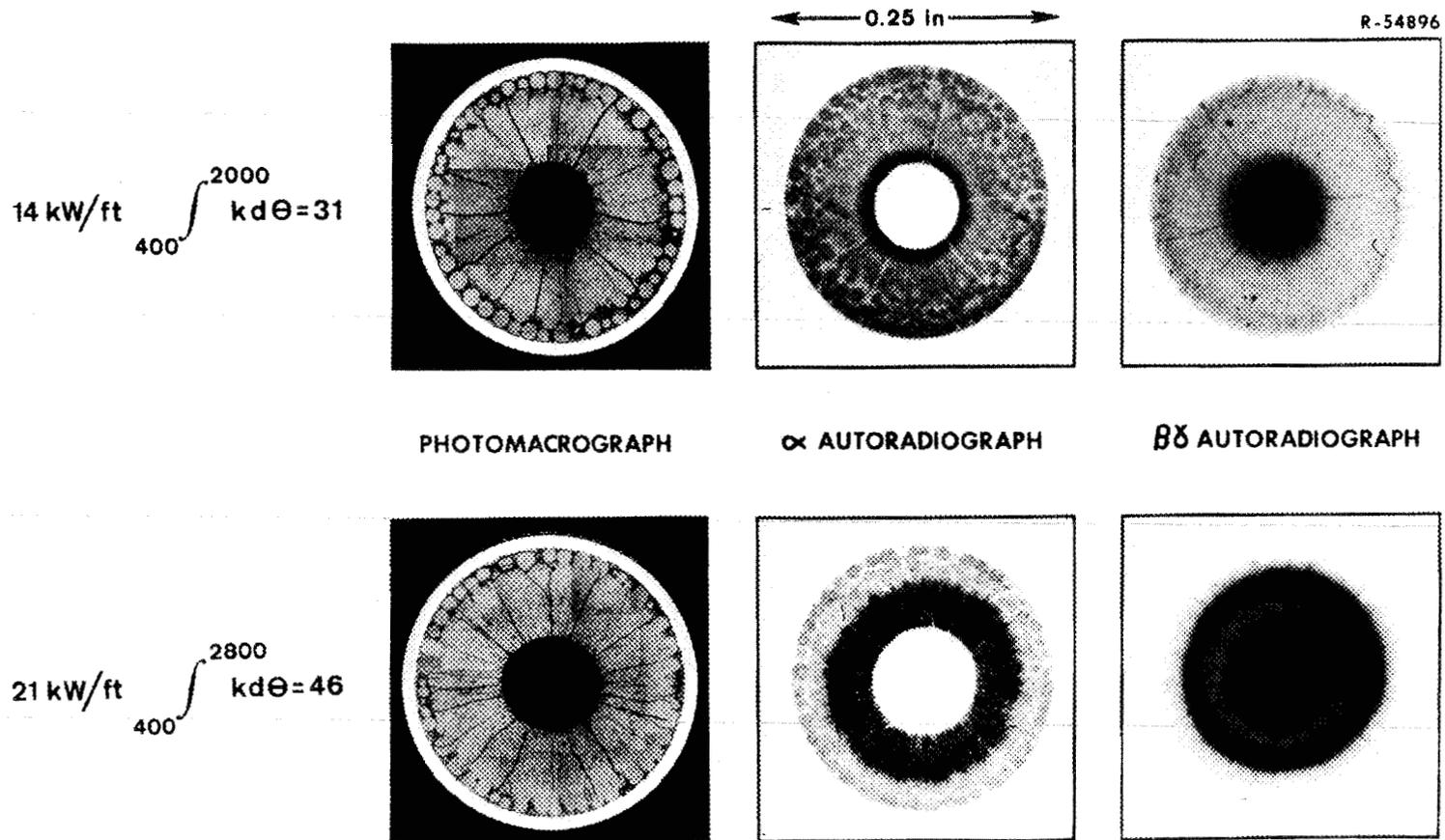


Fig. 9. Comparison of Postirradiation Autoradiography with Fuel Structure for Two  $U_{0.85}Pu_{0.15}O_2$  Pins Irradiated for 174 Effective Full Power Days at Two Different Heat Rates.

islands in the cooler regions of the columnar grains, although they are less distinct than those in Fig. 9(a). The major difference between this pin and the one with lower heat rate is in the plutonium concentrations near the central void. The pin with the higher heat rate does not show the sharp rise in concentration at the void, but, in fact, indicates a small decrease. Such a decrease cannot be attributed to solidification from the melt because this process would yield a peak in plutonium content for the last segment to solidify. Thermal diffusion<sup>15</sup> is another means of establishing a plutonium gradient but would not explain a peak away from the void surface. We feel that the vaporization-condensation process can explain both observations and we will show our reasoning.

As a first step in developing this model of actinide redistribution, we calculated the vapor pressures of Pu, PuO, PuO<sub>2</sub>, U, UO, UO<sub>2</sub>, and UO<sub>3</sub> over different solid compositions as functions of temperature and the H<sub>2</sub>O:H<sub>2</sub> ratio in the gas phase, using the thermodynamic data of Rand and Markin.<sup>16</sup> Figure 10 shows the vapor composition over a hypostoichiometric U<sub>0.8</sub>Pu<sub>0.2</sub>O<sub>2-x</sub> fuel with different H<sub>2</sub>O:H<sub>2</sub> ratios. Similar curves have been calculated for different solid compositions, and the effect of the solid composition at a constant H<sub>2</sub>O:H<sub>2</sub> ratio can be seen in Fig. 11. The total combined pressures of all uranium- and plutonium-bearing species are shown in Fig. 12. We were unsuccessful in attempting to calculate radial variations in plutonium contents for the solid by the method proposed by Aitken and Evans.<sup>17</sup> Contrary to their results, our calculation for hypostoichiometric fuels predicts that the plutonium-to-uranium ratio in the gas phase increases with increasing temperature

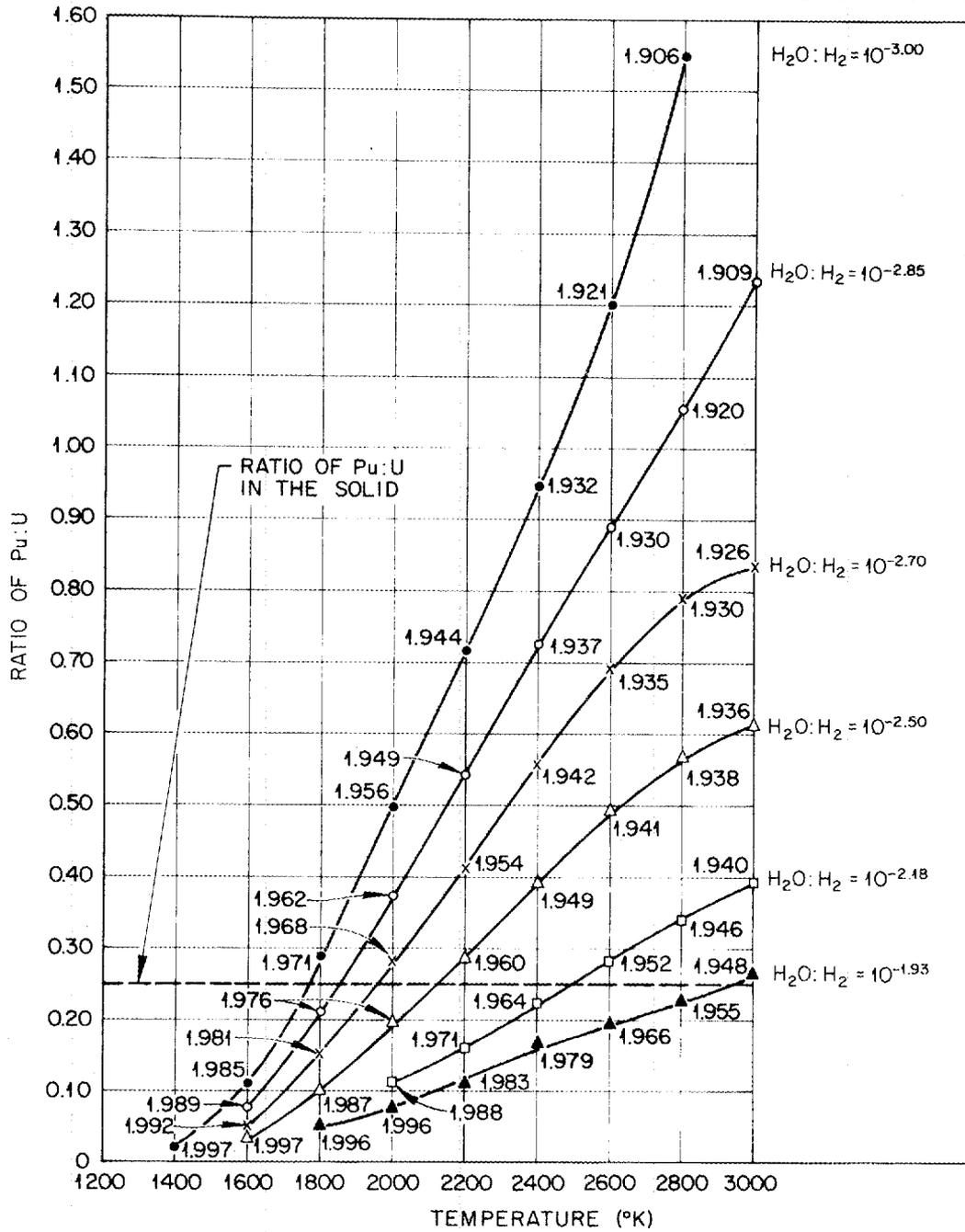


Fig. 10. Ratio of Plutonium to Uranium in the Gas Phase over  $U_{0.8}Pu_{0.2}O_{2-x}$  as a Function of Temperature.

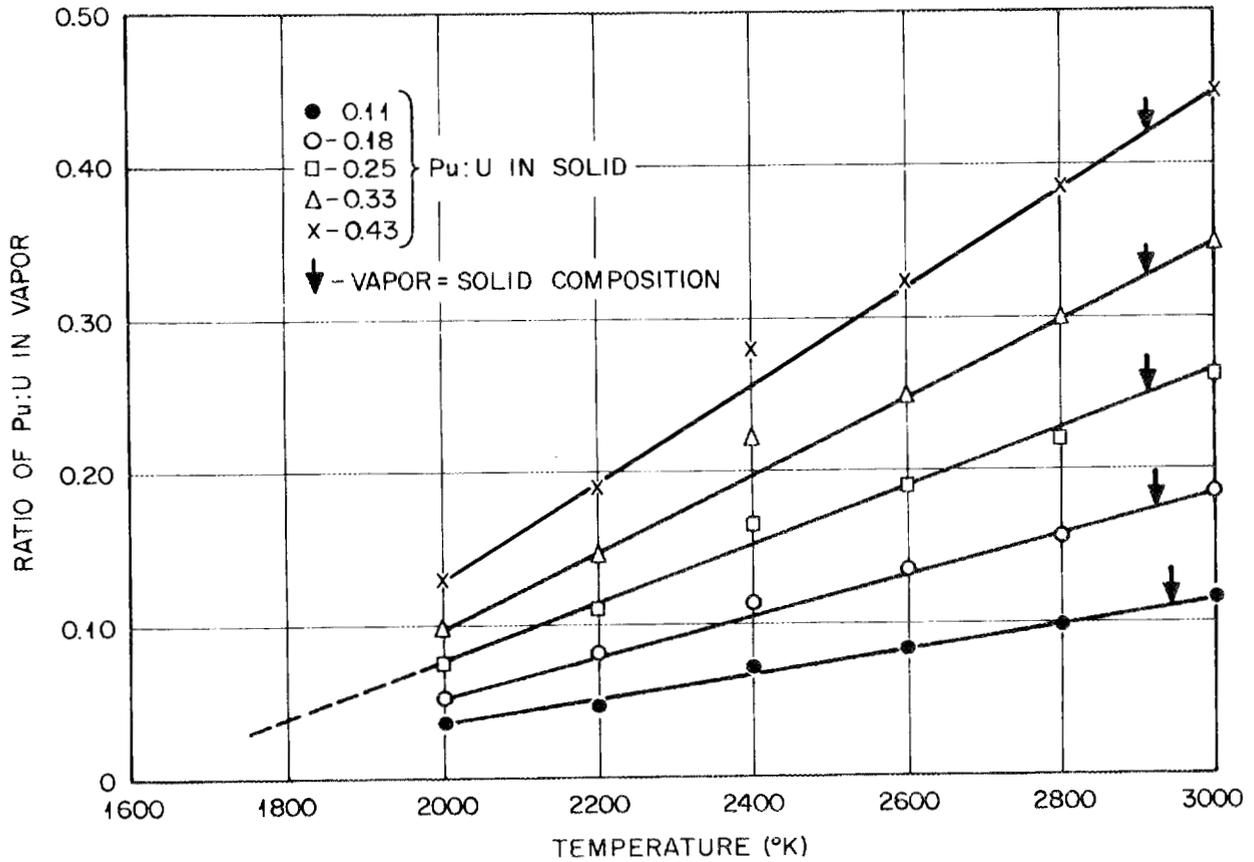


Fig. 11. Calculated Ratio of Plutonium to Uranium in Vapor Phase over Various Solid-Phase Compositions at an Oxygen Activity Equivalent to  $\text{H}_2\text{O}:\text{H}_2 = 10^{-1.93}$ .

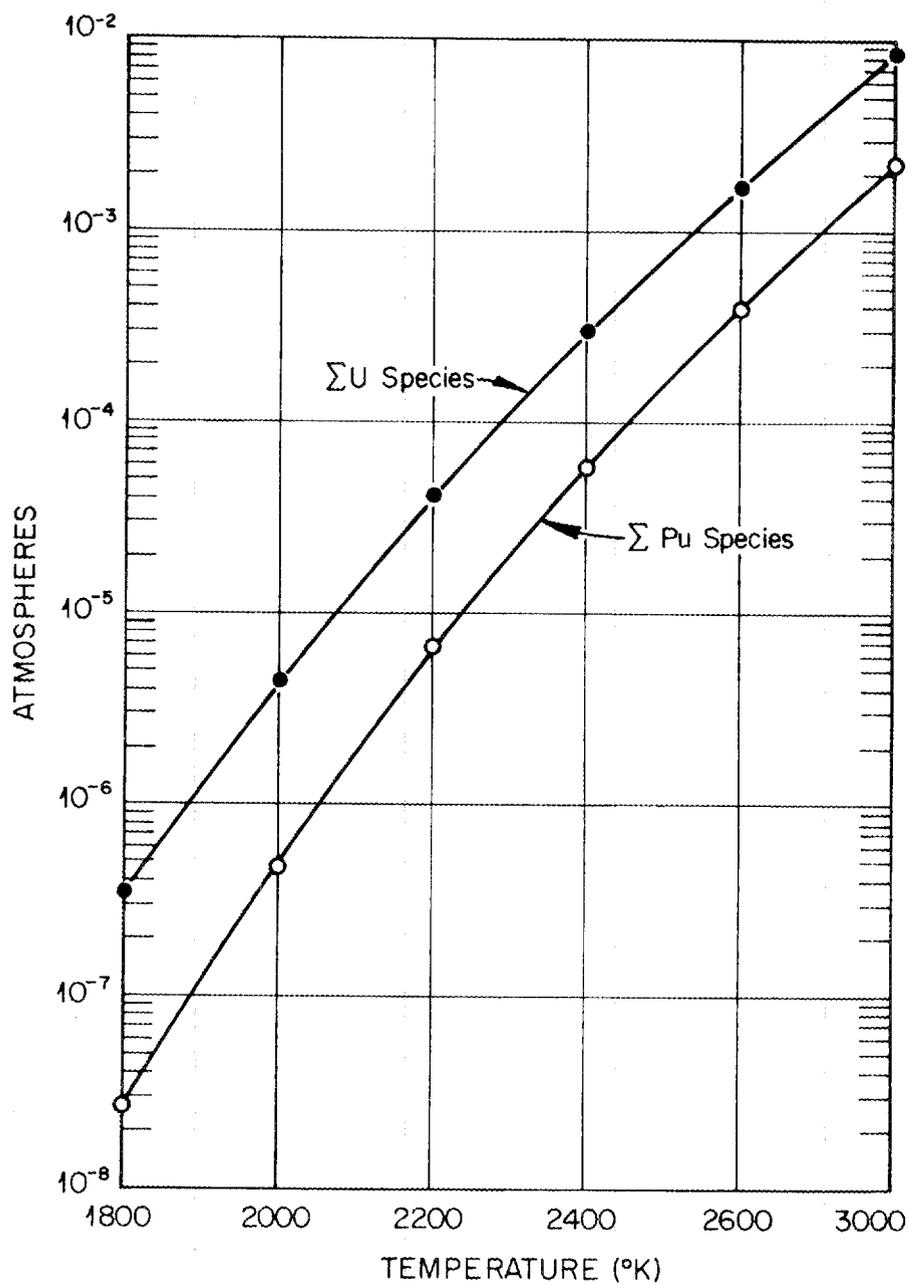


Fig. 12. Calculated Pressures of Uranium- or Plutonium-Bearing Vapor Species over  $U_{0.8}Pu_{0.2}O_{2-x}$  for  $H_2O:H_2 = 10^{-1.93}$ .

for any given water-to-hydrogen ratio in the range  $10^{-1.93}$  to  $10^{-3.0}$ . We have no explanation for the difference between our results and Aitken's.

On the basis of our observations and calculations, we postulate the following qualitative model:

1. Vaporization-condensation will occur in the steep temperature gradients ( $10^3$  to  $10^4$ °C/cm) of fuel under irradiation, since the fuel vapor partial pressures change by approximately an order of magnitude for every 200°C.

2. On the basis of currently available thermodynamic data, the vapor composition over a  $(U,Pu)O_{2-x}$  fuel will become enriched in plutonium-bearing species as the temperature increases.

3. The temperature at which the plutonium content in the vapor exceeds the plutonium content in the evaporating solid decreases with decreasing oxygen activity in the atmosphere (lower  $H_2O:H_2$  or  $CO:CO_2$  ratios, as determined by initial fuel oxygen-to-metal ratios).

4. If a fuel is irradiated under conditions such that the center temperature is below the temperature at which the plutonium content in the vapor equals the plutonium content in the solid, there will be a net migration of uranium down the temperature gradient, leaving a higher plutonium content in the higher temperature regions.

5. If the fuel is operated with center temperatures above the temperature at which the plutonium content in the vapor exceeds that in the solid, the plutonium content will peak at some temperature below the peak temperature.

We have only a limited amount of microprobe data available at this time, but the data to date support this postulate. Figure 13 shows the plutonium-to-uranium ratio in an irradiated fuel near the central void. The initial oxygen-to-metal ratio for the fuel was between 1.99 and 2.00, so an oxygen activity equivalent to a  $H_2O:H_2$  ratio less than  $10^{-2}$  is expected<sup>18</sup> in the gas atmosphere. With a center temperature of approximately  $2000^\circ C$ , a rise in plutonium content to a maximum at the edge of the central void is expected and was observed.

Figure 14 shows the microprobe analysis for the pin operated with center melting at the start of life and with center temperatures about  $2700^\circ C$  ( $3000^\circ K$ ) at the end of life. The initial fuel was the same, so the oxygen activity in the gas would be similar. However, at this higher temperature a peak in plutonium content would be expected in the columnar grains away from the center void. A reported microprobe analysis for a fuel irradiated in the EBR-II with central temperatures near the melting point shows a peaking in plutonium away from the central void, further confirming our evidence on the temperature effect.<sup>19</sup> We still see the decrease in plutonium content at the outer edge of the columnar grain region; the 1400- to 1600- $\mu m$  distance in the figure.

We have not yet examined any fuel with a lower oxygen-to-metal ratio, so the expected effects of oxygen activity are less well substantiated. There is some indication in reports of the General Electric Company<sup>20-22</sup> that plutonium gradients vary with initial oxygen-to-metal ratios, but the test conditions (88 hr with central melting) were such that equilibrium was probably not attained. A preliminary report<sup>23</sup> indicated that in EBR-II irradiations of the same fuel in two different

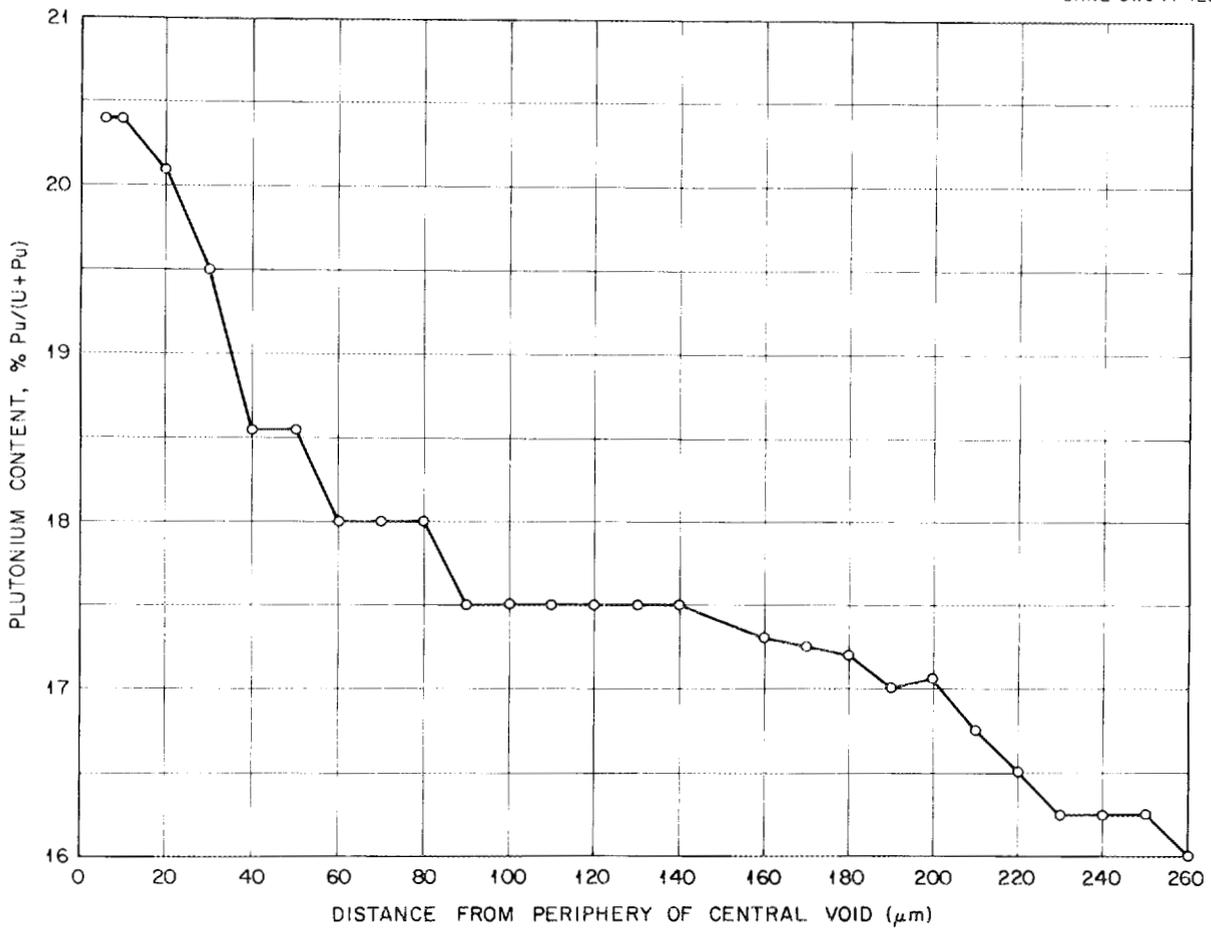


Fig. 13. Variation in Plutonium Content with Radial Position for  $\text{U}_{0.85}\text{Pu}_{0.15}\text{O}_{2.00}$  after Irradiation at a Linear Heat Rate of 13.6 kW/ft to 0.7% FIMA.

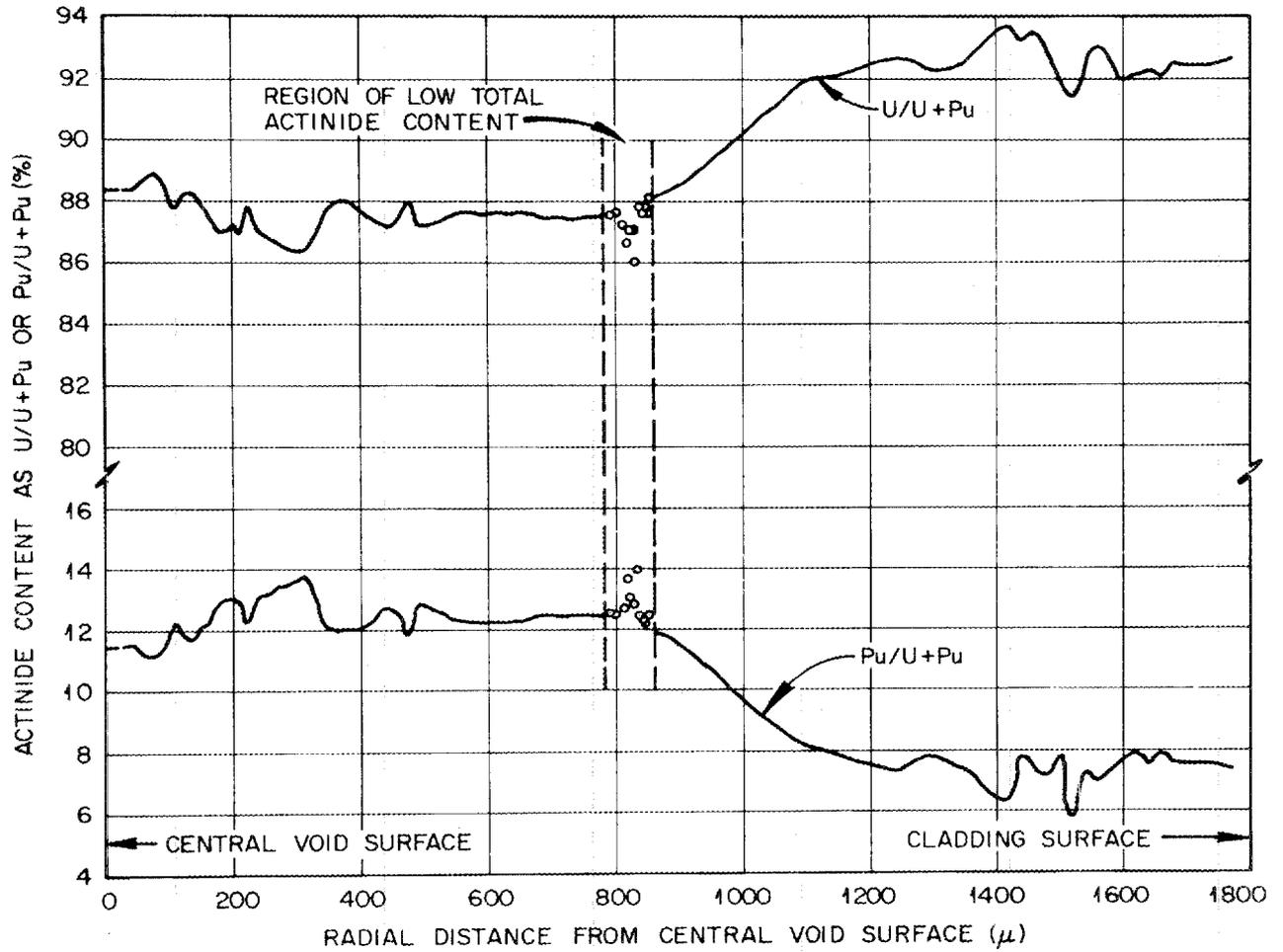


Fig. 14. Microprobe Analysis of  $U_{0.85}Pu_{0.15}O_2$  Fuel Pin with High Linear Heat Rate after Burnup to 6% FIMA.

claddings, one of which would act as an oxygen sink, distinctly different end-of-life plutonium contents were found in the intermediate columnar grain regions -- 31 compared with 26%, with the higher concentration being associated with the possibly lower oxygen activity.

#### DISCUSSION

The proposed mechanism for uranium and plutonium redistribution by a predominantly vapor phase transport mechanism is similar to that discussed by Bober<sup>24</sup> and Aitken.<sup>25</sup> The major differences are that in this case we show the effects of peak temperature on the equilibrium plutonium distribution and the effects of the compositional changes on restructuring forces (kinetics). The extent of actinide redistribution is initially established by the rapid decrease in vapor pressures with decreasing temperature. The composition of the solid state at various radial locations approaches a quasi-equilibrium when the uranium-to-plutonium and the oxygen-to-metal ratios in the solid are balanced with those in the vapor phase. The variation of equilibrium oxygen-to-metal with plutonium content is supported by the experimental observations of Ohse and Olson<sup>26</sup> and J. E. Battles et al.<sup>27</sup> who report differing oxygen-to-metal ratios for vapor pressure minimums (congruent vaporization) in out-of-pile experiments with two different compositions of solids. The pressure minimums occur at  $(U_{0.8}, Pu_{0.2})O_{1.92-1.93}$  and  $(U_{0.85}, Pu_{0.15})O_{1.97}$ . Since the total vapor pressures at all oxygen-to-metal ratios less than approximately 1.99 are essentially controlled by the partial pressure of  $UO_2$  (refs. 26 and 27), and this decreases with decreasing oxygen-to-metal ratios in the solid, the pressure gradient will be reduced as the solid compositions change in the higher temperature regions to higher plutonium

contents and lower oxygen-to-metal ratios. During restructuring the lowering of temperatures due to increased thermal conductivity resulting from fuel densification will lower the vapor pressure. The compositional changes will further lower these pressures so that the vaporization-condensation process essentially reaches equilibrium. The closing off of channels for vapor movement with densification will also reduce the possibilities for vaporization-condensation; but porosity sweeping up the temperature gradient will continue to provide a path along which such a process might proceed.

Even without the alternate possibility of thermal diffusion, the change in oxygen-to-metal ratios with burnup will provide a driving force for continued redistribution by the vaporization-condensation process. As the oxygen activity increases, the vapor pressures at all temperatures will also increase; and new quasi-equilibrium compositional distribution requirements will develop.

#### CONCLUSIONS

The analysis of our observations to date may be summarized as follows:

1. We have clearly defined vaporization-condensation as the principal process for restructuring in  $(U,Pu)O_2$  fuels for operating temperatures above  $1700^\circ C$  (the columnar grain-growth region).

2. Sintering processes and grain growth were shown to occur at all temperatures observed down to approximately  $900^\circ C$ , but these processes are of secondary importance in early-life restructuring.

3. Early-life restructuring has a distinct time dependence, which is an inverse function of the fuel operating temperatures. At a center temperature of 2000°C the restructuring is incomplete even after 28 days of full power operation.

4. The vaporization-condensation process can account for actinide redistribution. Either uranium or plutonium can concentrate in the fuel at the highest temperature, depending on the fuel operating temperature and probably on the oxygen activity in the gas phase.

#### ACKNOWLEDGMENT

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