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Transport Phenomena in Mixed Oxide Fuel Pins

by

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Abstract

Transport phenomena occuring in fast reactor mixed oxide fuels and their effect on the fuel pin behaviour under irradiation are discussed. In the initial phase transport of fuel constituents through evaporation-condensation processes and transport of oxygen via the gas phase and solid state thermal diffusion take place. Both processes depend strongly on the O/(U+Pu) ratio. With increasing burnup redistribution of fuel constituents by thermal diffusion becomes appreciable, which produces a continuously growing effect. Volatile fission poducts are transported through cracks and interconnected porosity channels toward the inner cladding surfaces. Soluble fission products migrate by thermal diffusion in the matrix and by evaporation condensation processes during healing of cracks. Insoluble solid fission products are moved with migrating pores.

Plutonium is transported to the fuel center producing an increase in central temperature of about 100°C in pins with a power rating of 500 W/cm. Oxygen transport processes establish a radial oxygen distribution with a nearly stoichiometric condition in the fuel periphery even for strongly hypostoichiometric mixed oxides. Volatile fission products together with the oxygen at the fuel periphery cause the attack on the cladding.

Transportprozesse in Mischoxid-Brennstäben

Zusammenfassung

Es werden Transportprozesse, die in Mischoxidbrennstäben schneller Reaktoren ablaufen, diskutiert und ihr Einfluß auf das Verhalten der Brennstäbe unter Bestrahlung beschrieben. In der Anfangsphase der Bestrahlung erfolgt eine Verschiebung der Brennstoffkomponenten durch Verdampfungs- und Kondensationsvorgänge und ein Transport von Sauerstoff in der Gasphase und durch Thermodiffusion in der festen Matrix. Beide Prozesse hängen stark vom O/(U+Pu)-Verhältnis ab. Die Thermodiffusion der Brennstoffkomponenten ruft einen kontinuierlich wachsenden Entmischungseffekt hervor, der mit zunehmendem Abbrand an Bedeutung gewinnt. Flüchtige Spaltprodukte werden über Risse oder Kanäle, die durch offene Poren gebildet werden, zur inneren Hüllwand transportiert. Gelöste Spaltprodukte wandern durch Thermodiffusion in der Matrix und durch Verdampfungs-Kondensations-Prozesse beim Ausheilen von Rissen. Unlösliche feste Spaltprodukte werden zusammen mit wandernden Poren verschoben.

Plutonium wird zum Zentrum des Brennstabes hin transportiert und ruft bei einer Stableistung von 500 W/cm einen Anstieg der Zentraltemperatur von ca. 100°C hervor. Die Transportprozesse des Sauerstoffs erzeugen auch in stark unterstöchiometrischem Mischoxid eine radiale Sauerstoffverteilung mit einem nahezu stöchiometrischen Zustand an der Brennstoffperipherie. Die flüchtigen Spaltprodukte verursachen zusammen mit dem Sauerstoff in der Brennstoffperipherie die Hüllrohrinnenkorrosion.

I. Introduction

Nuclear fuel materials are subjected to very high temperatures and steep temperatures gradients. Due to these extreme conditions various transport processes take place involving also ions and atoms which have relatively low mobilities in the fuel matrix. The known important transport processes are

a) Thermal diffusion in the solid state
b) Redistribution by evaporation-condensation processes in cracks and voids and during the migration of voids
c) Transport through the gas phase caused by equilization of the carrier gas pressure along cracks and interconnected porosity channels.

We distinguish the transport processes with regard to the physical mechanisms which cause the processes and with regard to the periods in which the processes generate a quasi steady state of redistribution. At first we describe the short term processes.

II. Short term processes

We consider three short term processes. The first is the redistribution of uranium and plutonium due to migrating voids during the initial phase of restructuring, the second is the redistribution of oxygen which achieves a quasi steady state in about one day and the third is the migration of the volatile fission products released through cracks and interconnected porosity channels to the cladding surface.

1) Redistribution of uranium and plutonium by vapor transport

The vapor transport of pores and voids in the temperatur gradient of a reactor fuel pin is accompanied with redistribution of uranium and plutonium if one of the fuel

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constituents evaporates preferentially. At temperatures above 1700°C the vapor transport of pores is a rapid process which can cause a significant redistribution during the short period of the initial restructuring of the fuel.

Since the redistribution of uranium and plutonium in migrating pores depends on the pressure of the vapor species over the uranium plutonium mixed oxide we have to consider the evaporation behavior of the fuel. Thermodynamical calculations show these pressures to be strongly dependent on the plutonium valency in the mixed oxide /1/. Experimental investigations of the evaporation behavior /2/ and observations of evaporation phenomena /3/ confirm the result of the calculations which show a preferential evaporation of uranium oxides above plutonium valencies of about 3.6 and a preferential evaporation of plutonium oxides at lower plutonium valencies.

The redistribution of uranium and plutonium in the fuel due to vapor transport of voids can be described in the following way. At the beginning of migration of each void there is an accumulation of the more volatile fuel constituent at the cooler side of the void. Parallel to this accumulation an enrichment of the less volatile constituent develops in a thin zone at the hotter side of the void. This zone moves up the temperature gradient together with the void while the accumulation of the more volatile component is left in the initial position of the void. Thus there is a tendency to enrich the less volatile constitutent in the center of the fuel and to accumulate the more volatile constituent in the cooler fuel regions where pore motion begins.

Considering the valency of 3.6, i.e. O/M=1.96 in $(U_{0.8}Pu_{0.2})O_{2-y}$, at which a turn in evaporation behaviour occurs we see that in mixed oxide fuels above this turning point an enrichment of plutonium and below this point an enrichment of uranium takes place around the central cavity. Figure 1 shows an enrichment of plutonium in the fuel center. It is a micrograph and an alpha autoradiograph of the cross section of a fuel pin irradiated in the RAPSODIE reactor. The initial mean plutonium valence was 3.93, i.e., uranium oxides evaporated preferentially. Besides the plutonium enrichment around the central cavity an enrichment of uranium can be seen in partially healed cracks /4/. If such radial cracks due to power cycling of the reactor appear repeateatly and heal subsequently by evaporation condensation processes this can contribute to the redistribution of the uranium and plutonium.

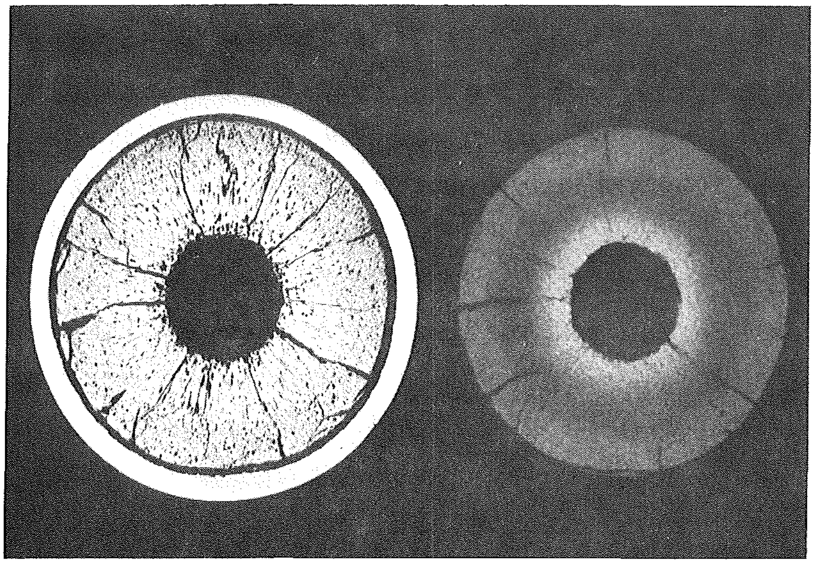

Fig. 1 Cross section and autoradiograph of a fuel pin irradiated in the RAPSODIE reactor. Burnup 5.77%, initial O/M=1.99, Pu/(U+Pu)=0.276/4/.

Microprobe analyses of fuel pins with central temperatures below 2000^{o}C indicate that redistribution by preferential evaporation occurs mainly in the initial irradiation period. No marked dependence of burnup could be observed in this cases where diffusion of uranium and plutonium is insignificant due to the low fuel temperatures /5/. However in a survey of the examination data of fast reactor fuel pins a clear dependence of the enrichment in plutonium on the O/M ratio was found at the fuel element centerline. The maximum enrichments were 65% at the centerline of nearly stoichiometric fuels /6/.

2) Redistribution of oxygen

We restrict our consideration to hypostoichiometric mixed oxide fuels which are favored because of their better compatibility with cladding materials.

Considerations on oxygen transport processes in the temperature gradient which occur in hypostoichiometric mixed oxide fuel pins of fast reactors reveal a strong reduction of the O/(U+Pu) ratio around the central cavity and a nearly stoichiometric condition near the fuel periphery /7-9/. Recent postirradiation examinations of mixed oxide fuel pins irradiated in the EBR-II reactor show the predicted oxygen distribution with the reduction in the inner dense fuel region /10/. The estimation of the radial oxygen distribution in the fuel pin bases on microprobe analysis of the local ratios of MoO_2 dissolved in the mixed oxide matrix and Mo metal accumulated in pores and voids together with the fission product metals Tc, Rh, Ru and Pd. We shall compare the results of the postirradiation examinations with oxygen distributions calculated from thermal diffusion.

In the dense inner fuel region the transport models of oxygen, which base on the equalization of the CO/CO_2 and H_2/H_2O ratios via the gas phase /7/ or on a combined gas/solid state mechanism /8/, are not applicable since the steady state of oxygen distribution establishes rapidly through the solid matrix in this range with temperatures above 1500°C /11/.

Since a solid state transport process must be responsible for the redistribution of oxygen observed in the dense fuel region we calculate the oxygen distribution on the basis of thermal diffusion in the solid state. This is performed with a heat of transport of oxygen vacancies of

$$Q_v^* = -(30 \pm 15) \text{ kcal/mol}$$

found in a thermal diffusion experiment with $(U_{0.85}Pu_{0.15})O_{2-y}$ in the temperature range of 1900 to 2350°C with a mean O/(U+Pu) ratio of 1.98/26,9/. The calculation of the transport of vacancies is advantageous because the vacancies can be assumed to be in a dilute solution in the oxygen sublattice. It corresponds to an equivalant transport of oxygen ions in the opposite direction. So Q_v^* allows us to calculate the oxygen distribution. Since the oxygen vacancies migrate rapidly we can restrict the calculation to the steady state distribution.

The steady state oxygen distribution in the radial direction of a fast reactor fuel pin can be discribed with the equation

$$\frac{1}{y}\frac{\text{grad } y}{\text{grad } T} = -\frac{Q_v^*}{RT^2} \qquad (1)$$

which holds for a dilute solution of oxygen vacancies in the mixed oxide /9/. In Eq.(1) y=2-O/(U+Pu) means the deviation from the stoichiometric condition, T is the absolute temperature, R the universal gas constant. Q_v^* represents the heat of transport of oxygen vacancies in hypostoichiometric mixed oxides, which includes the influence of the thermoelectric potential caused by the temperature gradient.

The result of the calculation with Eq.(1) is shown in Fig.2 for a $(U_{0.8}Pu_{0.2})O_{2-y}$ fuel with different stoichiometric conditions at the fuel periphery at which a temperature of 1150 K is assumed. Equation (1) reveals the oxygen

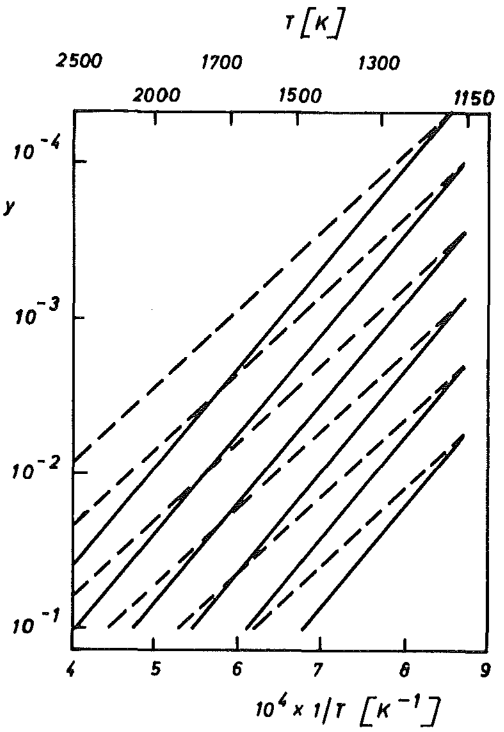

Fig. 2 y as a function of 1/T calculated from thermal diffusion for different stoichiometric conditions at the fuel periphery related to $(U_{0.8}Pu_{0.2})O_{2-y}$ fuel with a surface temperature of 1150 K. Solid lines: $Q^* = -30$ kcal/mol, dashed lines: $Q^* = -23$ kcal/mol.

distribution to depend only on the temperature. The slopes of the straight lines in the log y/T^{-1} diagram of Fig.2 are proportional to Q_V^*.

Now we turn to the comparison of the results of the post-irradiation examinations /10/, represented by the circles in Fig.3, with the calculated oxygen distributions. The diagram shows the O/(U+Pu) ratio as a function of a relative fuel radius based on the temperature distribution given for the irradiation experiment /10/. The solid and the

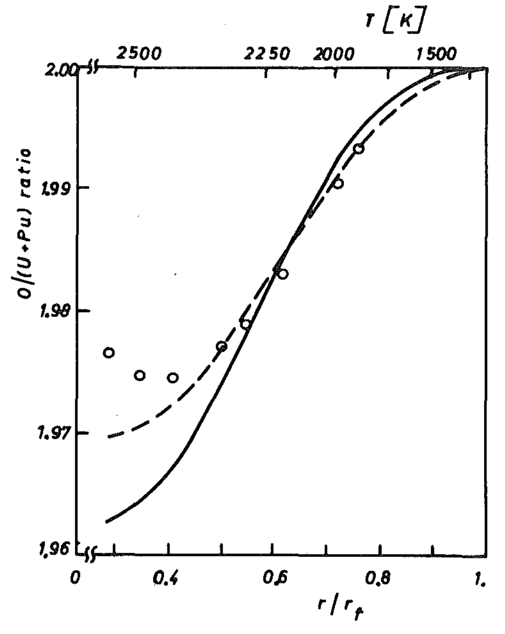

Fig. 3 Comparison of results of postirradiation examinations of an EBR II fuel pin (circles) (4) with radial oxygen distributions calculated from thermal diffusion with $Q^* = -30$ kcal/mol (solid line) and -23 kcal/mol (dashed line). Fuel: $(U_{0.8}Pu_{0.2})O_{2-y}$, mean O/(U+Pu) ratio: 1.991, fuel surface temperature: 1150 K.

dashed line represent oxygen distributions calculated for a mean O/(U+Pu) ratio of 1.991 with values of Q^* of -30 and -23 kcal/mol, respectively. We see, the line calculated with $Q^* = -30$ kcal/mol does not fit the distribution estimated from the postirradiation examinations. But a fairly good approximation is achieved with a mean effective heat of transport for oxygen vacancies of

$$Q^* = -23 \text{ kcal/mol}$$

which can serve to calculate the radial variation of the O/(U+Pu) ratio in reactor fuel pins. The rise in the O/(U+Pu) ratio represented by the circles in the vicinity of the central cavity in Fig.3 cannot be explained by a model of oxygen redistribution in a temperatur gradient, but the influence on the fuel pin behavior of this deviation from the calculated curve is negligible.

Although the dependence of Q_V^* on the temperature and on the O/(U+Pu) ratio is not known the mean value of -23 kcal/mol which allowed the fit shown in Fig.3 can serve to calculate roughly oxygen distributions in fuel pins. If values of Q_V^* are known as functions of temperature and O/(U+Pu) ratio more detailed calculations are possible.

3) Transport of volatile fission products

The volatile fission products cesium, tellurium, selenium rubidium and iodine are transported down the temperature gradient through vapor transport via interconnected porosity channels and cracks. They cannot be transported by solid state diffusion in the unrestructured fuel region since they are relatively immobile in the solid mixed oxide matrix.

The pressure of the saturated vapor of the volatile fission products are high at temperatures above 1200°C (c.f.Tab.1) and allow high transport rates if there is a connection between the high temperature zones, where the vapor molecules are released, and the cladding surface. Postirradiation examinations show that cesium is released out of the zone of columnar grain growth and is transported through interconnected porosity channels and cracks to the cladding surface while it is retained in the outer fuel region in closed pores and voids /13-15/.

Tab. 1 Saturation pressures (in atm) of volatile fission products /12/

T °C	Cs	Te	Se	Rb	I
600	0.37	0.01	0.31	0.44	(73)
800	2.03	0.15	(3.6)	3.1	
1000	(6.25)	1.0	(19.5)	11.0	
1200	(13.7)	(3.7)	(66.3)	(26.9)	
1400	(24.5)	(9.2)	(168.5)	(51.7)	

( ) extrapolated values

III. Long term processes

Long term transport processes which are controlled by diffusion mainly concern the migration of relatively immobile ions or atoms in the solid state; but we have also to consider the influence of the continuously growing amount of fission products and of the change in the oxygen potential on the transport processes during burnup.

1) Thermal diffusion of plutonium and uranium

Because of the low mobilities of uranium and plutonium significant redistribution of uranium and plutonium by thermal diffusion is generated only at temperatures above 1900°C. The transport of plutonium and uranium is determined by the heats of transport Q^*, the direction of the transport by the sign of Q^* /16/. Since the steady state distribution cannot be reached in the life time of the fuel pin the concentration distribution must be calculated with the flux equation. The flux of plutonium ions J_{Pu} - derived under specific assumptions applicable to mixed oxide fuels /9/ - is given by the equation

$$J_{Pu} = -nD_{UPu}\left[\nabla x_{Pu} + x_U x_{Pu}\frac{Q^*_{UPu}}{RT^2}\nabla T\right] \qquad (2)$$

where Q^*_{UPu} is the process heat of transport defined by

$$Q^*_{UPu} = \frac{D_{Pu}}{D_{UPu}}(Q^*_{Pu} - H_v) - \frac{D_U}{D_{UPu}}(Q^*_U - H_v) \qquad (3)$$

and D_{UPu} means the chemical interdiffusion coefficient

$$D_{UPu} = x_{Pu}D_U + x_U D_{Pu} \qquad (4)$$

The other quantities appearing in the equations are:

n = total number of atoms per unit volume of the solid,

x_U, x_{Pu} = molar fractions of uranium and plutonium oxides, respectively,

R = unsiversal gas constant,

T = absolute temperature,

D_U, D_{Pu} = chemical diffusion coefficients of uranium and plutonium, respectively,

Q^*_U, Q^*_{Pu} = molar heats of transport of uranium and plutonium, respectively,

H_v = molar enthalpy of formation of vacancies.

Starting with a uniform concentration distribution, the development of the concentration profile of plutonium can be calculated numerically with the equation of continuity, provided that the diffusion data and the heats of transport are known /9/.

The calculation is carried out with a suitable set of data of Q^*_{UPu} and D_{UPu} . The process heat of transport is assumed to be /17/

$$Q^*_{UPu} = -35 \text{ kcal/mol}$$

and the chemical interdiffusion coefficient (4)

$$D_{UPu} = 0.34 \exp(-111\,000/RT)\ \mathrm{cm^2/sec}$$

for the high temperature region. The temperature distribution in the fuel pin is obtained from a multizone formula taking into account the change in heat source distribution due to the plutonium transport to the fuel center (4).The result of the calculation for fuel pins with power ratings of 500W/cm and 600W/cm is given in Fig.4. The plutonium profiles obtained reveal that after longer irradiation periods a distribution is achieved which varies only slowly as a function of time, i.e. a quasi steady state is established. The calculated redistribution effects are in reasonable agreement with the effects found by microprobe analysis of irradiated fuels /14, 18-20/.

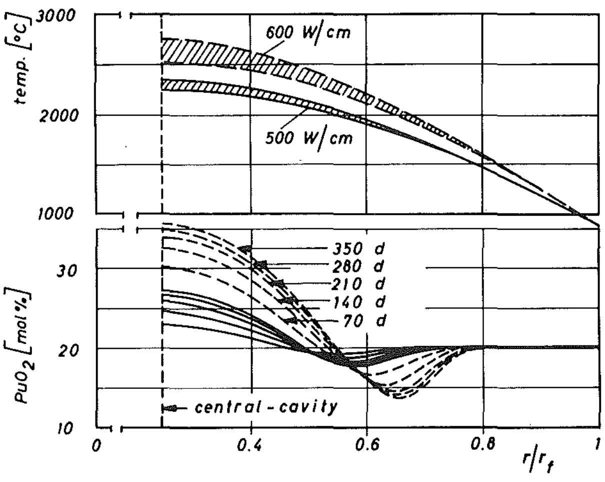

Fig. 4 Calculated plutonium distributions and temperature profiles in mixed oxide fuels containing 20 mol% of plutonium for irradiation periods of 70 to 350 days. The solid lines are related to a power rating of 500 W/cm, the dashed lines to 600 W/cm. Fuel radius: $r_f = 0.3$ cm.

The upper diagram shows the temperature distribution in the fuel pin and the influence of the plutonium-uranium redistribution on this distribution. The shadded area depicts the shift in the temperature distribution due to the redistribution of the actinides during burnup.

2) Transport of insoluble fission products

Insoluble fission products are released into pores and voids and are distributed in this form of small segregations over the region of equiaxed and columnar grains /14,20/. The inclusions range in size up to a few microns in diameter. They move together with the small pores up the temperature gradient. Corresponding to the size of the pores which contain the inclusions, the migration process bases on solid state diffusion of the surrounding matrix atoms. Thus, the transport of insoluble fission products is a long term process and therefore we can expect to find numerous metallic inclusions also in the high temperature region /10, 14, 20/. An indication of the migration of insoluble fission products to the fuel center was given by a post irradiation examination in which an accumulation of ruthenium was found in the central cavity /21/.

3) Influence of burnup on redistribution

One simple effect of burnup is the continuously growing mean concentration of all fission products. It turns the short term process of transport of volatile fission products in a long term process which lasts through the whole irradiation period. So we have a continuous transport rate which is discontinuously enhanced if new cracks arise due to a power cycling of the reactor.

A significant influence of burnup can be noticed on the radial oxygen distribution in the fuel pin. The mean O/M ratio of the fuel shifts in time to higher values without a significant change in the peripheral O/M ratio. The condition at the fuel periphery remains nearly stoichiometric when the fuel is hypostoichiometric in the interior of the pin. Only after a mean O/M ratio of about 2.00 is reached a more significant change in the peripheral stoichiometric condition takes place.

Under the assumption that no oxygen is consumed by the cladding the change in the mean plutonium valence v_{Pu} of the hypostoichiometric fuel is given by

$$v_{Pu} = \frac{v^0_{Pu} x_{Pu} + 4b\left[a + f_U - \Sigma_k S_k (f_U Y^U_k + f_{Pu} Y^{Pu}_k)/200\right]}{x_{Pu} + b(a - f_{Pu})} \qquad (6)$$

v^0_{Pu} = initial mean plutonium valence,

b = burnup (atomic fraction)

a = number of plutonium atoms produced by conversion, divided by the number of all fissions,

f_U, f_{Pu} = number of fissions of uranium and plutonium, respectively, divided by the number of all fissions,

Y^U_k, Y^{Pu}_k = number of atoms of the kth fission product related to 100 fissions of uranium and plutonium, respectively,

S_k = ratio of oxygen to metal atoms of the kth fission product oxide.

The yields of fission products Y^U_k and Y^{Pu}_k represent only those fission products which form oxides with oxygen potentials lower than that of the fuel.

The change in the mean plutonium valence is calculated with Eq.(6) as a function of burnup for the hypostoichiometric fuels. The values

$$f_{Pu} = 0.87,\ f_{U} = 0.13 \text{ and } a = 0.33$$

were roughly estimated from the RAPSODIE flux spectrum for a fuel with 20mol% of plutonium oxide using the nuclear cross sections of the actinide isotopes in the fuel /24/. For the calculation S_k is taken to be 1 for strontium and barium, 2 for zirconium and variable between 1.5 and 2 for the rare-earth elements, depending on the oxygen potential in the mixed oxide fuel. Fission product yields for fast fission of Pu-239 and U-238 after 1 year of decay /25/ are used ~~because the uranium was enriched with 83% of U-235~~. Fig.5 shows the calculated mean plutonium valences as a function of burnup for fuel pins with O/M ratios of 1.96 to 1.99. We see that all fuels gradually reach the stoichiometric state and are stabilized finnaly at an oxygen potential established by the formation of molybdenum oxide.

Recent post irradiation examinations give an indication that in mixed oxide fuel pins with stainless steel claddings the mean O/M ratio is established at 1.991 in $(U_{0.8}Pu_{0.2})O_{2-y}$ which is assumed to be an effect of the fuel cladding interaction. That means, the O/M ratio should not achieve the mean value of 1.991 in an early stage of irradiation to prevent a strong attack on the clad.

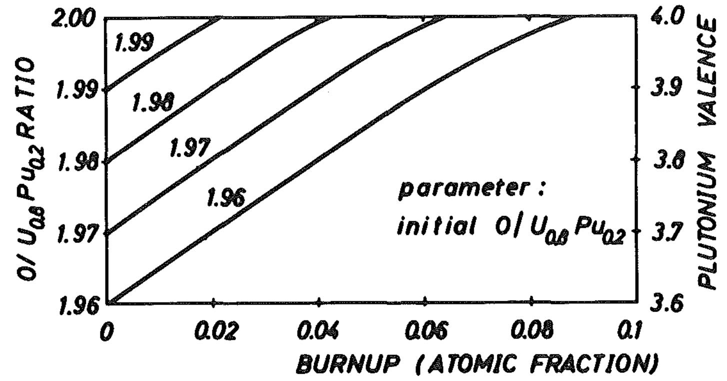

Fig.5 Calculated increase in the mean O/U+Pu) ratio and plutonium valence, respectively, of $(U_{o.8}\ Pu_{o.2})O_{2-y}$ fuel as a function of burnup in a fast reactor.

IV. Influence of the transport processes on fuel pin behavior

At first we consider the influence of uranium plutonium redistribution on the temperature profile. Fig.4 shows the variation in the temperature profile due to the enrichment of plutonium around the central cavity. In a fuel pin exposed to the extreme power rating of 600 W/cm an increase in central temperature of about 250°C must be anticipated due to an enrichment of about 75% of the initial plutonium concentration at the fuel centerline, calculated from thermal diffusion. At a power rating of 500 W/cm the calculated enrichment is about 30% which causes an increase in central temperature of 100°C. This redistribution effects caused by thermal diffusion develop after one year of irradiation under the condition underlying the calculation and are insignificant in the first stage of irradiation. The redistribution effect in the first stage of irradiation is determined by a vapor transport mechanism which was shown to generate in stoichiometric fuels plutonium enrichments up to 65% of the initial concentration / 6/. Thus an increase in central temperature up to about 200°C must be anticipated during the restructuring of the stoichiometric fuel because of the uranium-plutonium redistribution in voids migrating by vapor transport. This effect can be diminished by using hypostoichiometric fuels, e.g. $(U_{0.8}Pu_{0.2})O_{1.98}$ in which only enrichments of 20% to 45% were found / 6/. No redistribution by vapor transport appeared in $(U_{0.8}Pu_{0.2})O_{1.96}$. In such fuels only thermal diffusion contributes to redistribution which causes a slowly continuous increase in central temperature up to 100°C in maximum after 1 year of irradiation at power ratings of about 500 W/cm.

The influence of the redistribution of uranium and plutonium observed after irradiation and calculated from thermal diffusion on the doppler coefficient of fast reactor cores is negligible /22/.

The two main effects of oxygen redistribution on the fuel pin behavior concern the fuel cladding interaction and the redistribution of uranium and plutonium by vapor transport of voids. The calculations of the steady state oxygen distribution established by thermal diffusion show that even in strong hypostoichiometric mixed oxides the fuel periphery remains nearly stoichiometric while the fuel center is strongly hypostoichiometric. Fig.6 depicts the calculated oxygen distributions as function of the relative fuel radius for the typical temperature distribution of a mixed oxide fuel pin of a fast reactor. Already at a mean O/(U+Pu) ratio of 1.98 in $(U_{0.8}Pu_{0.2})O_{2-y}$ the O/(U+Pu) ratio in the columnar grain region is so low that no redistribution of uranium and plutonium by vapor transport takes place. Since the mean O/(U+Pu) increases in time with the burnup going on, the O/(U+Pu) in the center can increase to values at which redistribution by vapor transport becomes possible; but after restructuring of the fuel the contribution of vapor transport to redistribution is less significant. Thus, because of the redistribution of oxygen, we can prevent the fuel from significant redistribution of uranium and plutonium by vapor transport already by using a fuel with $(U_{0.8}Pu_{0.2})O_{1.98}$.

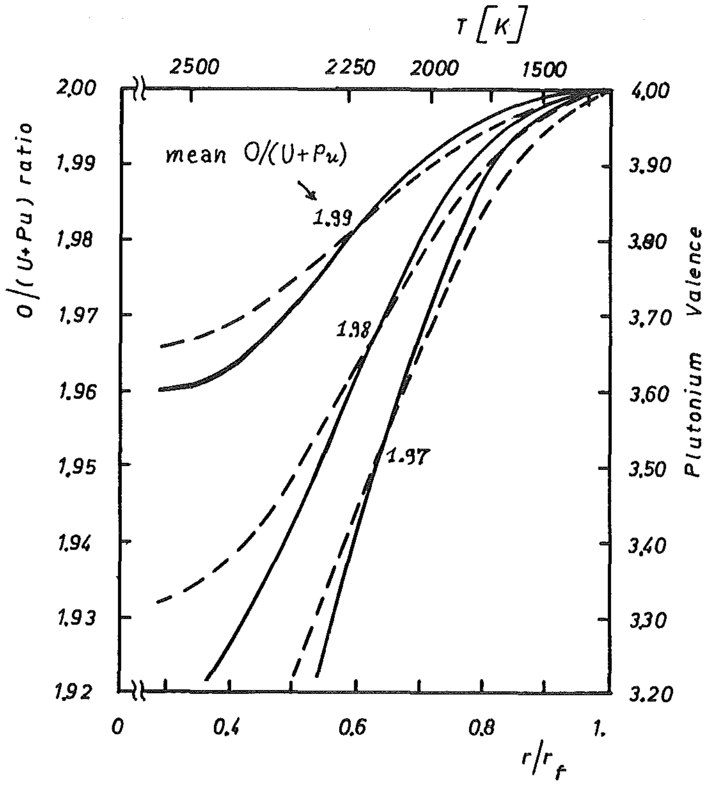

Fig. 6 Caclculated radial variation of the O/(U,Pu) ratio in $(U_{0.8}Pu_{0.2})O_{2-y}$ related to a fuel radius r_f=0.3 cm, Solid lines : $Q^*= -30$ kcal/mol, dashed lines: $Q^*= -23$ kcal/mol .

It is well known that the oxygen concentration at the fuel periphery influences strongly the fuel cladding interaction caused by volatile fission products /23/. The calculation of the oxygen distribution shows that over a wide range of hypostoichiometry no significant change in the oxygen potential at the fuel periphery takes place, since the periphery is always nearly stoichiometric. Recent post irradiation examinations /10/ indicate that oxygen is absorbed by the stainless steel cladding if the mixed oxide fuel exceeds the mean O/(U+Pu) ratio of 1.991. It seems that the stainless steel cladding stabilizes this stoichiometric condition where the calculated values of the O/M ratios in the center and at the periphery of the fuel are 1.97 and 1.99985 (theoretical), respectively. If we intend to prevent marked oxygen absorption of the cladding, accompanied with the atack of the volatile fission products, we have to start with a fuel of the composition of about $(U_{0.8}Pu_{0.2})O_{1.96}$. In this case the stoichiometric condition at which fuel cladding interaction becomes significant is achieved only in the last stage of irradiation.

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