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RESEARCH AT BATTELLE-NORTHWEST
ON TRANSPORT PROCESSES
IN OXIDE NUCLEAR FUEL

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RESEARCH AT BATTELLE-NORTHWEST
ON TRANSPORT PROCESSES IN OXIDE NUCLEAR FUEL*

by

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Chemistry and Metallurgy Division

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RESEARCH AT BATTELLE-NORTHWEST ON
TRANSPORT PROCESSES IN OXIDE NUCLEAR FUELS

J. A. Christensen

ABSTRACT

Current studies at Battelle-Northwest of mass and energy transport in fuels are directed toward coordinating precision laboratory measurements with in-pile experiments designed to yield specific materials property data. Results are applied to fuel systems design and behavioral model development. Emphasis has been on mixed uranium-plutonium oxides with lesser efforts on UO_2 , mixed nitrides, and uranium oxycarbides. Thermal diffusivities for various fuel materials have been measured at temperatures to 1700 °C. Results together with studies of associated thermodynamic properties, melting points, phase equilibria, electrical and thermoelectric properties, and materials synthesis methods, have been applied toward designing and interpreting a variety of irradiation experiments. Included among these have been: 1) measurements of stoichiometry and fabrication method effects on in-pile heat transfer in $(\text{U,Pu})\text{O}_{2+x}$ and UO_2 ; 2) determination of stoichiometry and heat rating effects on oxygen, plutonium, and fission fragment distributions in oxide fuels; 3) observations of relationships between void and fission fragment distributions and fuel thermal history; 4) characterization of irradiated mixed-oxide structures in terms of stoichiometry and heat rating; and 5) studies of dimensional stability.

Introduction

Ceramic fuel studies at Battelle-Northwest are directed toward developing improved materials and designs and understanding their performance limitations. This discussion relates to fuel performance and will be limited to oxide fuels since these are currently receiving the bulk of our attention.

Efforts described are sponsored by the United States Atomic Energy Commission under the Liquid Metal Fast Breeder Reactor Program and the Plutonium Utilization Program and are closely coordinated with Fast Flux Test Facility activities also in progress at Battelle-Northwest. The composite goal of the program may be summarized as to quantitatively anticipate the thermal and dimensional behavior of an oxide fuel core under any conditions it may experience in an LMFBR.

Irradiation Environments

Oxide fuels are interesting and complex materials - not because of strikingly unique properties but because of the unusual conditions to which they are exposed during use. These conditions are distinguished from more conventional applications in three important ways:

1. High temperatures (approaching or exceeding the melting point) are experienced for long periods of time, i.e., several months.
2. Extreme thermal gradients (as high as 8000 °C/cm) co-exist with the high operating temperatures.
3. The neutron flux and its associated fission events and ionizing radiation constitute a unique environment in terms of effects on mass and energy transport.

These conditions have the effects of providing both the potential and the driving force for a number of complicated, interrelated kinetic processes. Long times at high temperatures permit

favorable reactions to approach completion. The neutron flux has a similar influence in that it creates a population of dynamic defects which have an undefined temperature equivalence in stimulating transport processes. Stable defects also accrue from the irradiation environment. The most obvious of these are the fission products which may represent 15 to 20% of the total cation content of a fast fuel near the end of life. These high impurity contents can change important fuel properties in ways which vary continuously during the service life of the fuel. Structural defects, e.g., stable vacancy clusters, dislocation loops, etc., are also undoubtedly produced in the fuel during fissioning. These can have controlling effects on dimensional stability; but because they are much more difficult to observe and interpret than are the grosser manifestations of fuel irradiation, little is known about them.

Superimposed upon and interrelated with all of the thermal and flux effects are the effects of the thermal gradient. The steep thermal gradients, which reflect the low thermal conductivity of oxide fuels, provide the driving force for several processes which are fundamental in determining useful lifetimes and operating conditions. Most important of these is the redistribution of fuel constituents, oxygen in particular, to alleviate the chemical potential gradient set up by the thermal gradient. The resulting spacial variation in oxygen content is important because all fuel properties of technological interest, e.g., thermal conductivity, melting point, creep strength, vary significantly with O/M. These properties thus vary with initial O/M, heat rating, fuel size, burnup, impurity content, sheath material, etc. Similar arguments apply to redistribution of fuel cations and fission fragments. In addition to materials redistribution by thermal diffusion, movement of ions under the influence of the thermoelectric potential gradient is a potentially important transport process in fuels during irradiation. Neutron flux gradients in highly absorbing fuels may have similar effects.

The net picture is very complicated - materials with time and space varying compositions operating for long times at extreme temperatures in an irradiation environment effects of which are largely unknown and impossible to estimate with confidence. Multi-coupled-current mass transport processes activated by thermal and, perhaps, flux gradients determine the long-term behavior of these materials. These require empirical study in environments which can be specifically related to those in which they are used.

Direct application of laboratory results to such a complex system is rarely possible. A more productive approach is to combine out-of-reactor experimentation, which defines the separate processes, with irradiation experiments designed to determine interrelationships among processes. Identification of critical parameters and estimation of their interaction can be made tractable through use of the various analytical material behavior models being developed.

Categorization of separate transport processes in fuels is not realistic because of the almost universal interaction between these processes. For clarity, an arbitrary grouping of the current program into 1) fuel restructuring; 2) redistribution of fuel constituents; 3) energy transport; 4) fission gas effects; and 5) analytical modeling is employed in the following discussion.

Fuel Restructuring

Materials science is primarily concerned with the relationship between structure and properties. In oxide nuclear fuels, the structural changes which derive from service are especially interesting. The bulk of existing fuel materials information consists, in fact, of observations of these structural changes.¹ Studies of structural changes are useful because these changes are easily observed, direct manifestations of the transport mechanisms which control all of the kinetic processes of practical importance in fuels. General application of the results of these studies is, however, usually frustrated by the difficulty in quantitatively

correlating structure type with a specific property. To compensate this, the intermediary parameter "temperature" is evoked to cross-correlate structure with specific properties. This represents only an approximation of course since other variables function independently of, as well as in interaction with, temperature in determining structure-property relationships (e.g., time, temperature gradient, neutron flux, fission rate, chemical composition, impurity content, pressure). Also, it is difficult to precisely assign temperatures to discrete structural features - partly because of interference from the above effects and partly because accurate in-reactor fuel temperature distribution measurements are impeded by the high temperature and steep thermal gradients. Enough information has been accumulated, however, to permit the requisite structure-temperature-property correlations for some important properties.

Examination of a representative cross-section of a highly rated cylindrical oxide fuel element reveals as many as six regions with distinctly different grain morphologies (Figure 1). An equal number of mobile or stable pore types can also be identified. Grain morphology and associated pore type can be roughly correlated with temperature as has been done in Table I. The correlation is crude because grain boundary and pore movements are kinetic processes. Structure thus varies continuously with time. Table I applies to the steady-state achieved in a UO_2 -25 wt% PuO_2 fuel shortly after irradiation is initiated. Superficially, analyses of such a complex system of structures appears overwhelming. In fact, however, only two processes appear to be very important: 1) nominally equiaxed grain growth involving mass transport by point defects and 2) formation of grains by pores traveling along the thermal gradient. The first process can be represented by an Arrhenius relationship with an activation energy of approximately 125 Kcal/mole². The second process involves movement of pores up the thermal gradient by oxide vaporization from the hot side of the pore and condensation on the cold side³. This mechanism is unimportant at low temperatures because the vapor pressure is negligible. Vapor-deposited-grain growth rate also increases

FIGURE 1
 Structure Development in Highly Rated Mixed Oxide Fuels
 Material is Sintered, 0.257 in. Diam (0.75U, 0.25Pu)O₂.00 with an Initial Bulk
 Density of 94% T.D. Irradiated to 6x10¹⁸ f/cc with a Fuel Surface Temperature
 of 900°C and a Centerline Temperature Slightly Above Melting

TABLE I. Temperature, Grain Type, Pore Type Correlation for Lightly Irradiated, Highly Rated Oxide Fuels*

Temperature	Grain Morphology	Grain Size	Pore Morphology	Approximate Pore Size	Illustrated By
900 °C- 1200 °C	Equiaxed, As-sintered	~20 μ	Equiaxed, Random Distribution	5 μ	Figure 1F
1200 °C- 1700 °C	Equiaxed, Normal Grain Growth	20 μ -40 μ	Radially Migrating or Grain Boundary Spheres	5 μ	Figure 1E
1700 °C- ~2000 °C	Columnar Grains	20 μ x 80 μ	Lenticular	3 μ x 50 μ	Figure 1D
~2000 °C- Melting	Radially Elongated	150 μ x 600 μ	Random Spheres, Chevrons	10 μ -30 μ 1 μ x 50 μ	Figure 1C
Molten Region,** Outer Portion	Radially Elongated	150 μ x 600 μ	No Pores		Figure 1B
Molten Region,** Central Portion	Radially Elongated		Random Tubular	20 μ x 100 μ	Figure 1A

* Observations made on 94% T.D., 0.26 in. diameter UO_2 -25 wt% PuO_2 irradiated at ~30 kW/ft to 6×10^{18} f/cc. Observations apply in general to other oxide fuels but temperatures and dimensions may differ.

** Grains in these regions are mutually epitaxial from one region to the next or may extend through all three regions.

exponentially with temperature, however, and with an activation energy equal to the enthalpy of vaporization which is significantly higher than the activation energy for ordinary grain growth⁴. At some intermediate temperature vaporization controlled grain growth thus becomes the dominant process. The rate of pore movement by vaporization-condensation is also a function of the thermal gradient. The gradient decreases to zero at the thermal center of a fuel core, thus in the central region of the fuel, diffusion controlled grain growth again predominates. This competitive interaction between diffusion and vaporization controlled grain growth, illustrated diagrammatically by Figure 2, gives rise to the familiar fuel structure consisting of central and circumferential regions of normal grain growth separated by an annulus of narrow columnar grains formed by migrating pores. Pore formation and migration requires a source of pores. In a virgin fuel, on the order of 5% porosity seems to be required to yield significant columnar grain growth⁵. Thermal stresses established during thermal cycling of fuels which are not too heavily restrained cause circumferential cracks which serve as regenerative sources of migrating pores. Fission gas bubbles may function in this capacity also.

A second type of mobile porosity is shown in Figure 1E. Small ($<5\mu$) spherical pores are observed to form and migrate up the thermal gradient in the equiaxed grain region adjacent to the outer edge of the columnar grains. These are not fission gas bubbles - the specimen shown had a burnup too low to attribute the volume in the pores to fission gas. Many of these pores become trapped on grain boundaries but some escape and proceed into the matrix of the adjacent grain. These pores, which have been observed only in nearly stoichiometric urania-plutonia fuels of fast reactor compositions, are of a size appropriate for migration by a surface diffusion mechanism. They do not form new grains in their wake presumably because to do so would require an increase in grain surface area. These pores may be instrumental in fuel redistribution and fission product migration processes, and their transport characteristics can probably be related to fission gas

FIGURE 2
Interaction Between Different Growth Mechanisms to Yield
Columnar Grain Annulus Characteristic of High-Rated Oxide Fuels

bubble movements. They do not, however, cause apparently important structural modifications.

On a finer scale, fission gas porosity is the subject of intensive study. Transmission, replica, and scanning electron microscopy are being applied toward characterizing bubble size distributions in irradiated oxide fuels.

Redistribution of Fuel Constituents

In addition to restructuring, complex chemical redistribution occurs as a product of the high temperature, high thermal gradient irradiation environment. Redistribution of fuel components or fission products may occur by some of the same transport mechanisms responsible for grain restructuring or may reflect parallel processes which may or may not interact with restructuring. The potentially most important effect is oxygen migration. Thermodynamic calculations illustrate the feasibility of oxygen redistribution in fuels with an initial average O/M $\neq$ 2.00⁶. Since UO_2 - PuO_2 fast fuels characteristically are fabricated with O/M's less than 2.00 and irradiation to burnups of several percent causes a thusfar undefined shift in O/M, measurements of oxygen distributions characteristic of typical fuel operating conditions are required.

Radial sampling and analyses of irradiated UO_{2+x} specimens showed that when partial fuel melting occurred the excess component (uranium or oxygen) migrated toward the fuel center⁷. This was interpreted in terms of an expected increased solubility of the excess component in the melt relative to the solid which it contacted. A subsequent analysis considered the existence of small quantities of carbon - an unavoidable impurity - which on oxidizing creates an oxygen pressure which varies with radial position in the fuel because of the temperature dependence of the $\text{CO}_2 \rightleftharpoons \text{CO} + 1/2 \text{O}_2$ reaction⁸. In agreement with the experiments, this mechanism predicts an oxygen concentration gradient of the same sign as the thermal gradient for hypostoichiometric fuel and of opposite sign for hyperstoichiometric fuel. A critical test to differentiate between the two proposed mechanisms is to determine the direction of

the oxygen concentration gradient in a hyperstoichiometric fuel operating with no melting. This experiment was performed using $U_{2.02}$ with the results shown in Figure 3. Net movement of oxygen toward the fuel center occurred regardless of whether or not a molten phase was present. This eliminates the hypothesis that the melt functions as a sink in establishing composition gradients. Oxygen redistribution measurements for uranium-plutonium fuels have been limited to hypostoichiometric compositions because hyperstoichiometric materials are difficult to fabricate and have no direct practical use as fuels. The earliest results showed a slight outward migration in agreement with the analytical predictions⁹. Observations of microstructures developed in MO_{2-x} fuels during irradiation indicate a similar direction of oxygen migration¹⁰. Very recent measurements on UO_2 -25 wt% PuO_2 with an average O/M of 1.97 showed oxygen movement toward the fuel periphery also (Figure 3). It appears that the peripheral regions of an initially slightly substoichiometric fuel will approach stoichiometry during irradiation and that the central region will be correspondingly depleted. This is a favorable situation in that it tends to maximize the heat transfer capability of the fuel without impairing compatibility with the sheath. As fissioning progresses, the effective O/M is expected to increase which should cause progressive movement of the $MO_{2.00}$ constant composition line toward the center. If the average O/M increases above 2.00, the excess oxygen should concentrate near the fuel center. This is desirable for fuel-sheath compatibility but may cause a significant decrease in melting temperature. A crucial contemporary question concerns the extent to which fuel O/M changes during irradiation and the disposition of the excess of oxygen expected to be formed.

Cation redistribution also occurs during irradiation of mixed-oxide fuels; however, not to the extent observed for oxygen. Neither are the implications with respect to changes in important properties so severe as for oxygen redistribution. A convenient method for assessing plutonium redistribution is α -autoradiography¹¹.

FIGURE 3
Oxygen Redistribution During Irradiation of Non-stoichiometric
Oxides Both With and Without Central Melting

This technique together with microsampling has shown that molten fuel regions become enriched in Pu at the expense of the immediately adjacent, large grained oxide. In the absence of fuel melting, slow Pu migration to the center has also been observed, but not to an alarming extent in terms of effects on properties¹². On a micro scale, homogenization of initially, slightly heterogeneous mixed-oxide fuels has been studied for some time by autoradiography and more recently with the shielded electron microprobe¹³. The latter method is highly sensitive in detecting regions of varying Pu/U. Results show, as expected, that homogenization proceeds as a direct function of irradiation temperature.

Distribution and oxidation state of fission products are under intensive current study. Qualitative observation of fission product redistribution in highly rated fuels has progressed from the crude glass plate and photographic emulsion autoradiographs first employed at Hanford Laboratories in discovering the phenomenon¹⁴ through various stages of autoradiograph sophistication and microsampling technique development¹⁵. The latest innovation, developed in cooperation with LASL, has been microgamma scanning of irradiated fuel sections¹⁶. This technique is nondestructive, relatively inexpensive, and is capable of higher resolution than physical sampling methods. Figure 4 shows results of a typical fuel section survey for Ru¹⁰⁶ and Cs¹³⁷, two of the most mobile γ -emitting isotopes, along with a β - γ and an α -autoradiograph. The autoradiographs qualitatively illustrate the isotopic distribution and the central Pu enrichment discussed previously.

In general, it is observed that those isotopes which oxidize in preference to uranium or plutonium to non-volatile oxides and which dissolve appreciably in MO_2 do not redistribute markedly during irradiation. Zirconium-niobium, Ce, Y, La, and lanthanide rare earths are examples of such stable fission products. The most mobile fission products are those with high vapor pressure elemental states or which form unstable oxides, e.g., Cs and

FIGURE 4
Gamma Scanning and Microsample Determinations of Ru and Cs Distributions in
 UO_2 -2 w/o PuO_2 Vipak Fuel Irradiated to 9×10^{18} f/cc at 21 kW/ft Peak

Rb. A particularly interesting element with regard to fission product redistribution is ruthenium. Ruthenium metal is refractory (melting point ≈ 2500 °C) and has a vapor pressure about four orders of magnitude less than that of (U,Pu) O_2 . Ruthenium is thus not quickly ejected from the hotter fuel regions as is cesium for example. In addition, ruthenium oxide (Ru O_2) has a free energy of formation four to six times less than that of (U,Pu) O_2 thus Ru retains a zero valence state and does not go into solution in the oxide fuel matrix. Fission product ruthenium is thus generally retained throughout the fuel but largely as a second phase which is relatively more influenced by the transport processes which contribute to structural development than are those fission products which are in solution, e.g., Zr-Nb Ce, etc. Other common fission products which share these characteristics with Ru are Mo, Tc, Pd, and Rh¹⁷. Ruthenium is distinguished from these other elements in that both Ru¹⁰³ and Ru¹⁰⁶ are highly beta active - so much so that Ru is responsible for most of the distinctive features observed in a typical β - γ autoradiograph. The net result is that a relatively minor fission product (the combined fission yield for Ru¹⁰³ and Ru¹⁰⁶ from U²³⁵ fission is <4%) provides the largely illusory impression in autoradiographs that fission products in general undergo extreme redistribution during irradiation. One interesting manifestation of these effects is the much sharper contrast of β - γ autoradiographs from Pu²³⁹ enriched fuels relative to those from U²³⁵ enriched fuels. A major reason for the difference is that the ruthenium fission yield from Pu²³⁹ is a factor of three greater than from U²³⁵.

Fission product distribution, as detected by β - γ autoradiographs, is a useful tool in studying thermal performance of oxide fuels. The most reliable indicator of the extent of melting in highly rated fuels is a sharp discontinuity found on the autoradiograph⁷. Autoradiographs are also useful in characterizing fuel operating history. For example, rate of power reduction during the final thermal cycle of molten center oxide fuel cores determines the fission product distribution in the once-molten region¹⁸.

Ruthenium segregates to the fuel center as an inverse function of cooling rate and forms precipitates which seem to be associated with the surfaces of the porosity formed on solidifying the melt (Figure 5). This transport mechanism is thought to be responsible for the massive nuggets of Ru, Mo, Tc, Pd, and Rh sometimes found in the central void of high exposure fuel pins¹⁹. Figure 6 illustrates an extremely large nugget of this type²⁰.

An example of the interaction between solid state restructuring and fission product redistribution is shown in Figure 7. The two fuel pins shown were irradiated as part of the same assembly in precisely the same flux for approximately one day. The only difference between specimens A and B is that A had a sufficiently larger diameter than B to cause it to operate at a 20% higher heat rating. The assembly was discharged at a rate calculated to cause solidification of the molten zone in specimen A in 100 seconds. Some melting is evident in both specimens. Specimen A contains no narrow columnar grains presumably because they formed by lenticular void migration early in life and were subsequently annihilated by equiaxed grain growth. Specimen B has an abundance of columnar grains and their associated lenticular voids (Figure 1D is from the columnar grain region of this specimen). The β - γ autoradiograph from Specimen B shows fission product (principally ruthenium) relocation down the thermal gradient to the region at which columnar grain growth initiates. No redistribution of this sort is evident in the specimen with no columnar grains. Apparently fission products were transported by a vaporization-condensation process which yielded a net outward displacement of most of the ruthenium in a given volumetric unit for each lenticular void which passed inward through that unit. The vapor pressure of ruthenium is low enough to be consistent with such a mechanism. Quantitative analyses of this type can be applied to each of the major fission products and their extent of relocation up or down the thermal gradient computed as a function of time at temperature and concentration, i.e., burnup.

FIGURE 5
Cooling Rate Effects on Fission Product Distribution in
Molten Center UO_2 Fuels

FIGURE 6
Large Fission Product Metal Nugget in Central Void of UO_2 -25 W/O PuO_2 Fuel Pin
Irradiated in EBR-II to 19×10^{20} f/cc at ~ 30 kW/ft Peak; Longitudinal Section

FIGURE 7
Effect of Columnar Grain Growth on Fission Product
Distribution

Energy Transport

Inasmuch as the primary function of a nuclear fuel rod is as a heating element, the heat transfer characteristics of materials employed in its construction are of paramount concern. In the case of oxide nuclear fuels, the cumulative effort expended internationally in measuring and interpreting thermal conductivity is staggering. Probably more money and man-hours have been invested in studying heat transport in UO_2 (or $\text{UO}_2\text{-MO}_2^*$) than in any other material. There are two reasons for this:

1. In terms of properties, the low thermal conductivity of oxide fuels is their greatest single disadvantage. Fuel designs typically are limited by the heat ratings which can be achieved without exceeding unacceptably high temperatures. The thermal conductivity must thus be known very accurately so that arbitrary heat rating limitations may be minimized.
2. The principal thermal energy carriers in dielectric crystals such as oxide fuels are phonons. Lattice anharmonicity and therefore phonon mobility are directly proportional to the difference in cation and anion mass. This mass difference is maximized in the fissile oxides; and, as a consequence, their thermal conductivities are the lowest of the pure, crystalline oxides - roughly a factor of ten lower than the best of the oxide conductors. There are, however, other conduction mechanisms which are not impaired by this large difference in cation and anion mass. Because the phonon conductivity is low, these other mechanisms are relatively more important in contributing to the total thermal conductivity.

* M refers to any of a variety of cations including, principally, Pu, Th, and Zr.

Radiant transfer through the material and energy transport by charged particles, e.g., electrons or ions diffusing preferentially in a thermal gradient, have received the most study. Assuming the proper conditions, however, any conceivable mechanism of energy transport in solids might contribute sufficiently to the thermal conductivity to be of practical importance in the design of fuels.

In spite of all the possibilities, the current consensus is that for design purposes, phonon transport is the only practically important heat conduction mechanism in oxide fuels²⁵. Uncertainties still exist, however, as to the magnitude of the conductivity at temperatures much in excess of 2000 °C. Also, effects of irradiation, fabrication variables, and composition require further definition. These problems are being studied by both laboratory and in-reactor experiments.

The method most relied upon at BNW for absolute conductivity measurements is the laser flash diffusivity technique. This method measures diffusivities which must be multiplied by the volumetric specific heat for the same temperature to yield conductivities. Measurements of specific heats using drop-weight calorimetry are therefore also in progress.

Diffusivities have been measured for various oxide fuel materials at temperatures from well below room temperature to nearly 2000 °C²¹. A recent study concerned temperature and oxygen content effects on the diffusivity of UO_2 -25 wt% PuO_2 ²². For temperatures between 100 °C and 1600 °C and $1.93 \leq \text{O/M} \leq 2.00$, the reciprocal diffusivity obeyed the function:

$$1/\alpha = 0.11T + 1.08 \times 10^{+3}X + 4 \text{ sec/cm}^2$$

where T is in °C and X is the unknown in the value of the subscript in MO_{2-X} .

The corresponding reciprocal thermal conductivity, obtained from the preceding relationship by multiplying by the density and the specific heat is:

$$1/\lambda = 0.031T + 3.55 \times 10^2 X + 1.5 \text{ cm } ^\circ\text{C/w}$$

Comparison of these results with the best data for UO_2 and results of similar studies on mixed-oxides shows little difference between the materials.

Effects of varying Pu contents in UO_2 - PuO_2 solid solutions were also measured²³. Results show a small but consistent decrease in diffusivity up to 20 to 30 wt% PuO_2 and a more pronounced decrease for oxide containing 45 wt% PuO_2 . At 900 °C, the thermal diffusivity for UO_2 -20 wt% PuO_2 is approximately 20% less than that for UO_2 . At higher temperatures, this difference becomes smaller.

Laboratory measurements of thermal conductivity cannot be applied unequivocally to an oxide fuel. Composition gradients, fuel restructuring, and the accumulation of fission product impurities and structural defects alter the conductivity substantially. These effects are evaluated by experiments performed in-reactor using specimens which experience temperatures and thermal gradients typical of operating fuels. Effects of stoichiometry departures on the melting heat rating for UO_2 and UO_2 -25 wt% PuO_2 have been determined in this manner. For UO_2 with a fuel surface temperature of 500 °C, a 25% linear decrease in melting heat rating in going from $\text{O/U} = 1.90$ to $\text{O/U} = 2.15$ was measured⁷. In UO_2 -25 wt% PuO_2 with a 900 °C surface, varying O/M from 1.94 to 2.00, had no significant effect on melting heat rating²⁴. This range of O/M 's includes all values of interest from a mixed-oxide fuel fabrication point of view. It thus seems likely that O/M may be controlled on the bases of fuel - sheath compatibility and oxidation from fissioning without great regard for attendant effects on heat transfer.

The melting point is generally stipulated as the maximum permissible fuel temperature. Studies of melting are thus relevant to heat transfer in that peak allowable heat ratings reflect a combined consideration of thermal conductivity and melting point. Of most concern, are the possible effects of accumulated fission product impurities on end-of-life allowable

fuel heat ratings. Several relevant measurements have been made^{25,26,27} the most conservative of which²⁶ suggests a nearly 30% reduction in melting heat rating after 3×10^{21} f/cc exposure in a typical oxide fast fuel. The least conservative studies reported show a nearly negligible effect²⁷. The most recent study of irradiation effects on melting point shows no change at burnups to 2×10^{20} f/cm³ in UO₂ irradiated at <100 °C²⁸. Obviously, more precise methods of making melting point measurements on highly irradiated fuels are called for. The need is expected to be filled by thermal arrest methods currently being developed.

Electronic transport processes can influence both mass and energy transfer in uranium base, oxide fuels. These materials are electronic semiconductors, which can conduct heat by transport of the kinetic and potential energy of excited, conduction band electrons as these diffuse down the thermal gradient. A comprehensive study of UO₂ at temperatures to 2700 °C showed electrical conductivity to be extrinsic, p type to 1100 °C indicating that the material contained a slight amount of excess oxygen²⁹. Above 1100 °C, conductivity was intrinsic and obeyed the relationship:

$$\alpha = 3.57 \times 10^3 \exp (-1.15 \text{ eV/kT})$$

Measured conductivities in the vicinity of the melting point are high enough to predict a substantial increase in high temperature thermal conductivity if a simple band structure and optimum ambipolar diffusion of conduction band electrons are assumed³⁰. Such assumptions are not valid, however, and the bulk of irradiation experience with oxide fuels tends to deny large, high temperature increases in thermal conductivity.

Seaback coefficient measurements for UO₂ at temperatures to 2200 °C are consistent with the electrical conductivity results: below 1100 °C, the material is p-type while at higher temperatures it transforms to intrinsic, n-type³¹. The coefficients are not large, but the density of conduction electrons apparently does not

become sufficiently high to cause degeneration. As a result, the electrical potential across the radius of a typical oxide fuel increases continuously with surface-to-center ΔT to rather large maximum values - on the order of 2 volts. This potential gradient is sufficient to significantly influence ionic migration. Calculations have shown that the rate of diffusion of oxygen by this mechanism exceeds that by bulk thermal diffusion by approximately an order of magnitude³². Measurements of thermoelectric powers for $\text{UO}_2\text{-PuO}_2$ and extension of the analytical model to consider surface diffusion and vapor transport are required to assess likely effects in fast fuels.

Measurements of fuel-sheath heat transfer characteristics are providing particularly gratifying results. A recent study of xenon contamination effects on gap conductance showed surprisingly slight effects³³. Four $\text{UO}_2\text{-25 wt% PuO}_2$ fueled pins were evacuated and back-filled with one atmosphere of, respectively, 100% He; 60% He, 40% Xe; 40% He, 60% Xe; and 100% Xe. These were encapsulated in the same assembly which was then irradiated for four hours in a flat, uniform flux at approximately 35 kW/ft. Postirradiation measurements of extent of melting and grain growth (Figure 8) showed maximum gap conductance decreases of 15%, 25%, and 45% for 40%, 60%, and 100% Xe, respectively, based on a calculated conductance of $0.85 \text{ w/cm}^2 \text{ }^\circ\text{C}$ for pure He. No change in gap size was apparent in the irradiated pins.

Fission Gas Effects

The gaseous fission products are responsible for the principal stresses imposed upon a fuel element sheath. These stresses originate from two sources: 1) released gas exerts pressure which must be restrained entirely by the sheath and 2) retained gas precipitates as bubbles which are restrained by surface tension forces, the creep strength of the fuel, and the hoop strength of the sheath. In high burnup fuels, the latter process has the most serious implications since pressure from released gases can be relieved by incorporating larger plenum volumes into the fuel rod. Optimum fuel rod design requires a knowledge of what fraction of the fission gas produced is released and what fraction is retained as bubbles.

FIGURE 8
Xenon Effects on Fuel-Sheath Gap Conductance UO₂-25 W/O PuO₂ Irradiated at
35 kW/ft, 0.007 in. Diametral Cold Gap

Fractional gas release was measured for twenty, eight-foot long fuel elements³⁴. Burnups and heat ratings represented were 3×10^{18} to 1.5×10^{20} f/cc and 5 to 27 kW/ft, respectively. Results correlate well with volumetric average fuel temperature. A maximum gas release of 92% was observed for the highest rated fuel rod (27 kW/ft; $\bar{T}_v = 2200$ °C). Comparisons between pelletized and vipac fuel showed no significant difference.

Continuous measurements of gas release from four different eight-foot long vipac fuel elements are in progress³⁴. Results from these experiments, which have achieved 3×10^{20} f/cc at peak heat ratings to 17 kW/ft, show that internal pressure increased at a nearly constant rate with burnup. Essentially all of the gas release occurred during thermal cycling presumably because of gas dissolution or fuel cracking on shutdown. Sorbed gases (mostly water) were either largely retained in the fuel or reacted rapidly with the Zircaloy sheath to form non-volatiles. Results from all four experiments are consistent with postirradiation gas release measurements.

Resistance of the fuel to creep is the primary restraint which controls growth of fission gas bubbles. Creep data, as well as irradiation effects on creep which are postulated to be substantial, are thus fundamental to design of fuels. The stress dependency for creep rate of UO_2 suggests a Nabarro-Herring, diffusional process to be rate controlling at stresses $< \sim 5000$ psi. At higher stresses, the mechanism is less well defined but is thought to reflect dislocation climb, cyclic generation and degeneration of dislocation loops, or some other dislocation dependent process. Activation energies are approximately the same as for uranium self-diffusion suggesting this as the rate limiting step. Deviations in grain size and, in particular, O/M cause large changes in creep rate. Changing the O/M by 0.01, for example, may cause an increase in creep rate of two orders of magnitude.

Irradiation enhancement of creep rate may occur by a variety of mechanisms. Neutron bombardment can hypothetically

introduce sufficient metastable, mobile Frenkel pairs to substantially increase creep rates below about 1200 °C for materials in which diffusional creep is predominant. Coalescence of dislocation loops into networks which act as "short circuit", high diffusivity paths may also have an effect. The high temperatures associated with individual fission spikes are a third possible source of creep enhancement. Probably the most likely source of irradiation induced creep is variation in O/M during fissioning or by oxygen redistribution in mechanically critical, fuel temperature regimes. In-reactor experiments directed toward evaluating these processes in UO_2 - PuO_2 are in the final stages of construction³⁵. These experiments are coordinated with current laboratory measurements in terms of test conditions and specimen characteristics to permit separation of the irradiation influence from composition, grain size, and temperature effects.

Analytical Modeling of Fuel Element Performance

The preceeding discussion attempts to illustrate some of the interrelationships between kinetic processes in oxide fuels. The description that emerges is very complex - too complex in fact to permit quantitative manual manipulation of the various interacting mechanisms which determine fuel behavior at high burnups. To achieve the required performance predictions, two approaches might be employed: 1) perform experiments under all conceivable fuel service conditions and 2) devise computerized procedures to analytically characterize effects of prescribed combinations of operating parameters. Information of the first type will be obtained for present reactor concepts but only from years of operating experience with thousands of fuel elements. Near-term design and development of concepts which transcend current testing capabilities must thus rely substantially upon model development and computer simulations. Most of the effort in this area has been directed toward developing models which describe macroscopic dimensional changes in

fuel rods as functions of burnup and fluence³⁶. These models incorporate mechanical behavior of fuel and sheath, fuel swelling, temperature distribution, and amount and distribution of porosity and void space. They range in sophistication from end-of-life models which utilize only average operating conditions and total burnup through steady-state models which consider the rate dependence of deformations in terms of viscous flow processes, to pseudo-transient models which describe the fuel in more realistic, continuum mechanical terms. Current efforts on continuum formulations which consider interactions between transport phenomena and mechanical deformation will greatly enhance the versatility of pseudo-transient models³⁷. All of these types of simulations require further development and empirical verification. Good agreement between calculated and observed behavior has been achieved in some instances, however.

A common characteristic of macroscopic models is that they require as input independent descriptions of the kinetic processes which influence mass transport in the fuel. These processes (e.g., bubble growth and migration, associated effects on heat transfer and deformation fields, mechanisms of void formation and closure, etc.) must be quantitatively determined either analytically or empirically. Stochastic models which relate measurable parameters and the precision with which they must be known to the parameters required by continuum models for describing deformations are useful in these efforts³⁸. These models have the potential for greatly expanding the amount of useful information obtainable from a given experiment. Evaluation of various potential transport mechanisms by iterative comparison of their predictions with macroscopic observations is also a possibility.

Contemporary models of fuel materials behavior are still substantially removed from reality. The field is progressing rapidly, however, and probably represents the best hope for eventually interpreting the complex interactions of kinetic processes which limit fuel element performance.

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