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Fuel Performance Improvement Program

Thermal Conductivity of Sphere-Pac Fuel

July 1981

Consumers
Power
Company

EXXON NUCLEAR
COMPANY, Inc.

Battelle

Pacific Northwest Laboratory

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FUEL PERFORMANCE IMPROVEMENT PROGRAM

**Thermal Conductivity of
Sphere-Pac Fuel**

July 1981

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ABSTRACT

Progress in understanding the thermal conductivity of sphere-pac fuel beds has been made both at Oregon State University and Exxon Nuclear Company supported by the Fuel Performance Improvement Program (FPIP). FPIP is sponsored by the U. S. Department of Energy and is being performed by Consumers Power Company, Exxon Nuclear Company, and Pacific Northwest Laboratory. The purpose of the program is to test and demonstrate improved light water reactor fuel concepts that are more resistant to failure from pellet-cladding interaction during power increases than standard pellet fuel.

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1.0 SUMMARY AND INTRODUCTION

This report details a theoretical model for calculating the thermal conductivity of sphere-pac fuel consisting of a ternary mixture of fuel microspheres. Numerical results are provided for the application of this model to the sphere-pac fuel particle diameters, and packing fractions of the FPIP program. A comparison of experimental data with calculated results for thermal conductivity shows agreement to within 20% for unrestructured fuel. The calculated results provided for the FPIP sphere-pac fuel are extensive and include the effects of temperature on thermal conductivity as well as the effects of fuel restructuring, fission gas release, and fuel rod internal pressure. The calculated results show

- an increase in thermal conductivity with restructuring due to an increase in the contact area between adjacent fuel microspheres
- a reduction in thermal conductivity with fission gas release as the thermal conductivity of fill gas is degraded
- an increase in thermal conductivity as fuel rod internal gas pressure is increased owing to the Knudsen domain effect in the gas gap between fuel microspheres.

The determination of the temperature distribution in sphere-pac fuel requires knowledge of the thermal conductivity of the sphere-pac bed. The sphere-pac beds to be considered are made up of three size fractions.⁽¹⁾ The coarse fraction consists of large UO_2 spheres. This is surrounded by medium spheres (or medium fraction), small spheres (or fine fraction) and the fill gas. Initially, the fill gas is helium.

For evaluating the effective conductivity of sphere-pac fuel, one dimensional algebraic models have been developed to predict the thermal conductivity of a sphere-pac array that includes spheres of the same size.^(2,3) However, these models were found to be inaccurate for packed beds in which the thermal conductivity of the solid phase is much larger than that of the gas phase.⁽⁴⁾ Moreover, such models are limited since they often include empirical constants and fitting parameters that are sometimes difficult to determine. In this report, a general two-dimensional numerical model for evaluating the thermal conductivity of sphere-pac fuel is presented. This model is based on the work of Ades et al^(4,5) developed for two-fraction sphere-pac carbide fuel. The model was modified and extended for evaluating the thermal conductivity of three-fraction sphere-pac oxide fuel. The model employs a unit cell approach to account for the physical phenomena that are important with respect to the thermal conductivity of sphere-pac fuel. The basic model enables the effective conductivity to be determined as a function of fuel temperature and fill gas pressure. Moreover, the model can handle physical changes in the sphere-pac bed such as

1. Fission gas content in the fill gas.
2. Sintering of the fuel microspheres.

2.0 THEORETICAL MODEL

2.1 GENERAL DESCRIPTION AND APPLICATION TO FINE FRACTION UNIT CELL

The present theoretical developments for sphere-pac fuel are for the case where three distinct sphere sizes are mixed together within the fuel rod. The sizes are referred to as coarse, medium and fine. The diameter of the medium spheres is several times that of the fine spheres, and the diameter of the coarse spheres is several times that of the medium spheres. The evaluation of the effective thermal conductivity of sphere-pac fuel is based on the assumption that the conductivity of the randomly packed spheres corresponds to that of a ternary unit cell. Figure 2.1 (*iii*) shows an idealized two-dimensional ternary unit cell that consists of the coarse fraction surrounded by medium and fine fraction and fill gas. The medium fraction in the ternary unit cell is further represented by an idealized binary unit cell (Figure 2.1 (*ii*)). Similarly the fine fraction unit cell consists of two fuel hemispheres of UO_2 surrounded by fill gas, as shown in Figure 2.1 (*i*).

In evaluating the thermal conductivity of the ternary mixture the theoretical treatment of each unit cell is fundamentally the same. Each unit cell consists of two fuel hemispheres in contact. Hence the length of the unit cell over which heat transfer is occurring is equal to the sphere diameter, $2r$. The cylindrical radius of the insulated boundary of each unit cell is somewhat larger than the sphere radius as indicated by the radial length, a , in Figure 2.1 (*i*). Thus as shown in Figure 2.1 (*i*) the unit cell is a cylindrical volume having a radius of $r + a$ and an axial extent of $2r$.

The unit cell model used in the present work is intended to represent all major physical phenomena related to the thermal conductivity of sphere-pac fuel. Some effects occurring in practice such as the irregular heat transfer path between the contact points of a large number of randomly packed spheres, or the fact that some adjacent spheres may not actually be in contact, are not handled explicitly but rather are implicitly accounted for by making the radius of the unit cell somewhat larger than the sphere radius. The selection of the increase in cell radius for the present application is discussed further in Subsection 2.2. The effective thermal conductivity of the unit cell is defined in terms of the heat flux, q , through the cell by the following equation

$$k_{\text{eff}} = \frac{q}{\pi(r+a)^2} \frac{2r}{\Delta T} \quad (2.1)$$

where ΔT is the temperature difference between the axial boundaries of the unit cell at $z = 0$ and $z = 2r$ as shown in Figure 2.1.

Determination of the effective thermal conductivity of each unit cell and hence the ternary mixture thus corresponds to a three dimensional steady-state conduction problem with cylindrical symmetry. The solution sought is the heat flux across the unit cell that corresponds to a prescribed temperature difference ΔT . Note that the unit cell contains two media of different thermal conductivities, the two fuel hemispheres being one and the surrounding continuum being the other. This problem while relatively simple

does require a numerical solution such as by standard finite difference or finite element techniques. Since a numerical solution approach is used the evaluation of the effective conductivity of the ternary unit cell begins with first evaluating the conductivity of the fine fraction unit cell. This conductivity is then used to evaluate the conductivity of the binary mixture and ultimately that of the ternary unit cell. The remaining part of this subsection provides further details of the theoretical approach and its application to the fine fraction unit cell.

The boundary conditions for the fine fraction unit cell are uniform temperatures T_1 and T_2 at the upper and lower axial boundaries respectively. Hence,

$$T(r, z = 0) = T_1 \quad (2.2.a)$$

$$T(r, z = 2r_s) = T_2 \quad (2.2.b)$$

The boundary condition for the insulated surface is

$$\frac{\partial T}{\partial r}(r = r_s + a, z) = 0 \quad (2.3.a)$$

and from the cylindrical symmetry of the cell

$$\frac{\partial T}{\partial r}(r = 0, z) = 0 \quad (2.3.b)$$

Since the present interest is to determine the effective thermal conductivity of the ternary unit cell internal heat generation is not considered. The basic steady-state heat conduction equation to be solved is

$$\nabla \cdot k \nabla T(r,z) = 0 \quad (2.4)$$

In the present application the spheres are made of solid UO_2 with a porosity that is typically about 1.0% or less. For the fine fraction unit cell the media surrounding the two fuel hemispheres is a gas. This gas is either the original fill gas (helium) or a mixture of the fill gas and the released fission gases (xenon and krypton). In the case of unrestructured fuel the two clean hard hemispheres of the unit cell are assumed in physical contact with no interfacial pressure. Since there is only one point of contact essentially all the axial heat flow must pass through the gas gap region between the spheres. Conduction across the gas gap region offers the major resistance to heat transfer and hence represents a key aspect of the problem being considered. Thermal radiation across the gas gap region will provide some heat transfer across the gap region but this effect is significant only at temperatures above 2000°C and when fuel rod internal pressure is less than 1.0 atmospheres. Thermal radiation heat transfer has not been explicitly handled in the present work. It is noted that at the elevated temperatures where thermal radiation becomes a potential consideration, restructuring of the UO_2 microspheres is a much more important high temperature effect. The strong effect of fuel restructuring on thermal conductivity is treated in Section 2.4.

In the three-dimensional unit cell conduction problem being considered the temperature distribution throughout the cell is continuous. The temperature gradient however is discontinuous at the sphere boundaries because of the difference in thermal conductivity of the spheres and the surrounding medium. In the case of the fine fraction unit cell with a gaseous medium between the spheres the heat conduction through the fill gas is affected by the Knudsen domain particularly near the sphere contact point as the separation distance between the fuel spheres becomes small. The Knudsen domain is generally assumed for any gap width less than 100 mean free paths.⁽⁶⁾ For this situation, the thermal conductivity of the gas is corrected by using the expression of Kennard and reported by Olander.⁽⁷⁾

$$K_{\text{gap}} = \frac{K_g}{1+\sigma} \quad (2.5)$$

where

$$\sigma = 4 \frac{2-\alpha}{\alpha} \frac{\lambda}{\gamma+1} \frac{1}{Pr_g} \frac{\lambda_0}{P} \frac{T}{273.15} \frac{1}{t_{\text{gap}}}$$

K_g = thermal conductivity of the fill gas, W/cm°C

α = thermal accommodation coefficient of the fill gas on the sphere fuel.

Pr_g = Prandtl number of the fill gas

γ = C_p/C_v (= 5/3 for monoatomic gas)

P = gas pressure, atm.

T = gas temperature, °K

λ_0 = mean free path of the gas molecules at 1 atm. and 0°C, atm-cm.

t_{gap} = separation distance spheres, cm.

2.2 NUMERICAL CALCULATIONS

The determination of the heat flux across the unit cell and hence the effective thermal conductivity is determined by standard numerical methods. In the present applications a finite difference scheme was used. The ratios of the cylindrical unit cell radius to sphere radius for the fine, medium and coarse unit cells were respectively 1.86, 1.95 and 1.14 in the present numerical calculations. These ratios are based on the packing factors for each size fraction taken individually without consideration of the volume displaced by larger sized spheres. It should be noted that a fully acceptable criteria for the cell to sphere radius ratio has not been established. Hence the present values for the cell to sphere radius ratios should be recognized as tentative values that could be improved upon in future work.

The unit cell is first divided into regions. These regions are formed by lines parallel to the r and z axis. Each region is assigned a material with known properties. A total number of 20 radial and 26 axial nodes are used to define the mesh in the unit cell. Constant temperatures T_1 and T_2 are assigned to the top and bottom boundaries of the unit cell respectively. Typically the temperature difference ($T_1 - T_2$) is small, on the order of 20°C in order to avoid second order temperature dependencies.

Appropriate temperature-dependent properties are used. Using the successive overrelaxation method with a dominant-error-mode extrapolation technique to accelerate convergence⁽⁸⁾, the two-dimensional temperature field is determined for each node in the unit cell.

2.3 APPLICATION TO THE BINARY AND TERNARY UNIT CELLS

The effective conductivity of the binary unit cell consisting of the medium fraction surrounded by the fine fraction and the fill gas is solved by substituting the computed value of the effective conductivity of the fine fraction unit cell in the filling portion between the two medium hemispheres (Figure 2.1 (ii)). The analytical technique used to solve for the conductivity in the binary unit cell is identical to that used for the fine fraction unit cell.

The evaluation of the effective conductivity of the binary involves, however, a basic difference with that used for the fine fraction. This difference is due to the distance from the contact point, where fine fraction spheres cannot reside. Instead, the fill gas is included in this region and therefore, yields to a decrease of the effective conductivity of the binary. The distance between the fuel medium spheres, L_1 , where fine spheres cannot reside is shown in Figure 2.2 and can be expressed as

$$L_1 = 2 (r_m - \sqrt{r_m^2 - x_1^2}) \quad (2.6)$$

where

$$x_1 = [(r_m + r_s)^2 - r_m^2]^{1/2} - r_s$$

X_1 = minimum distance from the edge of the unit cell where fine spheres can reside

r_m = radius of the medium sphere

r_s = radius of the fine sphere.

Furthermore the effective conductivity of the binary mixture can be evaluated by using the model described in Section 2.1 with

- (1) The conductivity of the fill gas when the distance between the boundaries of the coarse fraction is smaller than L_1 .
- (2) The effective conductivity of the fine fraction unit cell when the distance separating the fuel sphere is larger than L_1 .

The effective thermal conductivity of the ternary unit cell consisting of the coarse fraction surrounded by the binary mixture and the fill gas is solved by substituting the computed value of the respective effective conductivities in the filling portion between the two coarse hemispheres (Figure 2.1 (iii)). The technique used to solve for the conductivity ternary unit cell is identical to that used for the fine fraction unit cell. The distances L_2 and L_3 where medium and fine fuel fractions respectively cannot reside between the two coarse fuel hemispheres are accounted for in a manner similar to that for the binary unit cell. These distances are expressed as

$$L_2 = 2 (r_L - \sqrt{r_L^2 - x_2^2}) \quad (2.7)$$

and

$$L_3 = 2 (r_L - \sqrt{r_L^2 - x_3^2}) \quad (2.8)$$

with

$$x_2 = [(r_L + r_m)^2 - r_L^2]^{\frac{1}{2}} - r_m$$

$$x_3 = [(r_L + r_s)^2 - r_L^2]^{\frac{1}{2}} - r_s$$

where

r_L = radius of the coarse sphere.

By denoting by L the distance separating the two coarse hemispheres, the assumption consists therefore of

- (1) Using the fill gas conductivity in the region between the spheres where $L < L_3$.
- (2) Using the effective conductivity of the fine fraction unit cell for $L_3 < L < L_2$.
- (3) Using the effective conductivity of the binary unit cell for $L > L_2$.

2.4 EFFECT OF EXPOSURE

2.4.1 Fuel Restructuring

Depending on pin power and cooling conditions, sphere-pac fuel can undergo substantial restructuring. "Initial stage" restructuring is

said to occur when the sphere-pac form is progressively lost due to thermal processes such as sintering. In sintering the fuel microspheres fuse each other. When initial stage restructuring is completed in a given fuel region, the fuel has the appearance of a porous pellet.

As part of the prediction of the thermal behavior of sphere-pac fuel undergoing sintering, it is necessary to determine the temperature distribution within the fuel pin. For such calculations, the thermal conductivity of the sphere-pac fuel must be known as sintering proceeds. Sintering of sphere-pac fuel is a thermal process in which solid particles in contact adhere and coalesce together into a solid compact. As a result the surface area of the particles decreases thus giving rise to densification of the compact. Sintering also can result in pore shape changes between the particles, pore shrinkage, and grain growth. Sintering can be enhanced by an applied compressive stress. This is usually referred to as hot-pressing. Stress-free sintering is due to non-equilibrium mechanical conditions that develop between the two particles in contact. The sintering process develops as a result of the reduction in free energy of the particle surface when there is a decrease in stress due to curvature and equalization of vapor pressure. This may be followed by the elimination of vacancy concentration in the immediate vicinity of the contact area between the particles.^(9,10)

The important physical changes which occur during the sintering process between spherical particles are characterized by stages.

A stage of sintering is referred to an interval of geometric change in which the interparticle pore shape change is totally defined (initial stage), or an interval of time during which the pore remains constant in shape, but decreases in size (final stage).⁽¹¹⁾

The initial stage of sintering is that during which the inter-particle contact area starts increasing. This initial stage is frequently referred to as the neck growth stage, where a "neck" is formed between the two particles (see Figure B-1). The region between the two particles, called the neck region, has a contact area of πx^2 , where x is the neck radius. The neck ratio defined as x/r has a particular significance in estimating the sintering stage. According to Coble⁽¹²⁾, the initial stage is achieved once the neck ratio is equal to about 0.15.

The intermediate stage begins with grain growth and pore shape changes. Densification of the compact continues. The shape of the spheres changes significantly as the porosity develops and forms interlinked channels.

The final stage of sintering begins when the pore phase in the aggregate of particles is pinched off. Individual pores of finite shape and size are created in the fuel while the interlinked porosity decreases gradually. At this stage, the initial sphere-pac fuel configuration has been entirely lost and replaced by a pellet-shaped densifying material. The transition between the intermediate and final stages is generally difficult to determine.

2.4.2 Effective Conductivity of Sphere-Pac Fuel Undergoing Sintering

Five basic sintering mechanisms have been identified. These are described in Appendix B. However, for UO_2 fuel, the dominant sintering mechanism is still unknown. Several investigators, though, have shown that under typical thermal operating conditions, diffusional mechanisms such as surface, grain boundary and volume diffusion are significant.

In the absence of the dominant sintering mechanism, a parametric study will be made to evaluate the effect of restructuring characterized by the increase in neck ratio between the particles on the effective conductivity. Upper and lower bounds of the effective conductivity will be obtained by assuming

- (1) that only the fine fraction sinters,
- (2) that the extents of sintering of the medium and coarse fractions is equal to the sintering occurring between the fine fraction.

In both of the above cases, shrinkage of the particles due to sintering is neglected. Finally, as an example, it will be shown that an assumed diffusional sintering mechanism, surface diffusion, yields an effective conductivity included in the upper and lower bounds previously determined.

It should be noted, however, that though the surface diffusion mechanism may not be dominant within the whole temperature range of interest, this sintering mode has been found to be significant for UO_2

fuel.⁽⁶⁾ The method used to evaluate the effective conductivity of sphere-pac fuel undergoing sintering is based on the model of Ades et al⁽¹³⁾ and uses the unit cell model described in Section 2.1. Figure 2.3 shows schematically a ternary unit cell with both the fine, medium and coarse fractions undergoing sintering. As a result of the neck growth of the fine fraction, the effective conductivity of the fine fraction unit cell increases. The extent of sintering undergone by the medium and coarse spheres that include neck growth is, however, much less significant since the medium and coarse sphere sizes are much greater than the size of the fine fraction.

As a first step, the contact area between the fine fraction spheres is increased and is equal to πx^2 , where x is the neck radius (Figure 2.4). Shrinkage of the unit cell is neglected. The temperature distribution in the unit cell is then evaluated by the method outlined in Section 2.1 and the effective conductivity is computed accordingly. Furthermore, the value of the effective conductivity of the fine fraction undergoing sintering is then substituted into the binary unit cell where a neck ratio is assumed between the medium spheres. The effective conductivity of the binary unit cell is then evaluated and substituted into the region of the binary mixture in the ternary unit cell. This then allows the evaluation of the effective conductivity of the ternary mixture for which a neck ratio value is assumed between the coarse spheres undergoing restructuring. For the present calculations, shrinkage of the unit cells are neglected, so that the volumes of the unit cells remains unchanged.

Using the above method, the effective conductivity of the ternary mixture can then be calculated for each temperature as a function of the neck ratios assumed between the different fuel fractions.

2.4.3 Fission Gas Release

In the model described in Section 2.1, it was assumed that the fill gas in the fuel rod consists solely of helium, the gas with which the rod is initially filled. This assumption may be correct for using the model to evaluate the effective conductivity of the ternary unit cell at the initial stage of irradiation, i.e., when only helium constitutes the fill gas. As fuel irradiation proceeds, the fill gas consists of a mixture of the initial helium gas and the gaseous fission products such as xenon and krypton. Only two basic modifications are made to the original model to account for the gas mixture. These modifications are:

- (1) The thermal conductivity of the gas mixture outside the Knudsen domain.
- (2) The thermal conductance of the gas mixture in the gap region within the Knudsen domain.

The thermal conductivity of the gas mixture outside the Knudsen domain is modeled by using the Brokaw model⁽¹⁴⁾ and is expressed as

$$K_{\text{mix}} = \sum_{i=1}^n \frac{K_i}{1 + \sum_{\substack{j=1 \\ j \neq i}}^n \psi_{ij} \frac{X_j}{X_i}} \quad (2.9)$$

where

$$\psi_{ij} = \phi_{ij} \left[1 + 2.41 \frac{(M_i - M_j)(M_i - 0.142 M_j)}{(M_i + M_j)^2} \right] \quad (2.10a)$$

and

$$\phi_{ij} = \frac{[1 + (k_i/K_j)^{1/2} (M_i/M_j)^{1/4}]^2}{2^{3/2} [1 + (M_i/M_j)]^{1/2}} \quad (2.10b)$$

where

n = number of components in the gas mixture

M_i = molecular weight of the i th component

X_i = mole fraction of i th component

K_i = thermal conductivity of the pure gas for the i th component.

For the present calculations

$$n = 3$$

Therefore, by using equation (2.5) for helium, xenon and Krypton separately, the thermal conductivity of each mixture component in the gap region is obtained. After substituting these conductivity values into equation (2.9), the thermal conductivity of the gas mixture in the gap region can be evaluated. The gas mixture conductance in the gas gap region affected by the Knudsen domain is thus expressed as

$$h_{\text{gap}} = \frac{K_{\text{mix}}}{t_{\text{gap}}} \quad (2.11)$$

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- (i) FINE FRACTION SURROUNDED BY FILL GAS
- (ii) MEDIUM FRACTION SURROUNDED BY FINE FRACTION AND GAS
- (iii) COARSE FRACTION SURROUNDED BY FINE AND COARSE FRACTIONS AND FILL GAS

FIGURE 2.1. Schematic Representation of a Sphere-Pac Unit Cell

FIGURE 2.2. Scheme for Determining the Distance L_1 Between the Surfaces of Medium Fuel Spheres Below Which Fine Spheres Cannot Reside

FIGURE 2.3. Ternary Unit Cell With Coarse, Medium and Fine Fractions Undergoing Sintering

FIGURE 2.4. Fine Fraction Unit Cell with Fine Spheres Undergoing Restructuring

3.0 COMPARISON OF THEORETICAL MODEL WITH DATA

In order to assess the accuracy of the model developed for predicting the thermal conductivity of sphere-pac fuel, the model was used to evaluate the thermal conductivity of ORNL sphere-pac fuel. The characteristics of this fuel are shown in Table 3.1. Predicted values of the thermal conductivity were furthermore compared with available experimental data provided by R. L. Beatty of Oak Ridge National Laboratory.

Figures 3.1, 3.2 and 3.3 show the computed effective conductivity of ORNL sphere-pac fuel in the temperature range of $100 \leq T \leq 800^{\circ}\text{C}$ for various fill gas pressures of 1., 3. and 6. atmospheres. Experimental data of the measured conductivity are also shown accordingly.

Comparison between predicted and measured conductivity values indicate that good agreement is obtained when the fill gas pressure is at 1 atmosphere. For higher pressure values at 3 and 6 atmospheres, however, the measured conductivity values are higher than those predicted.

Though both predictions and data show the same trend of the temperature dependence, the predicted values do not exhibit the strong fill gas pressure dependence of the measured conductivity. Nevertheless, the discrepancy between measured and predicted values is smaller than 20%. This result suggests that the model developed is adequate, though further improvements are needed to account for the stronger pressure dependence of the thermal conductivity that is obtained from the experimental measurements. Some recommendations for further theoretical work are given in Section 5.0.

TABLE 3.1. Physical Characteristics of ORNL Sphere-Pac Fuel
Including a 3-Fraction Mixture at 86.6% Smear
Density^(a)

Parameter	Size Fractions		
	Coarse	Medium	Fine
Size Range (μm)	1190-1410	280-320	25-45
Ave. Sphere Density (gm/gc)	10.82	10.89	10.81
% T. D.	98.6	99.3	98.6
Packing Factor (%)	50.94	17.44	19.32

(a) From R. L. Beatty of Oak Ridge National Laboratory

FIGURE 3.1. Effective Conductivity of Ternary Mixture Versus Temperature for ORNL Fuel

FIGURE 3.2. Effective Conductivity of Ternary Mixture Versus Temperature for ORNL Fuel

FIGURE 3.3. Effective Conductivity of Ternary Mixture Versus Temperature for ORNL Fuel

4.0 APPLICATION OF MODEL TO SPHERE-PAC FUEL

4.1 BASE CALCULATIONS

Computational results of the effective conductivity of sphere-pac fuel in its initial configuration resulting from the fine, binary and ternary unit cell models described in Section 2.1 are shown in Figures 4.1, 4.2, and 4.3 respectively. The curves obtained show the variation of the effective conductivity with temperatures ($500 \leq T \leq 2500^{\circ}\text{C}$) as a function of fill gas pressure ($1 \leq p \leq 20$ at.). In all cases, the porosity of the fine, medium and coarse fractions was assumed to be 1%. The packing factors of the fine, medium, and coarse fractions are 51.6%, 17.58%, and 19.33%, respectively.

The effective conductivity of the fine fraction, binary and ternary mixtures exhibit initially a strong dependence on the fill gas pressure at all temperatures. This dependence decreases with increasing gas pressure. As shown in Figure 4.3, the conductivity of the sphere-pac bed is much less pressure dependent and levels off as the fill gas pressure reaches about 8. atmospheres. This result can be explained by the fact that the pressure dependence resides in the gas gap separating the fuel spheres. As the pressure increases, the gap conductivity increases as well and reaches an asymptotic value (Eq. 2.5). Beyond this value, there is almost no change of the gap conductivity with gas pressure.

Figure 4.4 shows the effective conductivity of the ternary mixture vs. temperature for various fill gas pressures. As can be seen, the

effective conductivity decreases with increasing temperature for $T \leq 2000^{\circ}\text{C}$ and increases with increasing gas pressure. For higher temperatures ($T > 2000^{\circ}\text{C}$) the effective conductivity increases with temperature and in a lesser extent with fill gas pressure. As observed earlier, it can be seen that for a gas pressure higher than about 8. atmosphere, there is no significant changes of the thermal conductivity with gas pressure (less than 5%).

4.2 EFFECT OF FISSION GAS RELEASE

Figure 4.5 shows the conductivity of the sphere-pac ternary mixture versus fill gas pressure and composition at 1000°C . The fill gas composition was varied from 100% helium to 20% helium, the remainder being fission gas xenon and krypton. As can be seen, by increasing the fission gas content in the fill gas results in a decrease of the effective conductivity for a given gas pressure. This decrease is higher for a variation of fission gas content from 0 to 40% than from 40 to 80%. Also, increasing the fission gas content in the gas mixture decreases the dependence of the effective conductivity on gas pressure. The breakaway pressure beyond which the effective conductivity levels off is reduced as the fission gas content is increased.

Figure 4.6 shows the variation of the conductivity of the sphere-pac ternary mixture with temperature and fill gas composition. The fill gas is at 5 atmospheres. As can be seen, increase of the fission gas content in the fill gas yields a decrease in the effective conductivity in the temperature range considered ($500 \leq T \leq 2055^{\circ}\text{C}$).

4.3 EFFECT OF RESTRUCTURING

As an example of the behavior of sintering of the fuel particles, Figures 4.7 and 4.8 show the neck growth between fuel spheres as a function of time and temperature ($T = 1500$ and 2500°C) for various particles size (40, 300, and $1280\text{ }\mu\text{m}$). The neck growth is evaluated by assuming surface diffusion as being the sintering mechanism of the fuel particles. As can be seen, the neck ratio (x/r) increases rapidly within a short period of time, and then levels off. It is important to note that

- (1) the neck ratio increases with increasing temperature,
- (2) the neck ratio increases with decreasing particle size.

The last result outlines that in the sintering process of the sphere-pac ternary mixture, sintering that occurs between the fine fraction will be predominant and thus, will enhance the thermal conductivity of the fuel undergoing restructuring. It should be outlined that this is expected to be valid for all existing sintering mechanism assumed (diffusional and non-diffusional mechanisms, see Table B.1).

Figures 4.9 - 4.11 show the variations of the effective conductivity of the sphere-pac ternary mixture with temperature and neck ratio of the fine fraction (surface diffusion is assumed as the sintering mechanism for the medium and coarse fractions). The results are shown for a fill gas of 100% helium at 1., 5., and 20 atmospheres, respectively. As can be seen, as the neck ratio of the fine fraction (x/r) is increased from 0. to 0.8, the effective conductivity of the ternary mixture increases. It should be noted,

however, that this increase is more important at low fill gas pressure (such as 1 atmosphere, see Figure 4.9) than at higher pressures (see Figure 4.10 and 4.11). It can therefore be concluded that sintering will be more effective in increasing the effective conductivity of sphere-pac fuel more at low than at high fill gas pressure.

Figures 4.12 and 4.13 show the variation of the effective conductivity of the ternary mixture with temperature and fission gas content in the fill gas for neck ratios between the fine spheres equal to 0.2 and 0.4, respectively. Surface diffusion is assumed for the other fuel fractions. The fill gas is at 1 atmosphere. As can be seen increasing the fission gas content in the fill gas results in a decrease of the effective conductivity of the ternary mixture. However, this decrease is less important at high values of the neck ratio ($x/r = 0.4$) than at lower values ($x/r = 0.2$). This result therefore suggests that restructuring may offset the effect of fission gas content in the fill gas by reducing the decrease of the effective conductivity due to the presence of fission gas in the fill gas.

Figure 4.14 shows the variation of the effective conductivity of the sphere-pac ternary mixture with the neck ratio (x/r) of the fine fraction. The temperature and fill gas pressure are 1500°C and 5 atmospheres, respectively. The fraction of fission gas in the fill gas is 20%. The lower conductivity curve was obtained by assuming sintering of the fine fraction only, the remainder being unsintered fuel spheres. The upper curve was obtained by assuming that the extent of restructuring occurring between all fuel

fractions is equal, i.e., by setting the neck ratios of the fine, medium and coarse fractions equal. As can be seen, the effective conductivity increases significantly with increasing the neck ratio ($x/4$). This increase is even more important as the extent of restructuring of all fuel fractions are set equal (upper curve).

Also shown is the effective conductivity dependence on the neck ratio by assuming the surface diffusion mechanism as the dominant sintering process. In this case, the curve of the calculated conductivity is bounded by the upper and lower curves.

More generally, due to the strong effect of restructuring on the effective conductivity, the results indicate a need to determine the sintering mechanism of the UO_2 sphere-pac fuel. As shown in Figure 4.14 upper and lower bounds of this dependence can be established, but further theoretical efforts are needed to assess with more accuracy the variation of the effective conductivity with the extent of restructuring occurring in the sphere-pac bed.

4.4 ALGEBRAIC CURVE FITS OF THE CALCULATED NUMERICAL RESULTS

Numerical correlations have been established to calculate the effective conductivity of sphere-pac fuel for FPIP, in its initial configuration (without restructuring) with key parameters that are:

- (1) The fuel temperature.

(2) The pressure of the fill gas in the fuel rod.

(3) The composition of the fill gas that includes initially helium and ultimately consists of a mixture of helium and fission gas (xenon and krypton).

The basic correlation relating the thermal conductivity to temperature with a fill gas of 100% helium at 1. atmosphere is expressed as:

$$K_T = 2.0934 \cdot 10^{-2} - 9.131429 \cdot 10^{-6}T + 1.357143 \cdot 10^{-9}T^2 + 2.8 \cdot 10^{-13}T^3 \quad (4.1)$$

where

K_T = Thermal Conductivity, W/cm °C

T = Temperature, °C

The correlation of the thermal conductivity with temperature and fill gas pressure is expressed as:

(a) For $1 < P \leq 5$ at

$$K(T,P) = K_T \left[1 + f_1(T) \frac{P-1}{4} \right] \quad (4.2.a)$$

(b) For $5 < P \leq 10$ at

$$K(T,P) = K_T \left[1 + f_1(T) + f_2(T) \frac{P-5}{5} \right] \quad (4.2.b)$$

(c) For $10 < P \leq 20$ at

$$K(T,P) = K_T \left[1 + f_1(T) + f_2(T) + f_3(T) \frac{P-10}{10} \right] \quad (4.2.c)$$

with:

$$\begin{aligned} f_1(T) &= 2.9802 \cdot 10^{-1} - 3.087143 \cdot 10^{-4} T + 3.059714 \cdot 10^{-7} T^2 - 6.2 \cdot 10^{-11} T^3 \\ f_2(T) &= 6.7 \cdot 10^{-2} - 8.305238 \cdot 10^{-5} T + 6.982857 \cdot 10^{-8} T^2 - 1.373333 \cdot 10^{-11} T^3 \\ f_3(T) &= 4.168 \cdot 10^{-2} - 5.101905 \cdot 10^{-5} T + 4.122857 \cdot 10^{-8} T^2 - 7.866667 \cdot 10^{-12} T^3 \end{aligned}$$

where:

T = Temperature, °C

P = fill gas pressure, at.

The correlation of the thermal conductivity with temperature, fill gas pressure and composition is expressed as:

(a) For $0. < f \leq 0.20$

$$K(T, P, f) = K(T, P) \left[1. - 0.117 \frac{f}{0.2} \right] \quad (4.3.a)$$

(b) For $0.20 < f \leq 0.40$

$$K(T, P, f) = K(T, P) \left[0.8830 - 0.0991 \frac{f-0.2}{0.2} \right] \quad (4.3.b)$$

(c) For $0.40 < f \leq 0.60$

$$K(T, P) = K(T, P) \left[0.7839 - 0.0998 \frac{f-0.4}{0.2} \right] \quad (4.3.c)$$

(d) For $0.60 < f \leq 0.80$

$$K(T, P, f) = K(T, P) \left[0.6841 - 0.0569 \frac{f-0.6}{0.2} \right] \quad (4.3.d)$$

where:

f = fractional value of fission gas in fill gas mixture.

FIGURE 4.1. Effective Conductivity of Fine Fraction Fuel Versus Fill Gas Pressure

FIGURE 4.2. Effective Conductivity of Binary Mixture (Fine and Medium Fractions) Versus Fill Gas Pressure for Various Temperatures

FIGURE 4.3. Effective Conductivity of Ternary Mixture (Fine, Medium and Coarse Fractions) Versus Fill Gas Pressure for Various Temperatures

FIGURE 4.4. Effective Conductivity of Ternary Mixture (Fine, Medium and Coarse Fractions) vs. Temperature for Various Fill Gas Pressures

FIGURE 4.5. Effective Conductivity of Ternary Mixture Versus Fill Gas Pressure and Composition at $1000^{\circ}C$

FIGURE 4.6. Effective Conductivity of Ternary Mixture (Fine, Medium and Coarse Fractions) vs. Temperature for Various Contents of Fission Gas in Fill Gas at 5. Atmospheres

FIGURE 4.7. Sphere-Pac Fuel Sintering Due to Uranium Surface Diffusion at 1500°C

FIGURE 4.8. Sphere-Pac Fuel Sintering Due to Uranium Surface Diffusion at 2500°C

FIGURE 4.9. Effective Conductivity of Ternary Mixture Versus Temperature for Various Neck Radii (x/r)

FIGURE 4.10. Effective Conductivity of Ternary Mixture Versus Temperature for Various Neck Ratios (x/r)

FIGURE 4.11. Effective Conductivity of Ternary Mixture Versus Temperature for Various Neck Ratios (x/r)

FIGURE 4.12. Effective Conductivity of Ternary Mixture Versus Temperature and Fission Gas Content in the Fill Gas

FIGURE 4.13. Effective Conductivity of Ternary Mixture Versus Temperature and Fission Gas Content in the Fill Gas

FIGURE 4.14. Effective Conductivity of Ternary Mixture Versus Neck Ratio of the Fine Fraction

5.0 CONCLUSION AND RECOMMENDATIONS

In this report, a two-dimensional finite difference model was developed to evaluate the thermal conductivity of FPIP ternary mixture sphere-pac fuel. This model employs a unit cell approach. Fine, medium and coarse fractions can be treated separately. Furthermore, the effective conductivity can be determined as a function of fuel temperature and fill gas pressure. Moreover, the model was extended to incorporate physical changes in the sphere-pac unit cell such as

- (1) Fission gas content in the fill gas,
- (2) Sintering of the fuel microspheres.

Computational results have shown that the temperature dependent thermal conductivity

- (1) Increases by increasing fill gas pressure,
- (2) Decreases by increasing the fission gas content in the fill gas,
- (3) Increases with the neck growth induced by the sintering of the fuel particles.

Comparison of the predicted thermal conductivity with experimental data has shown that the model developed is adequate, though the predicted values do not exhibit the strong pressure dependence that is observed. Nevertheless, it was found that the discrepancy between computed and measured conductivity values do not exceed 20%.

5.1 RECOMMENDATIONS FOR FURTHER THEORETICAL STUDIES

The model presented in this report should be improved so that the observed pressure dependence of the effective conductivity can be reproduced more accurately. In particular, the present model does not account for the different open gas gaps that are formed as a result of the contact between fuel microspheres of different sizes. The gas conductivity in these gaps is pressure dependent (Equation 2.5) and should be implemented in the model. Similarly, the gas gap model as reported by Olander⁽⁷⁾ is presently used in the model to account for the conductivity drop in rarefied gas regions (open gaps). Other gas gap models should be investigated and implemented in the new model with a view to improve the predictions of the pressure-dependent conductivity of the sphere-pac bed.

As outlined in Section 2.4.2 the mechanism with which the UO_2 particles restructure is still unknown. The sintering mechanisms reviewed in this report all predict that the extent of restructuring increases as the particle size decreases, and increases with temperature and time. Other studies have shown that restructuring is enhanced by an applied stress on the sphere particles.⁽¹⁵⁾ For design considerations it is thus necessary to determine under what conditions, sphere-pac fuel restructures. A detailed sintering analysis of UO_2 sphere-pac fuel is therefore needed to quantitatively describe the neck growth between fuel particles as a function of key parameters, such as temperature and time. The method of the sintering diagrams reported by Ashby⁽¹⁶⁾ could be used for this purpose and thus allow to determine the dominant sintering mechanism for different fuel temperature and neck ratio values.

5.2 RECOMMENDATIONS FOR FURTHER EXPERIMENTAL STUDIES

The main source of data used to assess the validity of the analytical model presented in this report is the data base provided by R. L. Beatty of Oak Ridge National Laboratory. These data, however, cover a narrow interval of temperatures and fill gas pressures (Section 3.0). Also these data are only for the ternary fuel mixture, with no information on the behavior of the thermal conductivity of the fine fraction and the binary mixture. Subsequently, further experimental work is needed to

- (1) Provide experimental data of the conductivity of the fine fraction and binary mixture for various temperatures and fill gas pressure.
- (2) Provide experimental data of the conductivity of the ternary mixture for a wide range of temperature, fill gas pressure and composition.

The above experimental tasks are at present partially undertaken at Oak Ridge National Laboratory. The results of these experiments will be used to test more accurately the validity and the accuracy of the thermal conductivity model developed.

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APPENDIX A

PHYSICAL PROPERTIES USED IN THE NUMERICAL CALCULATIONS

APPENDIX A

PHYSICAL PROPERTIES USED IN THE NUMERICAL CALCULATIONSA.1 THERMAL CONDUCTIVITY

The thermal conductivity of UO_2 is reported as ⁽¹⁾

$$K_{95} = \frac{38.24}{402.4 + T} + 6.126 \cdot 10^{-13} (T+273)^3 \quad (A-1)$$

where

K_{95} = thermal conductivity for 95% TD fuel, W/cm °C

T = temperature, °C

For fuel with densities other than 95% TD, the thermal conductivity at the desired fuel density is calculated with the following expression⁽²⁾

$$K_p = K_{95} [1-2.5P] / (0.875) \quad (A-2)$$

where

P = fractional porosity.

The thermal conductivity of helium is ⁽³⁾

$$K_{He} = 3.366 \cdot 10^{-5} (T+273.)^{0.668} \quad (A-3)$$

where

T = temperature, °C

K = thermal conductivity, w/cm °C

The thermal conductivity of xenon and krypton are expressed as (4)

$$\begin{aligned} K_{\text{xe}} &= 0.72 \cdot 10^{-6} (T+273)^{0.79} \\ K_{\text{kr}} &= 1.15 \cdot 10^{-6} (T+273)^{0.79} \end{aligned}$$

A.2 MEAN FREE PATH

The mean free path of helium molecules at 1 atm. and 0°C is (5)

$$\lambda_0 (\text{He}) = 1.74 \cdot 10^{-5} \text{ atm-cm.}$$

Similarly, the mean free path of xenon and krypton molecules are (5)

$$\begin{aligned} \lambda_0 (\text{Xe}) &= 3.6 \cdot 10^{-6} \text{ atm.-cm.} \\ \lambda_0 (\text{Kr}) &= 8.75 \cdot 10^{-6} \text{ atm-cm.} \end{aligned}$$

A.3 THERMAL ACCOMMODATION COEFFICIENT

The thermal accommodation coefficient of helium on UO_2 is obtained by using the correlation of Ades⁽⁶⁾ developed from the data of Ullman et.al.⁽⁷⁾.

This correlation is expressed as

$$\alpha (\text{He}) = 26.5 \cdot 10^{-12} T^3 - 0.0834 \cdot 10^{-6} T^2 - 28 \cdot 10^{-6} T + 0.31$$

where T = temperature, °C.

Similarly, the thermal accommodation coefficients of xenon and krypton are expressed as

$$\alpha (\text{Kr}) \approx \alpha (\text{Xe}) = 27.7 \cdot 10^{-12} T^3 - 0.0863 \cdot 10^{-6} T^2 - 136.4 \cdot 10^{-6} T + 0.7994$$

A.4 MOLECULAR WEIGHTS AND MOLE FRACTIONS

The molecular weights of helium, xenon and krypton are respectively

$$M(\text{He}) = 4.0026$$

$$M(\text{Xe}) = 131.3$$

$$M(\text{Kr}) = 83.8$$

The mole fraction of xenon and krypton in the gas mixture are expressed as

$$X(\text{Xe}) = 0.86774F$$

$$X(\text{Kr}) = 0.13226F$$

where

F = fractional value of fission gas in the gas mixture.

APPENDIX B

SPHERE-PAC FUEL SINTERING MECHANISMS

APPENDIX B

SPHERE-PAC FUEL SINTERING MECHANISMS

Five basic sintering mechanisms have been previously identified and discussed by Kuczynski⁽⁸⁾, Kingery and Berg⁽⁹⁾, Coble⁽¹⁰⁾, Kingery and Berg⁽⁹⁾, and Kingery, et.al.⁽¹¹⁾ (see Table B-1). These mechanisms are shown in Figure B.1 where the direction of material movement from the source to the sink is indicated for each mechanism. The different mechanisms shown are summarized in Table B.1 where the neck growth and shrinkage rate between the particles are formulated. As shown in Figure B.1, material transfer occurs by atomic migration from a source such as the convex particle surface to the concave surface in the neck region. As summarized in Table B.1, sintering may occur through

(1) Solid-state diffusion that includes

- (a) Surface diffusion. The mechanism is based on the vacancy concentration gradient existing between the concave surface at the neck and the convex surface of the fuel spheres. The neck region includes an excess of vacancies which migrate along the particle surface towards the convex surface. By equating the change in material volume that accumulate in the neck to the corresponding form of Fick's law for vacancy migration to the convex surface, the equation describing the neck growth is obtained and is expressed as ⁽⁸⁾

$$\frac{x}{r} = \left[\frac{28\gamma a^4 D_s t}{KTr^4} \right]^{1/7}$$

where x = neck radius, cm

r = sphere radius, cm

γ = surface energy, ergs/cm²

a = atomic diameter, cm

t = time, sec

K = Boltzmann's constant, ergs/°K

T = absolute temperature, °K

D_s is the surface diffusion coefficient in cm²sec. For UO_2 , D_s is expressed as (3)

$$D_s = 4.10^5 \exp \left[- \frac{1.08 \cdot 10^5}{RT} \right]$$

where R = gas constant, cal/mole°K

By this mechanism, transport of material occurs along the particle surfaces and shrinkage of the particles does not take place.

- (b) Volume diffusion. The mechanism for volume diffusion is based on the assumption that the interparticle grain boundaries act as a vacancy sink while the concave neck surface is the vacancy source. Vacancy diffusion occurs through the lattice rather than along grain boundaries between the spheres. The neck growth equation is derived by equating the rate of volume change in the neck region to the vacancy flux from the neck region. The neck growth is expressed as (9)

$$\frac{x}{r} = \left[\frac{80\Omega D_v \gamma t}{KTr^3} \right]^{1/5}$$

where Ω = atomic volume, cm^3 .

For UO_2 , D_v is expressed as (3)

$$D_v = 4.10^{-7} \exp \left[-\frac{69,368}{RT} \right], \frac{\text{cm}^2}{\text{sec}}.$$

By this mechanism, the resulting decrease of the distance between the fuel spheres due to shrinkage is as shown in Table B.1.

- (c) Grain boundary diffusion. The mechanism for grain boundary diffusion is based on the assumption that the grain boundaries at the particles interface act as a vacancy sink while the concave neck surface is the vacancy source. Vacancy diffusion occurs along the grain boundaries between the spheres. In this case, the neck growth is expressed as (10)

$$\frac{x}{r} = \left[\frac{96\delta D_b \Omega \gamma t}{KTr^4} \right]^{1/6}$$

where: δ = grain boundary thickness, cm.

D_b = grain boundary diffusion coefficient, cm^2/sec .

The change in the distance between the fuel spheres due to shrinkage is shown in Table B.1.

- (2) Vapor-solid phase diffusion that includes the mechanism of evaporation-condensation. This mechanism uses the basic assumption that the neck between the two particles grows as a result of mass transfer from the particle surface through the vapor phase and subsequent condensation in the neck region. The equation describing the neck growth by this mechanism is derived by using the Kelvin equation to account for the change in vapor pressure of the evaporating species on curved surfaces. The model also uses the Langmuir equation to describe the mass transfer between evaporating and condensing surfaces. By further using a mass balance relation between the evaporating and condensing material, the neck growth is obtained and is expressed as (9)

$$\frac{x}{r} = \left[\frac{3 \pi M^{3/2} P_0 \gamma t}{2 R^{3/2} T^{3/2} \rho^2 r^2} \right]^{1/3}$$

where M = atomic weight of the migrating species, g

ρ = density of migrating species, g/cm³

P_0 is the vapor pressure of the migrating species over a flat surface.

As for sintering the surface diffusion, the sintering process by evaporation-condensation does not yield to shrinkage of the sintering spheres.

- (3) Viscous flow. The mechanism of viscous flow refers to the densification process with the aid of a liquid phase. Due to

the negative pressure on the neck curvature compared with the surface of the particle, a viscous flow of material into the neck region develops. By an analysis similar to that derived for the diffusion processes, the rate of the neck growth can be expressed as (11,12)

$$\frac{x}{r} = \left[\frac{3\gamma t}{2\eta r} \right]^{1/2}$$

where η is the viscosity of the particle material. The expressions for shrinkage of the fuel particles during sintering is shown in Table B.1. More generally, shrinkage as a means of fuel densification during sintering is significant for volume and grain boundary diffusion as well as for viscous flow. However, both evaporation-condensation and surface diffusion result in only mass transfer which reduces the free energy of the particles but does not induce densification.

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Table B.1

Sintering Mechanisms, Neck Growth and Shrinkage Relations

No.	Sinter Mechanism	Neck Growth Equation	Material Transport Phase	Shrinkage Between Particle Centers	Ref.
1.	Surface Diffusion	$\frac{x}{r} = \left(\frac{28\gamma a^4 D_s t}{KTr^4} \right)^{1/7}$	solid	no shrinkage	8
2.	Volume Diffusion	$\frac{x}{r} = \left(\frac{80\Omega D_v \gamma t}{KTr^3} \right)^{1/5}$	solid	$\frac{\Delta L}{L_0} = \left(\frac{20\gamma\Omega D_v t}{\sqrt{2}r^3 KT} \right)^{2/5}$	9
3.	Grain Boundary Diffusion	$\frac{x}{r} = \left(\frac{96\delta D_b \Omega \gamma t}{KTr^4} \right)^{1/6}$	solid	$\frac{\Delta L}{L_0} = \left(\frac{3\delta D_b \gamma \Omega t}{r^4 KT} \right)^{1/3}$	10
4.	Evaporation-Condensation	$\frac{x}{r} = \left(\frac{3\sqrt{\pi} M^{3/2} P_o \gamma t}{\sqrt{2} R^{3/2} T^{3/2} P_p^2 r^2} \right)^{1/3}$	vapor-solid	no shrinkage	9
5.	Viscous Flow	$\frac{x}{r} = \left(\frac{3\gamma t}{2\eta r} \right)^{1/2}$	liquid	$\frac{\Delta L}{L_0} = \frac{3\gamma t}{4\eta r}$	11

KEY

- 1. SURFACE DIFFUSION
- 2. VOLUME DIFFUSION
- 3. GRAIN BOUNDARY DIFFUSION
- 4. EVAPORATION CONDENSATION
- 5. VISCOUS FLOW

FIGURE B.1. Alternative Paths Allowing Sintering Between Two Spherical Particles in Contact

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