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SUBJECT: Fission Product Transport Through Graphite Matrices

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FROM: R. B. Korsmeyer

ABSTRACT

The transport of fission products from points of origin in unclad graphite matrix-type fuels to the reactor circulating system involves, as one of the steps, diffusion through the graphite matrix to the fuel element surface. As pointed out by Rosenthal, the fraction of a given fission product chain actually reaching the fuel element surface will be small if the time for transport through the graphite is long compared to the half-lives of the volatile members. An important problem, therefore, is the determination of the effective transport rates of the various mobile elements and their daughter products of interest through various graphites suitable for use as fuel element compacts, as functions of temperature over the range of greatest immediate interest to reactor designers. The upper end of the range need not exceed about 1000°C.

The transport of helium and argon through various graphites has been the subject of considerable study by Watson, Evans, and others, and a preliminary investigation of the high temperature transport of some ordinarily non-volatile elements has been carried out by Saunders. This work is briefly reviewed in relation to the final problem and the areas in which further information is needed most by reactor designers is indicated.

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Larry E. Williams
Authorizing Official
Date: 05/23/2007

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DISCLAIMER

Nomenclature

α	empirical constant, erg/Å
β	empirical constant, erg/Å^6
A	superficial area normal to flow Q_1, cm^2
B_o	coefficient of viscous flow, cm^2
D_c	diffusion coefficient of non-volatile species c in the porous medium, cm^2/sec
D_{YZ}	mutual diffusion coefficient of gases y and z in the porous medium, cm^2/sec
D_{yz}	mutual free diffusion coefficient of gases y and z, cm^2/sec
E	porosity, dimensionless
$\hbar$	Planck's constant, erg-sec
k	Boltzmann's constant, erg/deg K
k_o	coefficient of viscous flow, millidarcy
K	apparent permeability coefficient, cm^2/sec
K_o	Knudsen flow coefficient, cm^2/sec
K_y	combined diffusion coefficient of gas y (Knudsen flow and mutual diffusion), cm^2/sec
L	nominal length of path over which Q_1 flows, cm
N_Y	mol fraction of gas y
n_Y	flow rate of gas y, mols/sec
n_T	net particle drift, mols/sec
P_1	pressure at which Q_1 is measured, dyne/cm^2 or atm.
$\bar{P}$	average pressure of gas flowing in the porous medium, dyne/cm^2 or atm.
ΔP	pressure drop associated with Q_1 in distance L, dyne/cm^2 or atm.

Nomenclature (Cont'd)

ΔP_Y	partial pressure drop of gas component y, dynes/cm^2 or atm.
Q_1	volume flow rate, cm^3/sec
Q_y	volume flow rate of gas y, cm^3/sec
q	tortuosity, dimensionless
R	gas constant, erg/mol "K or cm3atm/mol "K
r	atomic radius, Å
T	absolute temperature, °K
T_m	melting point, "K
$\bar{v}$	mean thermal molecular velocity, cm/sec
$\bar{v}_Y$	mean thermal molecular velocity of gas y, cm/sec
ΔV	energy required to fix the atom in the matrix, erg
x	point distance in direction of flow through porous medium, cm
μ	gas viscosity, dyne-sec/cm^2 or centipoise
Γ_i	rate of molecular jumping between surface sites, sec^{-1}

Fission Product Transport Through Graphite Matrices

The transport of fission products from points of origin in unclad graphite matrix-type fuels to the reactor circulating system involves, as one of the steps, diffusion through the graphite matrix to the fuel element surface. As pointed out by Rosenthal,(1) the fraction of a given fission product chain actually reaching the fuel element surface will be small if the time for transport through the graphite is long compared to the half-lives of the volatile members. An important problem, therefore, is the determination of the effective transport rates of the various mobile elements (e.g., Xe, Kr, I, Br, Cs, Rb, and Te) and their daughter products of interest (Sr, La, Ba, Ca, Nb, Y and Te) through various graphites suitable for use as fuel element compacts, as functions of temperature in the range of immediate interest to reactor designers.

Fuel element temperatures of greatest immediate interest do not exceed about 1000°C (i.e., 1800 to perhaps 2000°F). Costs of steam generators rise steeply for steam temperatures above about 1050°F because the higher temperatures require use of stainless steel instead of the lower-cost chrome-moly steels whose reliability and fabricating techniques for such service are so well established. Experience with fossil fired plants may show, eventually, that the economic advantage of the higher thermodynamic efficiency available at higher steam temperatures is offset

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(1) R. Korsmeyer, "Problems in Fission Product Disposition," Letter to A. P. Fraas, December 19, 1960. (Attached)

by the higher capital and maintenance costs of the stainless steel generators, These costs would be greater for nuclear plants. Thus, there is little incentive to raise coolant gas temperatures above about 1200°F, which is all that is required to reach the efficiencies of modern fossil-fired steam plants. Due to the higher inherent thermal conductivity of unclad, graphitic fuel elements and the relatively unrestricted heat transfer surface available (compared to clad elements), one can design these elements for rather low internal temperature drop (e.g., 100-150°F) for a total cost penalty of no more than perhaps 10% of the reactor capital charges,, or about 3% of the total plant capital charges, compared to the more idealized elements that operate at very high temperatures, Considering the orders-of-magnitude difference in difficulty of fission product retention, and other demands which must be placed on the materials of the fuel element at very high temperatures, the more attractive concepts of unclad gas-cooled reactors do not require fuel element temperatures above about 1000°C.

The transport of helium and argon through various graphites has been the subject of considerable study by Watson, Evans, and others,(2,3,4) and a preliminary investigation of the high temperature transport of some

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(2)Reactor Chemistry Division Annual Progress Report, January 31, 1960, ORNL-2931, pp 149-155.

(3)Gas-Cooled Reactor Project Quarterly Reports, March, June, September, and December, 1960, ORNL-2929, 2964, 3015, and 3049.

(4)Gas-Cooled Reactor Project Monthly Reports,.

ordinarily non-volatile elements has been carried out by Saunders.(5,6) The purpose of the present writing is to review this work briefly in relation to design problems and to indicate the area in which further information is most urgently needed.

Gas Diffusion Through Graphite

Background

The transport of gases such as Xe and Kr through porous media can be considered to consist of a mixture of two types of flow, viscous and diffusive, where the pore size is small and the pressure drop is insufficient to cause turbulence. For single gases flowing isothermally and in steady state, the flow is expressed by the following equation:(7)

$$Q_1 P_1 = A \frac{\Delta P}{L} \left[\frac{B_0}{\mu} \bar{P} + \frac{4}{3} K_0 \bar{v} \right] \qquad (1)$$

Eq. (1) includes a viscous flow term (Darcy's Law) characterized by the coefficient B_0, and a diffusive flow term involving the mean thermal molecular velocity, $\bar{v}$, and a coefficient K_0 (Knudsen flow). Although Knudsen flow applies strictly only where the mean free molecular path

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(5) Gas-Cooled Reactor Project Quarterly Report, June 1960, ORNL-2964.

(6) A. R. Saunders, "A Study of Fission Product Transport Mechanisms in High Temperature Gas-Cooled Reactor Fuel Elements," (Unpublished).

(7) P. C. Carman, Flow of Gases Through Porous Media, Academic Press, 1956, Chapter III.

is large compared to the pore diameter, the deviation from viscous flow (called slip flow) observed when the pore diameter approaches the mean free path is satisfactorily accounted for by the above expression. The coefficients B_o and K_o are characteristic properties of the porous medium.

The viscous flow is proportional to the average pressure level of the flowing gas in the porous medium and to the total pressure drop across the porous medium. The Knudsen flow is independent of pressure level and is proportional to the partial pressure drop (or concentration difference) of the gas component in question. For single gases, the pressure drop for Knudsen flow is, of course, the total pressure drop.

For pure Knudsen flow there is no interaction between different gases simultaneously diffusing through the porous medium, as the molecules collide essentially only with the walls. However, pure Knudsen flow is not realized under practical reactor conditions, and the mutual diffusion effect of different gases (e.g., coolant and f.p. gases) must be taken into account. The Knudsen coefficient in Eq. (1) is, therefore, replaced by a total diffusion coefficient. Since the resistances to flow (Knudsen and mutual diffusion) are additive, the terms (which correspond to conductances) are combined jointly as follows:

$$\frac{}{} = \frac{}{} \qquad \frac{}{\frac{\cdot}{3} K_o v_y} \tag{2}$$

where D_{YZ} is the mutual coefficient for the two gases y and z diffusing through the graphite and the diffusion rate of gas z is small compared to the forward flow. It is not necessary to measure D_{YZ} directly if the free diffusion coefficient for the two gases is known along with the tortuosity factor and porosity of the graphite, for Eq. (2) then becomes:

$$\frac{1}{K_y} = \frac{q}{\epsilon D_{yz}} + \frac{1}{4 K_o \bar{v}_y} \qquad (3)$$

(Actually, q and ϵ are measured from diffusion experiments in the case of some graphites, rather than from permeability experiments.)

Substitution of Eq. (3) in Eq. (1) to account for total viscous and diffusive flow of gas component y under isothermal conditions yields:

$$Q_y P_1 - Q_1 P_1 N_y = \frac{A}{L}\left[B_o \frac{\bar{P}}{\mu} \Delta P N_Y + K_Y \Delta P_Y\right] \qquad (4)$$

where ΔP is total pressure drop, ΔP_Y is the partial pressure drop of component y, and N_Y *is* the mol fraction of y in the stream Q_1.(8)

Since correction for so-called "pressure diffusion" (transport of heavier molecules in the direction of increasing pressure) is negligible for pressure drops occurring in reactor fuels,(9) it is possible to

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(8)Equation 4 implies that dP/dx is constant throughout the path length. For treatment of radial flow through cylinders, see H. L. Weissberg and A. S. Berman, "Diffusion of Radioactive Gases Through Power Reactor Graphite," KL-413, April 6, 1959.

(9)R. B. Evans, J. Truitt and G. M. Watson, "Superposition of Forced and Diffusive Flow in a Large Pore Graphite," om-3067, February 24, 1961.

calculate the isothermal transport rates of gaseous fission products through graphite from knowledge of the system pressure and temperature, the K_o, B_o, q and ϵ values for the graphite, and the viscosities and mutual free diffusion coefficients of the gaseous fission products and coolant gas, by making the conservative assumption that there is no interaction between the gases and the graphite (e.g., chemical reaction or chemi-sorption).

Reactor fuels do not operate isothermally, however, and the temperature gradient set up through the graphite could affect the fission product retention adversely through the phenomenon of thermal transpiration.(10) The transpiration of a light coolant gas, such as helium, toward the hot center of the fuel element might set up Poiseuille flow (viscous flow) in the centers of the larger pores in the opposite direction, thus tending to sweep fission product gases out of the fuel element. The combined phenomenon of thermal transpiration inward and viscous flow outward appears to be too complex to estimate indirectly, but net transport rates could be determined experimentally for particular combinations of graphites and gas pairs, as functions of temperature, pressure and temperature gradient.

Work Accomplished on Gas Transport

Essentially two types of graphite have been used in the gas diffusion studies to date: National Carbon AGOT needle coke and CEY. Tortuosities

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(10) P. C. Carman, ibid., page 140.

and porosities of these graphites have been measured, along with the viscous and Knudsen flow coefficients, using helium, argon and CO_2 gases. Also the apparent diffusion coefficients of helium and argon passing through these graphites have been measured. In general, these measurements were made at temperatures in the range of 20 to 100°C and pressures from 0.5 to 2 atm.

The permeability and diffusion measurements are summarized below:

	Permeability Coeff. K cm^2/sec		Apparent Diffusion Coeff. D_{YZ} cm^2/sec	
	He	A	He	A
AGOT	1.6×10^{0}	1.2×10^{0}	7.2×10^{-3}	2.3×10^{-3}
CEY	7.6×10^{-5}	2.6×10^{-5}	1.3×10^{-5}	0.4×10^{-5}

The average ratio of permeability-to-diffusion coefficients is 400 for AGOT and 6 for CEY. It is clear that the AGOT graphite is much too permeable for use in fuel compacts and is not considered further. The apparent diffusion coefficients for helium and argon are not numerically equal, as calculated, for the net drift of the gases (a phenomenon characteristic of the experimental procedure) was not taken into account. After accounting for the net drift by means of the following transport equation, the diffusion coefficients are essentially equal.

$$n_y = n_T N_y - D_{YZ} \cdot A \cdot \frac{P}{RT} \cdot \frac{dN_y}{dx} \qquad (5)$$

In Eq. (5), n_y represents the flow rate of component y (moles/sec), n_T is the net particle drift and N_Y is the mol fraction of component y. At any point, x, the individual flow rate of gas y is the sum of a mixed flow component and a mutual diffusion component. Although the flow rate of each component varies along x, the sum of the components remains constant with position and time.(9)

Permeability measurements in CEY graphite pipe have also been made using CO, in addition to He and A. The values measured at 25°C and 1.5 atm are shown below in terms of both the apparent permeability coefficients and the coefficient corrected for gas pressure and viscosity. The latter coefficient is expressed in the mixed units of the Darcy instead of c.g.s. units.

	He	A	cop
K cm^2/sec	7.78×10^{-5}	2.85×10^{-5}	3.37×10^{-5}
k_o - Millidarcy $(\frac{cm^2 - cp}{sec - atm} \times 10^{+3})$	2.08×10^{-3}	0.675×10^{-3}	0.506×10^{-3}

If k_o is plotted versus $1/P$ and extrapolated to $1/P = 0$, (i.e., infinite pressure) the coefficient, k_o, has the same value for all three gases, as would be expected for pure viscous flow.(11) Thus both viscous and slip flow exist, especially slip flow for helium.

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(11) W. T. Ward and H. V. Smith, ORNL-3049, ibid., page 301.

Work Remaining on Gas Transport

As nearly as can be foreseen at this time, the CEY graphite probably is fairly representative of practical graphites suitable for use as fuel element matrices,* although **it** is not a homogeneous material. The AGOT graphite, above, is clearly too permeable and, at the other extreme, the pyrolytic carbons are difficult to form, have some undesirable physical properties (e.g., considerable anisotropy in heat conductivity and apparently high shrinkage or warpage with fast flux exposure) and probably will be too costly for fuel element matrix material..

Using the CEY graphite as a reference stock material, the importance of thermal transpiration in the transport of fission gases should be examined. The same gas pair, helium and argon, would be suitable for the study. The temperature conditions of the experiment should closely simulate those to be expected in a reactor, both as to temperature level and gradient (e.g., T in range of 600 to 1000°C, dT/dx in range of 50 to 100°C/cm). If the evidence indicates that thermal transpiration is important, a further series of tests should be made with xenon and perhaps with krypton, depending on the confidence one has in the quantitative interpretation of the mechanism.

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*Graphites under investigation by General Atomic have helium permeabilities in the range of 10^{-5} cm^2/sec except: (a) Sic-coated graphite ~ 6.9 x 10^{-10} cm^2/sec, and (b) graphite for helium and cesium permeation tests at 1200°C, ~ 1 x 10^{-7} cm^2/sec. (GA-1982, December 31, 1960, Progress Report on 40 Mwe Prototype HTGCR, pages 56-57)

Incidentally, it would be well to re-run a couple of isothermal diffusion tests at this temperature level as a check against unanticipated changes in the graphite properties with temperature. A scouting check on the possibility of incurring irreversible changes in graphite properties with temperature is planned by the Reactor Chemistry Division in which permeation and diffusion tests are run at both room temperature and 200°C, before and after heating the graphite to high temperature.

As other apparently suitable graphites become available, their properties, as expressed by K_o, B_o, q and ϵ , will need to be determined, along with at least one high temperature check. If thermal transpiration is important, check determinations of the thermal coefficient expressed, perhaps, in terms of an additive fission gas diffusion coefficient, will need to be made.

Non-Volatile Element Transport Through Graphite

Transport of the essentially non-volatile fission products through graphite would not be expected *to* take place by simple gas diffusion characteristic of xenon and krypton. The fuel element temperature is high enough, however, that activated transport, such as the "random walk process" or two-dimensional gas diffusion(12,13) might be important.

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(12) G. Toos, Theoretical. Physics, Second Edition, Hafner Publishing Company, New York, 1950.

(13) P. C. Carman, ibid., pages 119-124.

Diffusion in AGOT graphite has been studied in one set of experiments. By utilizing an infinite plane geometry, rates of diffusion of Ba(6) and Sr(14) into AGOT graphite were measured at a temperature of 800°C. The data are not inconsistent with the earlier work of Findlay,(15) and Large & Walton.(16) The coefficients of diffusion for the fission products studied decreased with atomic radii in the manner expected of diffusion taking place as a random walk process.(12) The equations expressing this mechanism are as follows:

$$D_c = \frac{1}{2}\sum \Gamma_i (\Delta x_i)^2 \qquad (6)$$

where Δx_i is the distance between jumps. The rate of jumping is:

$$\Gamma_i = \frac{kT}{\hbar} e^{-\Delta v/kT} \qquad (7)$$

where

$$\Delta v = (\alpha/r) + (\beta/r^6) \qquad (8)$$

Δv is the energy required to fix the atom in the matrix.

Another group of experiments was started to study the overall migration rates of fission product elements in AGOT graphite at

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(14) ORNL-2964, ibid., pages 207-209.

(15) T. R. Findlay, "Diffusion of Fission Products from Graphite at 800°C," AERE-C/R-2683.

(16) Large & Walton, "Migration of Non-Gaseous Fission Products Through Graphite," AERE-C/M-346.

temperatures between 1200 and 1900°C. utilizing SrO, BaO, CeF_3, Y_2O_3 and ZrO sources. Rates of migration are much higher than at the lower temperatures, increasing by about 10^5 from 800 to 1500°C, thus indicating decreased energies of activation and movement in the open pores. The increases are not of the same order of magnitude for the different species. Yttrium and zirconium react with the graphite to form stable carbides and, as a result, the release rates are below 8%. On the other hand, barium is released rather rapidly with less than 5% remaining after heating to 1500°C for 16 hr.

A correlation of diffusion coefficient with temperature is indicated, to some extent, at the higher temperatures, by plotting the coefficient against the reduced temperature of the oxide in question, taking the oxide melting point as the datum. From such a plot of pairs of points for strontia, zirconia, barya, and yttria (one point) in the range of 1200 to 1700°C,(6) the following equation is derived:

$$D = 1.2 \times 10^{-3} \left(\frac{T}{T_m}\right)^{11.2} \qquad (9)$$

Due to the very large exponent, a considerable quantity of data is required to establish the numerical terms of the equation with sufficient accuracy to be useful. However, this type of plot is consistent with the increased transport by activated or surface diffusion to be expected in the neighborhood of the species' melting point.

Continuing Work on Non-Volatile Element Transport

The experimental work has indicated the possibility of a generalized correlation for diffusion of fission products at temperatures below, say, 1000°C, but there is no accurate correlation evident at higher temperatures.

up to 1000°C

As already indicated, a thorough knowledge of fission product transport behavior in graphites of fairly low permeability (order of 10^{-5} cm^2/sec) as a function of temperature up to about 1000°C is the most valuable and most urgent information that can be given to the reactor designer on this subject. The work Saunders started should be augmented and extended to include measurements of the diffusion coefficients of all of the other non-volatile elements listed at the beginning of this memorandum. If the applicability of the general equations characteristic of the random walk process can be established quantitatively at an early date, it may be unnecessary to measure the diffusitivities of all of the elements over the complete temperature range. Whereas further measurements in AGOT graphite may be desirable for purposes of correlation, the effort should be directed mainly toward obtaining the information in graphites of indicated usefulness for fuel element compacts. These are graphites having permeability coefficients in the range of 10^{-5} cm^2/sec. The CEY graphite is a commercial product within this class, although it seems to lack homogeneity.(17) Until other, better graphites become

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(17) ORNL-3049, ibid., page 300.

available with satisfactory indications of quality control and reasonable cost, the work should proceed with the CEY graphite.

In a similar manner, in-pile experiments should be conducted to determine the influence (if any) of radiation on the fission product transport, and particularly the perhaps indirect effect of fuel burn-up,*

Above 1000°C

As already discussed, there is little immediate interest in measurements of fission product transport through graphite at temperatures substantially above 1000°C. The economic incentive from potential reduction in capital charges is small and uncertain, and fission product transport is known to rise by orders of magnitude with significant temperature increases. On the other hand, the costs associated with maintenance of the reactor circulating system are influenced critically by the choice of direct versus indirect maintenance procedures. The designer is willing, therefore, to make concessions in almost every direction that will increase the fission product retention of the fuel.

Tests with Pyrolytic Carbon

In this memorandum the value of transport measurements through pyrolytic carbon has been discounted because the use of this material for the matrix of unclad fuel elements does not presently appear practical, However, the use of pyrolytic carbon (or other ceramics such as Al_2O_3,

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* It appears that the in-pile aspects can be included in the present programs for sweep capsule tests of coated fuel particles and loop tests for fission product deposition.

BeO, etc.) as coatings on small fuel particles embedded in the graphite matrix appears very attractive as a possible "front-line" barrier to fission product escape from the fuel itself. Knowledge of high temperature transport of fission products through pyrolytic carbon under this service is exceedingly important, but it is outside the scope of the present discussion.

Silicon-Carbide Coated Graphite

Although Si-C coated graphites show extremely low permeability, it is felt that transport studies would not be worth while until a coated fuel element is designed which will not crack under irradiation (perhaps associated with thermal cycling) as the cracking is characteristic of those made to date,

Summary of Work Needed

1. Determine the importance of thermal transpiration as a factor in fission product transport in CEY graphite, using helium and argon.

2. If thermal transpiration is important, determine the corresponding coefficients for helium and xenon or krypton.

3. Check the isothermal diffusion coefficients of helium and argon in CEY graphite at high temperature (e.g., 1000°C).

4. As other low-permeability graphites become available, determine K_o, B_o, q and ϵ for each type.

5. Determine the transport rates of the "non-volatile" fission product elements in CEY graphite at temperatures up to ~ 1000°C to

determine the applicability of the generalized correlation based on the "random walk" process (Eq. 6) or to develop some other correlation if Eq. (6) does not hold.

6. If the generality of Eq. (6) or some other correlation is established, determine the transport rates of enough of these elements in other graphites (as they become available) to permit a generalized correlation for all the fission products of interest.

INTRA-LABORATORY CORRESPONDENCE

OAK RIDGE NATIONAL LABORATORY

December 29, 1960

To: A. P. Fraas

Subject: Problems in Fission Product Disposition

I have reviewed the above subject with Murray Rosenthal in connection with the program on advanced reactors, At Murray's suggestion I have written up the attached statement of the problems he has had in mind and which he has discussed at some length with George Watson and co-workers in Reactor Chemistry Division.

R. B. Korsmeyer

RBK:dh
Attachment

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Fission Product Release and Disposition from Graphite Matrix Fuels

The transport of fission products from points of origin to surfaces in the reactor circulating system can be considered to take place in three steps: (1) escape from the fuel particle by fission recoil and/or solid state diffusion, (2) fluid diffusion through the graphite pores to the fuel element surface, and (3) transport by the gas stream to the surface of some component in the reactor system.

Step 1. Efforts to minimize release by the first step have led to the incorporation of fairly large, dense fuel particles in the graphite. The particle size is large enough to reduce escape by fission recoil to a small fraction of that generated, and coatings are applied to the particles to reduce the escape further by providing a dense barrier through which migration must be essentially by the comparatively slow process of solid state diffusion as long as the barrier remains intact. Thus the very short-lived products die out before escape, and the "non-volatile" products diffuse too slowly to be of concern.

Step 2. The longer lived fission products which experience has shown have some mobility, and hence escape the fuel particle in significant amounts, are Xe, Kr, I, Br, Cs, Rb, and Te. Having escaped the fuel particle and lodged in the pores of the graphite matrix, these elements and at least some of their daughter products can diffuse through the graphite pores to the fuel element surface by processes so much faster than solid state diffusion that their release to the circulating gas has been assumed almost instantaneous, in studies of the contamination problem.(1) However, the fraction of a given fission product chain actually reaching the fuel element surface will be small if the time for diffusion through the graphite is long compared to the half-lives of the volatile members. An important example is the mass 140 chain which has the decay scheme shown below:

$$16s\ Xe \rightarrow 66s\ Cs \rightarrow 12.8d\ Ba \rightarrow 40.2h\ La \rightarrow 32d\ Ce \rightarrow \text{stable } Br$$

The La^{140}, being a hard gamma emitter, is important with respect to shielding and maintenance, and its effective half-life is fairly long because of its 12.8d Ba^{140} precursor. If the La and Ba can escape only by migration of Xe and Cs, the effective half-life for escape is only 82 seconds. An average hold-up of 10 minutes would mean that only 0.6% of the mass 140 chain which is evolved from the fuel would escape the graphite.

Recent British work(2) indicates that the releases from graphite one might expect by extrapolation of the NAA data > 1500°C to lower temperatures(3) do not occur. Their data, however, can be explained if the longer-lived mobile precursors of the activities detected outside of the graphite are taken into account.(4) An important problem, therefore, is

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(1) W. B. Cottrell, et al., ORNL-2653.

(2) Large & Walton, "Migration of Non-gaseous Fission Products Through Graphite," AERE-C/M-346.

(3) W. B. Cottrell, et al., technique in ORNL-2653.

(4) M. W. Rosenthal's notebook on PBR, page 117.

the determination of the diffusion rates of the various mobile elements listed under Step 1 and their daughter products of interest (e.g., Sr, La, Ba, Ca, Nb, Y and Te) through various graphites suitable for use as fuel element compacts, as functions of temperature in the range of, say, 600° to 1200°C.

George Watson has pointed out in discussions with Murray Rosenthal that there is experimentation in this field both in the United States and abroad using irradiated fuels of interest. Rosenthal has suggested that much might be learned about this problem using non-radioactive or low activity forms of the above chemical elements and thus avoid having to handle irradiated fuel, since the problem is concerned with chemical and physical properties. He wondered if suitable graphite specimens couldn't be prepared by cyclotron bombardment with important nuclides, and the release rates determined by mass spectrometer analyses or activity measurements. Watson and co-workers independently had been considering a similar approach using the calutron, but there is some doubt whether the energies available from the calutron are adequate to imbed the nuclide deeply enough in the graphite. Watson, et al., are studying the problem with the intention of coming up with an experimental program.

In the meantime the problem can be attacked substantially by a calculation of the average time required for a volatile fission product (e.g., Xe, Kr, I, Br, Cs, Rb, and Te) to escape from a given type of graphite fuel element, based on the data being obtained by Bob Evans on diffusion of noble gases through graphite.

Step 3. The behavior of those fission products which do escape into the gas stream is of particular importance in estimating shielding and maintenance problems. Of interest is information on whether the non-volatile species can move through the reactor circulating system and, if so, under what conditions they are deposited. To what extent will the halides, alkali metals and tellurium be adsorbed on surfaces which are above the condensation temperature, and at what rate would it be possible to make a significant reduction in, say, the Cs and Te concentrations in the primary system by using a 1% by-pass cleanup system? Watson has suggested the possibility of providing ceramic-type filters in the main gas stream which have surfaces of high affinity for particular fission products. Although, as pointed out by others, the chemical behavior of elements in such extremely low concentrations as the fission products considered here is not generally predictable from known chemical properties, it is possible that this subject could also be investigated without the use of irradiated fuel. In particular, the chemical thermodynamicists should be working on establishing the equilibrium surface concentration of these fission products as a function of surface temperature, nature of the surface, gas temperature and concentration, first for single elements and then in combinations of high chemical activity (e.g., Cs and I).