CENTRAL RESEARCH LIBRARY
DOCUMENT COLLECTION

MARTIN MARIETTA ENERGY SYSTEMS LIBRARIES
3 4456 0364420 6

ORNL-3145
UC-80 - Reactor Technology

A STUDY OF FISSION PRODUCT TRANSPORT MECHANISMS IN HIGH TEMPERATURE GAS-COOLED REACTOR FUEL ELEMENTS

A. R. Saunders

CENTRAL RESEARCH LIBRARY
DOCUMENT COLLECTION
LIBRARY LOAN COPY
DO NOT TRANSFER TO ANOTHER PERSON
If you wish someone else to see this document, send in name with document and the library will arrange a loan.

OAK RIDGE NATIONAL LABORATORY
operated by
UNION CARBIDE CORPORATION
for the
U.S. ATOMIC ENERGY COMMISSION

Printed in USA. Price $0.75. Available from the
Office of Technical Services
Department of Commerce
Washington 25, D.C.

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

ORNL-3145

Contract No. W-7405-eng-26

REACTOR CHEMISTRY DIVISION

A STUDY OF FISSION PRODUCT TRANSPORT MECHANISMS IN HIGH TEMPERATURE GAS-COOLED REACTOR FUEL ELEMENTS

A. R. Saunders

DATE ISSUED

JUN 29 1961

OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee
operated by
UNION CARBIDE CORPORATION
for the
U.S. ATOMIC ENERGY COMMISSION

SYSTEMS LIBRARIES

3 4456 0364420 6

CONTENTS

Abstract 1

Introduction 2

Experimental 5

Rates of Diffusion for Fission Product Elements in Graphite Below 1000°C 5

Rates of Diffusion for Fission Product Elements in Graphite Above 1000°C 7

Discussion 10

Survey of Earlier Experimental Work 10

Structure of Matrix and Mechanisms of Fission Product Entry 13

Random Walk Theory of Solid State Diffusion 15

Activation Energies for Fission Product Diffusion . 21

Mechanisms of Transport Above 1000°C 22

Chemical Reactions in Graphite 24

References 28

A STUDY OF FISSION PRODUCT TRANSPORT MECHANISMS IN HIGH TEMPERATURE GAS-COOLED REACTOR FUEL ELEMENTS

A. R. Saunders

ABSTRACT

The experimental work on the diffusion of fission products in ceramic matrices at high temperatures is reviewed and the possible mechanisms of mass transport are discussed. Preliminary data are given on diffusion in these systems.

It is shown that the rates of diffusion for fission products introduced by a recoil process into the moderator lattice can be correlated based on a random walk solid state diffusion model. An expression is derived for the coefficient of diffusion in terms of the lattice parameters giving the coefficients of diffusion for different atomic species as an exponential function of the atomic radii.

The possibility of other controlling mechanisms above 1000°C and the effect of carbide formation and other reactions is evaluated.

INTRODUCTION

Containment and removal of the fission products produced in a power reactor is an important problem in the operation of nuclear reactors. At lower temperatures metal cladding of the fuel elements is used to contain the fission products. Production of fuel element claddings impervious at the operating temperatures of the high temperature gas-cooled reactors is a much more difficult technical problem for which a satisfactory solution can be found only through an understanding of the mechanisms of mass transfer under reactor operating conditions.

Gas-cooled reactors can be operated at relatively high temperatures with thermal efficiencies exceeding those of fluid fuel reactors. The design concepts for fuel element operating temperatures of 2000°F or higher utilize ceramic matrices fabricated from graphite or refractory oxides containing the fuel as uranium carbide or oxide.[1] Examples of this approach are the Oak Ridge National Laboratory's HGCR-1[1] with fuel elements of 200-micron particles embedded in graphite, the Philadelphia Electric-General Atomic-Bechtel Corp. HTGCR[2] fueled with cylinders of graphite containing the ThC-UC fuel, Sanderson and Porter-Alco Products Inc.'s Pebble Bed Reactor, PBR,[3] as

well as others.

The importance of a fission product in relation to the reactor operation and, hence, the need for its containment within the fuel is determined by its fission yield, half-life, gamma-activity and neutron absorption cross section. The fission products with long half-lives and high energy gamma-emissions, generated at a high yield, will cause the greatest radioactivity and contamination problems in the coolant gas stream to the extent that they escape from the fuel element.

Examination of a plot of fission yields of the products of U-235 fission[4] shows that the elements with mass numbers of 85-105 and 130-150 have the highest fission yields. Fission yields for these products range from 2 to 6 %.

Based on the amount of gamma-activity released as high energy decay products with long half-lives, decay chains 87-95, 129 and 131-143 are the most significant.[5,6] Examination of these fission chains indicates that Rb, Y, Sr, Zr, Nb are the most important radioactive products of chains 87-95, Sb, Te, and I of chain 129 and Sb, Te, I, Xe, Cs, Ba, Sr, La and Ce for chains 131-143.

Fission chains 147-161 produce rare earths with high neutron absorption cross sections. Rate of removal of

these elements will influence the neutron economy of the reactor.

The fission products formed in an unclad ceramic fuel element will diffuse from the fuel into the gas stream by solid state diffusion. Fragments of sufficiently high kinetic energy will recoil into the moderator matrix. The fission products reaching the moderator will be transported through this matrix by pore, surface or solid state diffusion. The controlling mechanisms are different for each of these processes.

In this report the experimental data obtained on high temperature solid state and surface diffusion of fission product elements in graphite and some of the mechanisms controlling these processes will be discussed and the results compared with some of the earlier work. The experimental work is of exploratory nature and it was designed to study a number of idealized systems and obtain comparisons with earlier work giving an insight into the mechanisms controlling fission product transport in fuel element matrices.

EXPERIMENTAL

Rates of Diffusion for Fission Product Elements in Graphite Below 1000°C

In this phase of the work, classical diffusion experiments were carried out utilizing an infinite plane geometry for measurements of the rates of diffusion of strontium and barium in graphite. The concentration gradients were determined by measurements of the gamma activity distribution in the samples.

Source matrix pellets of reagent grade BaO, SrO tagged with Ba^{133} and Sr^{85} were placed in contact with AGOT graphite pellets and held at constant temperature for several weeks in a furnace equipped with a Pyrovane temperature controller, passing a stream of helium gas over the diffusion couple during the experiment. A well-type scintillation counter was used to determine the gamma activities of source and matrix specimens before and after the experiment.

The coefficients of diffusion were evaluated using the solution of the diffusion equation for an infinite cylinder of unit cross section.[7] The one-dimensional form of the diffusion equation is

$$\partial C/\partial t = D\partial^2 C/\partial x^2 \qquad (1)$$

where C is concentration of the diffusing species, t is time, D is coefficient of diffusion and x is distance coordinate in the direction of the diffusion flux.

For the boundary conditions

$$C = C_0 \quad \text{for } x < 0 \quad \text{at } t = 0$$

$$C = 0 \quad \text{for } x > 0 \quad \text{at } t = 0$$

the solution is

$$C\ (x,t) = (C_0/2)\ \mathrm{erfc}\ (x/2\sqrt{Dt}\ )\ . \qquad (2)$$

Differentiation of this solution with respect to x and substitution of the result into the flux equation gives

$$J = -D\ (\partial C/\partial x)_{x=0} = C_0\ \sqrt{D}/2\ \sqrt{\pi t}\ . \qquad (3)$$

Integrating for a time period t over a cross sectional area A gives the total amount transferred as

$$S = A \int_0^t J\ dt = A\ C_0\ \sqrt{Dt/\pi} \qquad (4)$$

or solving for D

$$D = S^2\ \pi/A^2\ C_0{}^2 t\ . \qquad (5)$$

Using this equation the coefficient of diffusion can be determined for a known geometry from the initial source and final matrix compositions for diffusion over a fixed time period.

Experimental diffusion data obtained by this technique are given in Table I. An examination of the data indicates

migration through the matrix by a relatively slow process.

Rates of Diffusion for Fission Product Elements in Graphite Above 1000°C

Additional experimental data were obtained on the rates of transport of fission product elements at temperatures between 1000 and 1800°C. At these temperatures the rates of migration are considerably higher and reactions with the graphite matrix are more likely to take place.

In order to facilitate these studies diffusion of one component was measured in a well defined geometry. The release rates were measured from an enclosed cylindrical source containing the diffusing oxide through the surrounding AGOT graphite matrix. The specimens used in the experiments were 1/2-in. o.d. by 3/4-in. cylinders with a .062-in. by 1/4-in. source. The choice of a 1:8 inside to outside diameter ratio allows use of the equations for diffusion through a plane sheet as a close approximation.[8]

The source materials used in the experiments were BaO, SrO, Y_2O_3 and ZrO_2 with Ba^{133}, Sr^{85}, Y^{91} and Hf^{181} tracers. Selection of these elements gives a cross section of several of the important fission product decay chains.

The enclosed hollow cylinders containing the source material were placed in a high temperature vacuum furnace

and heated at constant temperature for a fixed time period. Measurement of the gamma activities of the cylinders before and after the experiment in a well-type scintillation counter allows determination of the rates of mass transport from the source through the porous graphite membrane. The experimental data obtained are given in Fig. 1.

These data indicate two types of behavior for these systems. Either all activity gradually diffuses out from the source through the membrane or after some initial release of activity a more or less constant portion remains in the cylinders.

The effective coefficients of diffusion can be calculated from these experiments from the expression

$$\frac{dQ}{dt} = DA \left.\frac{dC}{dr}\right]_{r=b} \qquad (6)$$

where Q is the activity released over a time period t from the cylinder, D is the coefficient of diffusion, A the area through which diffusional transport takes place, b the outer radius of cylinder and C the concentration of the diffusant at point r along the radius of the cylinder r. Assuming an average ΔC the coefficient of diffusion can be calculated when dQ/dt is measured in a known geometry.

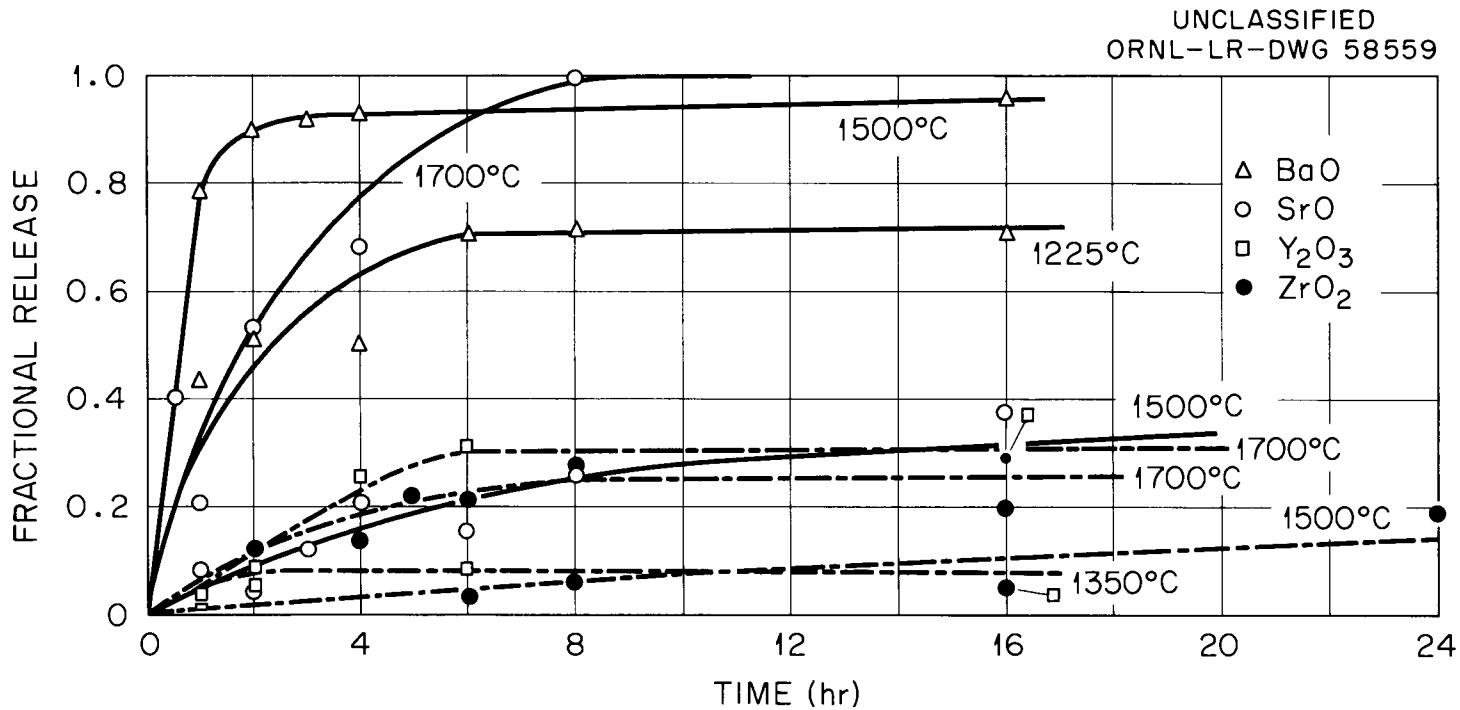

Fig. 1. Diffusion of Fission Element Oxides in Graphite Above 1000°C.

Values of the coefficients of diffusion D_{eff} and the energies of activation, ΔE derived from the experimental data are given in Table II. The considerably higher order of magnitude of the values indicates a mechanism of transport different from the diffusion process at lower temperatures.

DISCUSSION

Survey of Earlier Experimental Work

The measurement of fission product release rates from high temperature refractory fuel elements was the subject of several experimental investigations.[5] Release of fission products was studied from uranium oxide and carbide alone or in combination with graphite and other moderator matrices.

Transport in the moderator matrix will be controlled by the processes transferring the diffusant into the matrix and the structure of the matrix. Transfer of diffusing fission products can take place either by (1) recoil of the high kinetic energy particles from the fuel into the matrix or (2) diffusion from the fuel. The relative importance of these two transfer processes will vary depending on the structure and size of the fuel particles.

Table I. Results of Solid State Diffusion Experiments

Source Material	Coefficient of Diffusion $cm^2/sec \times 10^{-12}$ 800°C	850°C	950°C	1050°C	Energy of Activation, kcal/g mole
SrO	-	.882	1.03	41.60	52.1
BaO	.074	-	-	-	-

Table II. Rates of Mass Transport in AGOT Graphite Above 1200°C

Diffusant	Temp. °C	Effective Coefficient of Diffusion D_E, $cm^2/sec \times 10^{-5}$	Energy of Activation, ΔE, kcal/g mole
BaO	1225	1.11	16.4
BaO	1500	3.72	
SrO	1500	0.32	119.8
SrO	1700	2.93	
Y_2O_3	1350	0.62	6.3
Y_2O_3	1700	0.88	
ZrO_2	1500	0.024	136.8
ZrO_2	1700	1.23	

A large portion of the fission products will become imbedded in the moderator lattice and this portion will be released by a slow solid state diffusion process. Above 1000°C more rapid surface or grain boundary diffusion processes may also play a part in fission product transport in the moderator.

Largest portion of the experimental data on diffusion of fission products was accumulated in the temperature range between 1000 - 1900°C. The group with North American Aviation, Inc. studied the release of fission products in this range for graphite impregnated with a molten uranium salt converted subsequently by heating in vacuum up to 1900°C into the carbide. Then the specimens were irradiated and the rates of fission product release were measured by heating in vacuum after decay of the short half-life activity.

Cubicciotti[9] measured the rates of xenon release and Doyle[10] determined the rates of release for most of the other important fission products in uranium carbide impregnated graphite. Young and Smith[11,12] obtained more experimental data in the same area. Additional experimental work by Young[13] furnished data on the fission product retention on deuteron bombardment at 1000 - 1950°C.

Diffusion of fission products from uranium impregnated graphite and magnesium oxide matrices at 1500°C to 3000°C was studied by Cowan and Orth[14] at the Los Alamos Scientific Laboratory.

More recently Findlay[15] of the Atomic Energy Research Establishment, Harwell, obtained experimental fission product diffusion data at 800°C on molten uranyl nitrate impregnated irradiated graphite after leaching out the fission products and uranium with nitric acid and measuring the rates of diffusion on the fission products retained in the graphite.

Structure of Matrix and Mechanisms of Fission Product Entry

The structure of the matrix in which diffusional transport takes place will have an important influence on the mechanism of the migration process. The structure of graphite[16] can be described as an essentially random agglomerate of particles. The particles are composed of randomly arranged crystallites of about 150 Å cross section and up to 1000 Å long. The pore volume of about 20 - 30% is distributed around the particles. Within the crystallites the graphite has a layer structure forming a two

dimensional hexagonal mesh structure with each carbon atom bound to three other atoms. The distance between the carbon atoms in the layer is 1.42 Å and the separation between layers is 3.35 Å. Alternate layers are displaced relative to each other somewhat. The bonds within the layers are covalent while the layers are held together by weaker van der Waals' forces.

The processes resulting in the introduction of the diffusing elements into the solid matrix will have an effect on the mechanisms by which diffusional transport of these species will take place. Fission product recoil entry into the graphite lattice can be interpreted based on the hot-zone model of nuclear recoil initiated reactions proposed by Harbottle and Sutin.[17] The energy of the recoils is dissipated by producing displaced atoms and heating up to the melting point a small region of the matrix for a time period of the order of 10^{-11} sec. Some chemical reactions may have a chance to proceed during this period or the subsequent quenching period. Displacement of some atoms will produce vacancies or holes in the graphite crystal lattice and can introduce carbon atoms in interstitial positions. The fission fragments will be trapped in some locations in the lattice eventually. The

resulting structure would contain fission product elements substituted in positions in the graphite lattice as well as interstitially. In addition a number of more-or-less randomly distributed vacancies will be present in the graphite lattice. The radiation flux will produce similar effects in addition to the fission recoil penetration.

Random Walk Theory of Solid State Diffusion

Diffusional transport can be viewed as random walk of the diffusant through a matrix. Rayleigh demonstrated that using this model the diffusion equations can be derived and Zener[18] showed that the coefficient of diffusion can be interpreted in terms of the statistical theory of reaction rates.

Let us assume that diffusion takes place along the x-axis parallel to the crystal axis in jumps of length Δx of equal probability in both +x and -x directions with a mean time t between the jumps. Examining a segment of unit area for n(x,t) atoms passing through the x plane at time t the change for a time interval δt will be

$$n(x,t+\delta t) - n(x,t) = \left[n(x+\Delta x,t) - n(x,t) + n(x-\Delta x,t) - n(x,t) \right] \delta t/2t \qquad (7)$$

For $\delta t \rightarrow 0$ and $\Delta x \rightarrow 0$ the difference equation can be written as

$$\partial n/\partial t = D\ (\partial^2 n/\partial x^2) \tag{8}$$

with

$$D = (\Delta x)^2/2t = \frac{1}{2}\ (\Delta x)^2 R \tag{9}$$

Then the coefficient of diffusion for different jump lengths can be expressed as

$$D = \frac{1}{2} \sum_i R_i\ (\Delta x_i)^2 \tag{10}$$

where R_i is the rate at which an atom makes a particular jump i to the distance Δx_i.

From the statistical theory of reaction rates of Eyring[19] the expression for the mean rate of jumps can be given as

$$R = (kT/h)\ F^*/F \tag{11}$$

where F* is the partition function for the activated complex of the crystal lattice and diffusant atoms for the diffusing atom confined to a plane passing through the potential barrier and the two neighboring sites. No similar restraint is placed on the partition function F. The function F can be expressed as

$$F = F_L\ F_1{}^* \tag{12}$$

where F_L is the partition function of a single linear

oscillator representing the dominant part of F for vibration in a given site. For a linear oscillator F_L can be expressed as

$$F_L^{-1} = 2 \sinh (h\nu/2kT) \tag{13}$$

which gives for the cases of interest as good approximation

$$F_L^{-1} = h\nu/kT \tag{14}$$

Hence the rate of jumps can be set equal to

$$R = (kT/h)\ e^{-\Delta V/kT} = \nu\ e^{-\Delta V/kT} \tag{15}$$

where k is the Boltzman constant, h the Planck constant, T absolute temperature, ν the vibrational frequency of the atom held in a site in the lattice and ΔV the energy required to surmount the energy barrier between two successive positions in the lattice. For atoms held in a lattice by partially ionic forces this energy would be given by the Born-Lande equation[19]

$$U_0 = \frac{M\ Z_1 Z_2 e^2}{r} - \frac{B}{r^n} \tag{16}$$

or for the lattice energy at the equilibrium distance in the crystal

$$U_0 = \frac{M\ Z_1 Z_2 e^2}{r_0} (1 - \rho/r_0) \tag{17}$$

Here r_0 is the crystal radius, Z the ionic charge, M the Madelung constant which is related to the crystal structure

parameters, e, electronic charge, B, M and ρ constants.

In these expressions the first term represents the electrostatic attraction and the second gives the repulsive energy due to overlapping of the valence bands.

Combining the constants in Eq. (17) gives

$$U_0 = \frac{A\ Z_1 Z_2}{r_0} (1 - \rho/r_0) \qquad (18)$$

indicating that the lattice energy will be inversely proportional to the crystal radius.

For the atoms held in positions in the lattice the energy required for migration will be proportional to the energy barrier represented by the lattice forces. Hence

$$\Delta F \propto \frac{A\ Z_1 Z_2}{r_0} (1 - \rho/r_0) \qquad (19)$$

and the coefficients of diffusion should be a function of the crystal radii of the intermediates or those of the diffusing atoms.

The final expression for the coefficient of diffusion can be written as

$$D = \frac{1}{2} \sum_i (\Delta x_i)^2 \frac{kT}{h} e^{-AZ_1 Z_2 (1-\rho/r_0)/kTr_0} \qquad (20)$$

Here the simplifying assumption is made that the rate of jumps will be the same in any position in the lattice.

This may be only an approximation for a matrix containing a large number of fission fragments.

An examination of Fig. 2 showing the coefficients of diffusion for fission product recoils in graphite as given by Large and Walton[15] indicates that these coefficients of diffusion are a function of the atomic radius of the species as indicated by Eq. (20). The agreement is good considering that the data cover a range of 10^6 for the coefficients of diffusion. Xenon and cesium show the same relationship but the coefficients of diffusion are higher by a factor of 10^3. These elements would be above the boiling point at 800°C and this may account for their increased mobilities in the lattice. The behavior of iodine which is also above the boiling point can be attributed to this and its relatively long half-life tellurium precursor. The relationship is valid using either the atomic or the ionic radii of the diffusing species. The atomic radii were preferred since the structure of the activated complexes is not known and the bonds may be partially covalent in the intermediate states. In addition the radii will be somewhat different for the different isotopes. This effect may be significant in estimation of the rates of diffusion for the species in different fission chains but the available data are not sufficiently accurate for

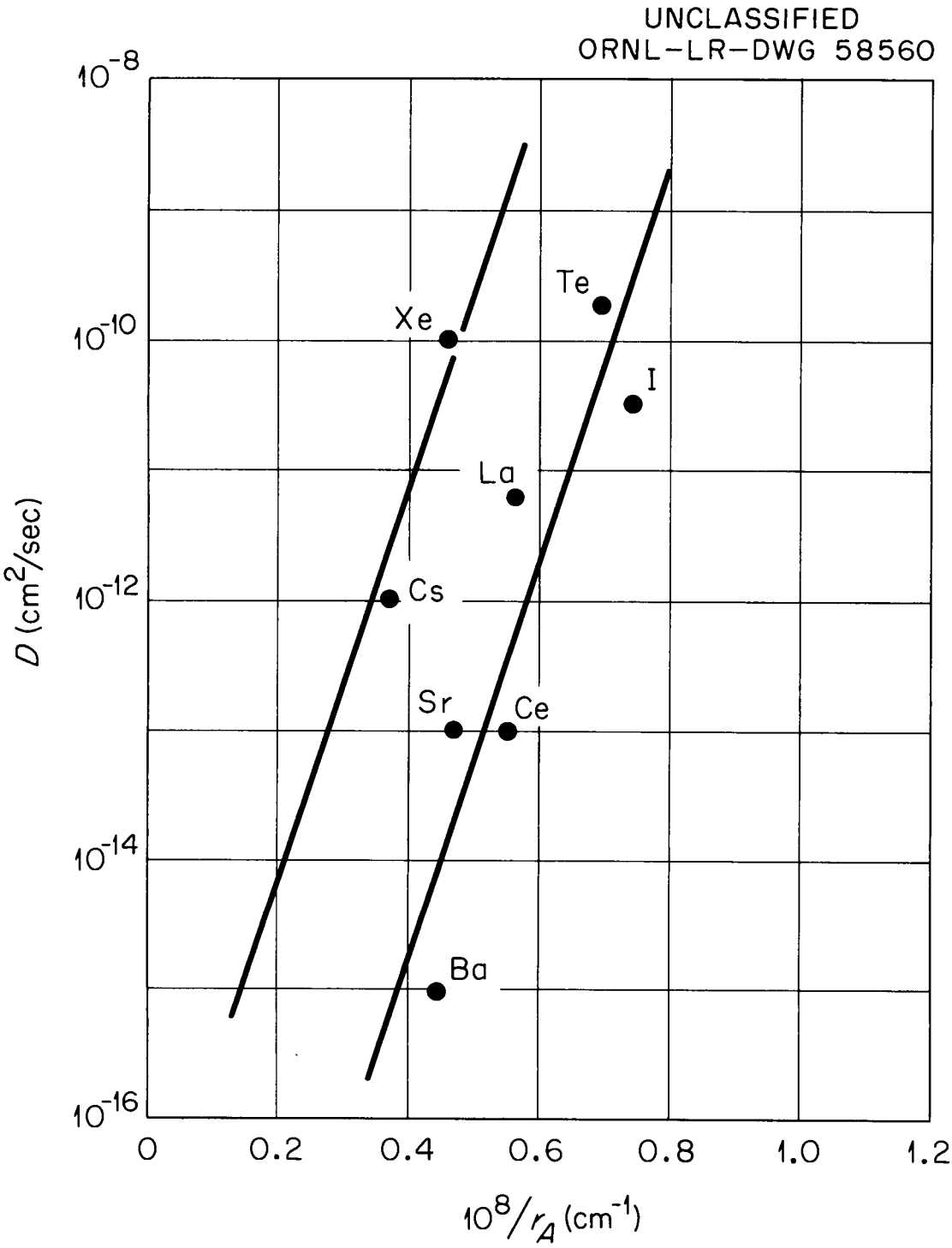

Fig. 2. Rates of Fission Product Diffusion in Graphite at 800°C as a Function of the Atomic Radii of the Diffusants.

taking it into account at the present time.

The slope of the coefficients of diffusion versus atomic radii plot can be utilized to calculate the Madelung constant for a particular intermediate configuration by Eq. (20). This calculation based on a slope value of 3.85×10^4 and approximate values for the unknown constants from empirical lattice energy correlations yields a value of 95 for M. The value is of the correct order of magnitude for a structure where a diffusant atom would occupy a position in the graphite lattice. With a wider range of additional experimental data available this relationship can be further tested, the jump lengths and frequencies calculated.

Activation Energies for Fission Product Diffusion

Further understanding of the mechanisms of fission product diffusion can be obtained through consideration of the energies of activation for diffusional transport. The available values for the energies of activation range from 20 - 100 kcal/mole for diffusion in graphite. Values given by Dienes[21] for different mechanisms of self-diffusion in graphite are somewhat higher. The calculated values for vacancy, direct interchange and interstitial diffusion are 119.4, 90.4 and 417.0 kcal/mole respectively.

However it was observed that the activation energies for chemical diffusion are usually lower than those for self-diffusion. Johnson[22] interpreted this as an indication that less work is required to produce a lattice vacancy adjacent to a solute or impurity atom at a particular lattice site. This in combination with the enhanced vacancy formation rate under irradiation would account for these lower values for the energies of activation for fission product diffusion in graphite or other matrices.

Mechanisms of Transport Above 1000°C

At temperatures above 1000°C fission product transport is controlled by processes that are different from the rate controlling steps at lower temperatures. Examination of the diffusion data obtained at these temperatures indicates diffusion combined with chemical reactions for Y and Zr. The comparatively low values obtained for the activation energies from the data indicate that diffusional transport may take place at least partially on the free surfaces of the crystallites rather than through the crystal lattice.

As Fig. 3 indicates the data for the effective coefficients of diffusion for all the systems studied can be correlated as a function of the reduced temperature T/T_M.

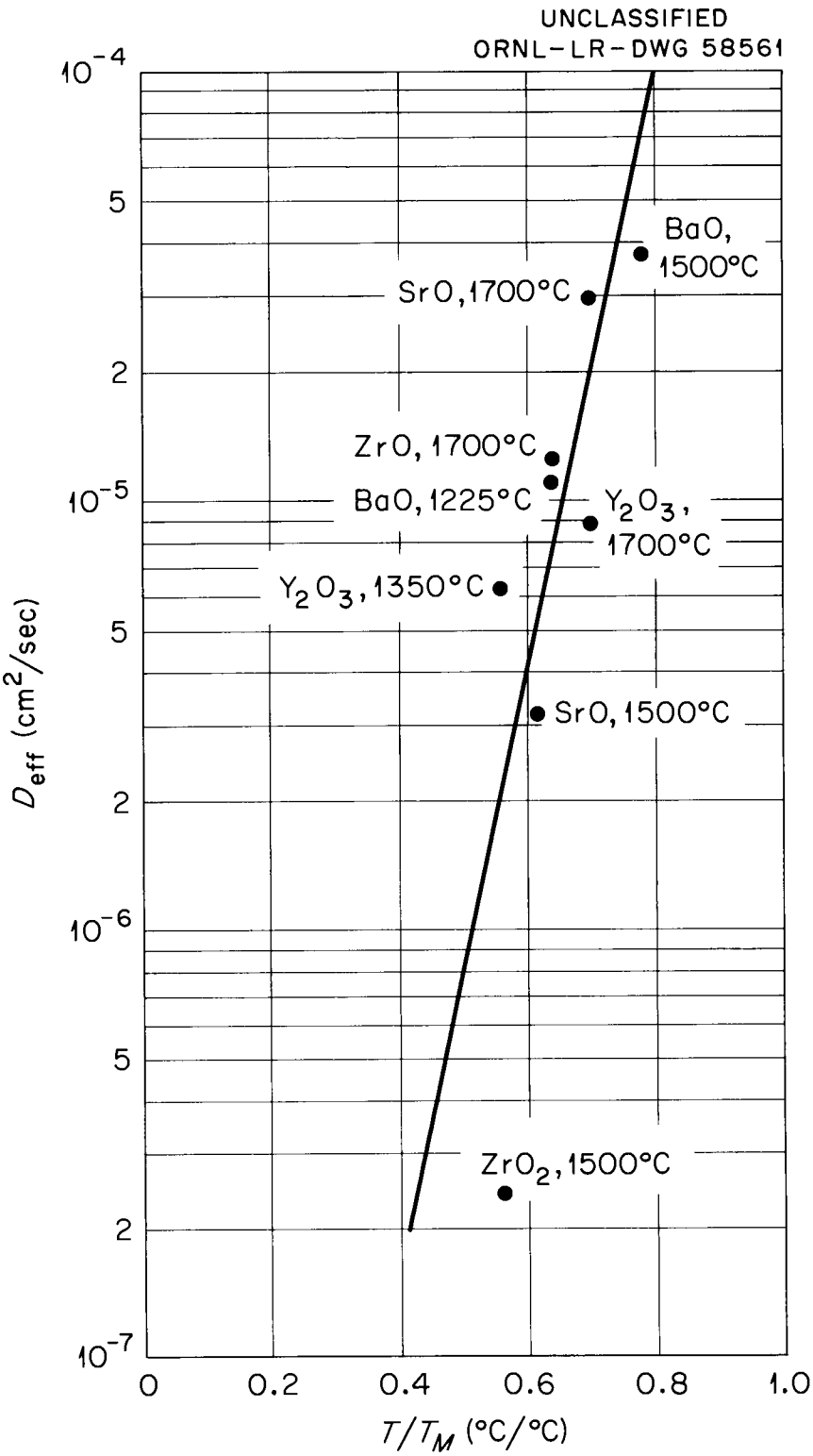

Fig. 3. Diffusion of Metal Oxides in Graphite as a Function of the Melting Point.

Diffusional transport can be described in this system as a two dimensional random walk through the surface imperfections. The mobility of the diffusing species will be proportional to the T/T_M ratio or the kinetic energy of the migrating species.

Chemical Reactions in Graphite

In addition to the aspects of fission product diffusion already discussed, the effects of chemical reactions between the diffusants and matrix should be also examined.

Graphite can form intercalation and residue compounds with a large number of elements and their salts. Recent work by Croft,[23] Hennig,[16] Rudorff[24] and others indicates that chemical species that can act as electron acceptors or donors will form ionic or covalent bonds with graphite. Since the conduction and valence bands are separated by a narrow energy gap with some overlap between the two bands a small change in the concentration of holes or electrons can have a comparatively large effect on the electrical properties of the graphite. X-ray crystal structure studies on these intercalation or interstitial compounds indicate that the intercalated substance is held between the graphite planes in units that have symmetries somewhat similar to the structure of the graphite planes with appreciably

larger spacings. The residual compounds appear to be atoms held near imperfections in the graphite structure. Although most of the intercalation compounds decompose below the temperatures a nuclear fuel element is exposed to, some exhibit surprising stability. Residual compounds[16] can be stable at temperatures up to 2000°C in polycrystalline graphite.

Formation of carbides by reactions between fission products and graphite becomes increasingly important at high temperatures. Since the available thermodynamic data for metal carbides are somewhat incomplete the heats and entropies of formations were estimated for the carbides of several rare earths and noble metals appearing as fission products. The values for the heats of formation were estimated from an empirical relationship between the thermodynamic properties of homologous series and the entropies of formation were obtained by Latimer's method[25] from the partial entropies of the elements. Since heat capacity data are mostly unavailable the relationship

$$\Delta F_{fT} = \Delta H_{f\ 298°K} - T\ \Delta S_{f\ 298°K} \qquad (21)$$

was used to calculate the free energies of the carbide formation reaction from the elements. Assumption of no change in the heat capacities should give reliable free

energy values for the systems considered.

The free energy values given in Table III indicate that formation of hard metal and rare earth carbides is most likely but the alkali earth and even the noble metals may form carbides at sufficiently high temperatures. A comparison with fission product diffusion data indicates a similar trend in the quantities of these elements retained in graphite matrices. The alkali earth elements are not retained in the graphite matrix but the carbides of these elements have melting points below 2000°C and their mobility in the matrix should be considerably higher than those of the refractory carbides.

Table III. Free Energies, Heats of Formation and Standard Entropies of the Carbides of Fission Products

Compound	$-\Delta H_{f\ 298^oK}$ kcal/mole	$\Delta S_{f\ 298^oK}$ cal/deg mole	$-\Delta F_{fT}$, kcal/mole at ^oK 298	1073	1473	1873
SrC_2 [a]	10	3.7	11.1	14	15.5	16.9
BaC_2 [b]	5	3.4	6	8.6	10	11.4
YC_2 [a]	23	7.5	25.2	31	34	37
LaC_2 [a]	23	3.4	24	26.6	28	29.4
CeC_2 [a]	24	3.4	25	27.6	29	30.4
PrC_2 [a]	26	3.4	27	29.6	31	32.4
ZrC [b]	45	8.5	47.5	54.1	57.5	60.9
Mo_2C [b]	- 4.2	2.4	11.3	30.0	39.6	49.2
Ru_2C [c]	-17	7.3	-14.9	- 9.2	- 6.3	- 3.3
Rh_3C [c]	-50	17.7	-44.8	-31	-23.9	-16.9
Pd_3C [c]	-54	17.9	-48.7	-34.8	-27.6	-20.5

a. Estimated from lattice parameters

b. High Temperature Technology, ed. by I. E. Campbell, John Wiley and Sons, Inc., New York, 1956.

c. Estimated by homologous series method. (O. Kubaschewski and E. L. Evans, Metallurgical Thermochemistry. John Wiley and Sons, Inc., New York, 1956.)

REFERENCES

1. The ORNL Gas-Cooled Reactor Advanced Concepts, ORNL-2510 (Sept. 18, 1958).
2. High Temperature Gas-Cooled Graphite-Moderated Power Station. General Atomics, GA-593, GACP-595, GA-598, (Nov. 1958).
3. Sanderson and Porter, Design and Feasibility Study of a Pebble Bed Reactor Steam Power Plant, S & P 1963A, Vol 1 (May 1958).
4. S. Glasstone and M. C. Edlund, The Elements of Nuclear Reactor Theory (67, Van Nostrand Company, Inc., New York, 1952).
5. T. A. Lane et al., A Study of Problems Associated with Release of Fission Products from Ceramic Fuels in Gas-Cooled Reactors, ORNL-2851 (Oct. 27, 1959).
6. T. O. Blomeke and M. F. Todd, Uranium-235 Fission-Product Production as a Function of Thermal Neutron Flux, Irradiation Time, and Decay Time, Part 1, Vol. 1, ORNL-2127 (Nov. 12, 1958).
7. W. Jost, Diffusion in Solids, Liquids, Gases, Academic Press Inc., New York, 1960.
8. T. Crank, The Matematics of Diffusion, Oxford University Press, London, 1957.
9. D. Cubicciotti, The Diffusion of Xenon from Uranium Carbide Impregnated Graphite at High Temperatures, NAA-SR-194 (Oct. 13, 1952).
10. L. B. Doyle, High Temperature Diffusion of Individual Fission Elements from Uranium Carbide Impregnated Graphite, NAA-SR-255 (Sept. 11, 1953).
11. C. A. Smith and C. T. Young, Diffusion of Fission Fragments from Uranium Impregnated Graphite, NAA-SR-72, (May 4, 1951).
12. C. T. Young and C. A. Smith, Preliminary Experiments on Fission Product Diffusion from Uranium Impregnated Graphite in the Range 1800° - 2200°C, NAA-SR-232 (June 17, 1953).

13. C. T. Young, High Temperature Diffusion of Individual Fission Products from Small Uranium Impregnated Graphite Samples under Deuteron Bombardment, NAA-SR-247 (Sept. 1, 1953).

14. G. A. Cowan and C. T. Orth, Diffusion of Fission Products at High Temperatures from Refractory Matrices. Second United Nations International Conference on the Peaceful Uses of Atomic Energy. A/Conf.15/P/613 (June 1958).

15. N. R. Large and G. N. Walton, Migration of Non-Gaseous Fission Products through Graphite, AERE C/M 346 (1958).

16. G. R. Hennig, "Interstitial Compounds of Graphite," p 125 in Progress in Inorganic Chemistry, Vol. 1. Interscience Publishers, Inc., New York, 1959.

17. G. Harbottle and N. Sutin, J. Phys. Chem. 62, 1344 (1958).

18. C. Zener, "Theory of Diffusion," p 289 in Imperfections in Nearly Perfect Crystals, ed. by W. Shockley et al., John Wiley and Sons, Inc., New York, 1952.

19. S. Glasstone, K. T. Laidler and Henry Eyring, The Theory of Rate Processes, McGraw-Hill Book Co., Inc., New York, 1941.

20. T. C. Waddington, "Lattice Energies and Their Significance in Inorganic Chemistry," in Advances in Inorganic and Radiochemistry, Vol. I, Academic Press Inc., New York, 1959.

21. G. T. Dienes, Jour. of Applied Phys., 23, 1194 (1952).

22. R. P. Johnson, Physical Review, 56, 814 (1939).

23. R. C. Croft, J. Appl. Chem. 2, 557 (1952).

24. W. Rudorff, "Graphite Intercalation Compounds," p 224 in Advances in Inorganic and Radiochemistry, Vol. I. Academic Press, Inc., New York, 1959.

25. O. Kubaschewski and E. L. Evans, Metallurgical Thermochemistry. John Wiley and Sons, Inc., New York, 1956.

ORNL-3145
UC-80 - Reactor Technology
TID-4500 (16th ed.)

INTERNAL DISTRIBUTION

1. Biology Library
2. Reactor Division Library
3. ORNL - Y-12 Technical Library, Document Reference Section
4-5. Central Research Library
6-26. Laboratory Records Department
27. Laboratory Records, ORNL R.C.
28. C. E. Center
29. A. M. Weinberg
30. J. P. Murray (K-25)
31. R. G. Jordan (Y-12)
32. J. A. Swartout
33. G. E. Boyd
34. W. R. Grimes
35. R. B. Briggs
36. D. S. Billington
37. F. F. Blankenship
38. A. L. Boch
39. J. H. Frye, Jr.
40. H. G. MacPherson
41. J. C. White
42. W. D. Manly
43. C. H. Secoy
44. E. G. Bohlmann
45. R. A. Charpie
46. F. L. Culler
47. W. H. Jordan
48. M. T. Kelley
49. J. A. Lane
50. H. F. McDuffie
51. M. J. Skinner
52. A. H. Snell
53. E. H. Taylor
54. G. M. Watson
55. C. E. Winters
56. G. C. Warlick
57. E. L. Compere
58. J. C. Griess
59. G. H. Jenks
60. R. B. Korsmeyer
61. H. C. Savage
62. M. Blander
63. S. Cantor
64. J. H. Shaffer
65. M. J. Kelly
66. C. J. Barton
67. W. L. Marshall
68. C. F. Baes
69. R. E. Thoma
70. R. A. Strehlow
71. R. B. Evans, III
72. G. W. Keilholtz
73. W. E. Browning
74. W. T. Rainey
75. L. G. Overholser
76-78. A. R. Saunders
79-104. Lois Curry
105. F. Daniels (consultant)
106. F. T. Miles (consultant)
107. F. T. Gucker (consultant)
108. Leo Brewer (consultant)
109. J. E. Ricci (consultant)
110. D. G. Hill (consultant)
111. G. W. Morey (consultant)
112. H. Eyring (consultant)
113. M. G. Fontana (consultant)
114. R. M. Fuoss (consultant)
115. N. Hackerman (consultant)
116. H. S. Harned (consultant)
117. H. Insley (consultant)
118. E. A. Mason (consultant)
119. T. N. McVay (consultant)
120. G. Scatchard (consultant)
121. T. F. Young (consultant)
122. J. S. Dahler (consultant)
123. J. W. Cobble (consultant)
124. P. H. Emmett (consultant)

EXTERNAL DISTRIBUTION

125. Division of Research and Development, AEC, Washington
126. Division of Research and Development, AEC, ORO
127. Division of Reactor Development, AEC, Washington
128. Division of Reactor Development, AEC, ORO
129-732. Given distribution as shown in TID-4500 (16th ed.) under Reactor Technology category (75 copies - OTS)