DIFFUSION OF FISSION PRODUCTS AT HIGH TEMPERATURES
FROM REFRACTORY MATRICES

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I. INTRODUCTION

Current interest in high temperature reactors has emphasized the need for measurement of fission product diffusion rates from refractory materials in the temperature region of 1500°C to 3000°C. In addition, detailed knowledge concerning diffusion rates of fission products at high temperatures permits the study of many short-lived early members of fission product chains which might otherwise be difficult or impossible to separate rapidly enough by chemical means.

A problem of particular importance concerns the possible loss of delayed neutron reactivity in advanced design reactors which are meant to operate at temperatures greater than 2000°C. Other important problems concern the rate of loss of fissile material, the gross rate of loss of fission products, and the rates at which individual fission products are lost at high temperatures.

II. SOME PREVIOUS INVESTIGATIONS

Early measurements of the diffusion of a few fission products were carried out at the Argonne National Laboratory.^{1,2,3,4,5} The Argonne investigators observed that the loss of xenon from 1 cm cubes of BeO-UO₂ varied from 0.1% per three hours of heating in flowing helium gas at 1000°C to a few percent in three hours at 1450°C. The total loss of gross fission product activity from these cubes was less than 0.1% in fourteen hours of heating at 1400°C.

Investigators at North American Aviation Laboratories studied the diffusion of fission products from graphite.^{6,7,8,9,10} When discs of AUF graphite, 0.84 cm in diameter and 0.064 cm to 0.52 cm thick, impregnated with

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uranium carbide to a concentration of about 520 mgs. per cubic centimeter, were heated to temperatures ranging from 900°C to 1500°C, loss rates of xenon varied from approximately 2% per three hours of heating in argon gas at 900°C to approximately 50% per three hours at 1500°C. The loss rates were independent of sample size. It was postulated that diffusion from graphite was largely through the interior of grains to the surface followed by a rapid escape to the argon stream. For the infinite cylindrical solution of Fick's equation and a graphite grain size of the order of 0.07 mm., the diffusion constant can be represented by the equation

$$D = D_0 e^{-E/RT}$$

where D_0 and E are 3×10^{-5} cm²/Sec and 49,000 calories/mole-degree, respectively. The slow rate of diffusion for xenon was thought to be due to a "steric hindrance" effect since the interlaminar distance of carbon atoms in graphite is 3.4 Å and the effective diameter of xenon is 3.8 Å.

The diffusion rates in AUF graphite discs for several major fission elements were measured in a vacuum at 1500°C to 1900°C. The discs were 0.625" in diameter and 0.1" thick. Barium and praseodymium appeared to have diffusion rates dependent on graphite specimen size but iodine, tellurium, and perhaps strontium, diffused at rates independent of specimen size. Approximate initial loss rates are summarized in Table I.

TABLE I
Initial Loss Rate of Fission Products
from Graphite*

	1500°C	1900°C
Iodine	0.6%/min	8%/min
Tellurium	1%/min	8%/min
Barium	0.2%/min	5%/min
Strontium	2%/min	> 10%/min
Cesium	> 10%/min	---
Praseodymium	---	1.5%/min
Cerium	---	0.5%/min
Ruthenium	---	0.13%/min

* North American Aviation results.

III. MATHEMATICAL CONSIDERATIONS

The principal refractory matrix used in these experiments was graphite. In addition, diffusion measurements were made with a magnesium oxide matrix.

Although the actual process of diffusion from these materials is a complex one, it has been observed that the fraction remaining of each fission product studied at a given temperature and in a given geometry can be approximately represented by an equation of the form

$$F_i = c_i e^{-D_i(1)t} + (1 - c_i) e^{-D_i(2)t} \quad (1)$$

The diffusion rates observed experimentally appear to be independent of sample size and are so represented here.

The representation of the diffusion process by a sum of logarithmic decay terms simplifies further mathematical treatment and is not inconsistent with equations derived by a more rigorous analysis of the diffusion problem.

This problem is somewhat analogous to one considered by Flügge and Zimens¹¹ concerning the rate of escape of gaseous emanation from radioactive material. In their model, escape occurs in two ways, by escape from interstitial positions, due to recoil of product atoms out of the grain surface, and by diffusion from within the grain to the surface.

In the case of fission fragments, every fragment has enough energy to escape from the interior of a grain. It is then trapped in another grain or is trapped between grains. Diffusion then occurs both from pores and, at a different rate, from the interior of grains. The rate of escape of volatilized material from pores should approximate a simple exponential decay law. However, diffusion from the grains themselves obeys a more complicated law.

The cylindrical solution of Fick's diffusion equation may be used to illustrate the theoretically derived time dependence of diffusion from grains.

For an infinite cylinder of radius r_0 , the concentration after a time t is given by

$$C/C_0 = 4 \left[\sum_{v=1}^{\infty} \frac{1}{\xi_v^2} e^{-\left[\xi_v^2 \frac{Dt}{r_0^2} \right]} \right] \quad (2)$$

For a cylinder with a length twice the diameter

$$C/C_0 = 8 \left[\sum_{n=0}^{\infty} \left\{ \frac{2}{\pi(2n+1)} \right\}^2 e^{-\left\{ \frac{\pi(2n+1)}{2} \right\}^2 \cdot \frac{x}{4}} \right] \left[\sum_{v=1}^{\infty} \frac{1}{\xi_v^2} e^{-\xi_v^2 x} \right] \quad (3)$$

where $x = Dt/r_0^2$ and ξ_v are the roots of the Bessel function $J_0(x) = 0$.

To a very good approximation, these functions can also be represented by equations of the form of equation (1). Therefore, the empirical representation of diffusion by equation (1) may be understood as having some basis in the facts that there are at least two important rate-controlling processes, that the simplest of these processes, diffusion from cracks or pores, can be **roughly approximated** by an exponential decay law, and that the more complicated process, diffusion from grains, can be empirically described as the sum of two simple exponential terms as though it consisted of a fast and slow process. It should be emphasized that the diffusion constants in equation (1) are not "true" diffusion constants but are chosen

to provide a best fit to the observed rates of diffusion.

In all further derivations of equations for the loss of fission products by radioactive decay and diffusion, the effective decay constant is taken as the sum of the radioactive decay constant and the diffusion constant.

IV. EXPERIMENTAL TECHNIQUES

A furnace was designed for obtaining temperatures in excess of 2600°C . The furnace is shown schematically in Fig. 1. The refractory slugs are inserted into a hollow graphite resistance heating element and heated indirectly.

These slugs were cylindrical rods, 0.6 cm in diameter and 2.5 cm long. They were loaded with 150 mg of U^{235} per cm^3 . This loading produced graphite containing 7% by weight of U^{235} . During graphitization the UO_2 was converted to uranium carbide. Changes in slug size demonstrated no significant size dependence for diffusion rates.

Temperature readings were made through a viewing port by a Leeds and Northrup optical pyrometer. No black body corrections were made. Unless otherwise stated, the furnace atmosphere was flowing helium gas.

The slug could be transferred pneumatically out of the furnace through the hollow copper electrode by helium gas entering through the hollow aluminum electrode. When short-lived delayed neutron emitters were counted, the slug was fired into a neutron counter as illustrated schematically in Fig. 2. In these experiments eight 13" G.E. B^{10} -coated neutron detectors were connected in parallel and the output fed to an amplifier. The amplifier output was fed to a fast scaler which recorded total counts and to a time-delay analyzer¹² which recorded counts in 0.1, 1, and 10 sec. intervals for periods up to 5 minutes. The decay data from the time delay analyzer were processed on an IBM-704 for principal decay periods.

When it was desired to study sweep gases, modifications were made in the heating element so that it held the slug in place and also in the solenoid valve arrangement so that the gas line could be initially evacuated. Evacuation of the gas line minimized gas transit time to the neutron counter.

After neutron bombardment and heating, slugs were destroyed in a mixture of perchloric and fuming nitric acid and radiochemical analyses were performed according to standard procedures.

V. EXPERIMENTAL DATA

A. Loss of Uranium

In studies made to ascertain the loss of uranium from refractory materials under experimental conditions, it was observed that loss was limited to erosion loss in the matrix material. At the highest temperature readily available in graphite, 2600°C , 0.6% loss in weight after 10 minutes of heating in He gas was observed in a graphite element. A similar loss occurred in the total uranium content of the graphite. Within experimental error, the concentration of uranium in the graphite remained the same.

FIG.1 ELECTRIC FURNACE

FIG.2 EXPERIMENTAL EQUIPMENT AT REACTOR

When the experiment was repeated at the same temperature in an ammonia atmosphere for five minutes, a 15% loss in total weight of the matrix was observed with no measurable loss of uranium.

B. Loss of Fission Products

1. Graphite Matrix

a. Mo^{99} .

Since loss estimates in all individual fission product studies were based on measurement of total fissions, it was necessary to find a reliable monitor for measurement of total fissions. Early measurements were made in which graphite slugs were bombarded both cold and at 2600°C , and the neutron flux was measured with foils attached to the outside of the furnace. Measurements were then made of the Mo^{99} activity in the slugs, normalized to the uranium content and the neutron exposure. Within an experimental error of 1%, the Mo^{99} activity per fission per gram of uranium was constant up to 2700°C , for 5 minute heating and simultaneous bombardment intervals. Therefore, it was concluded that no loss of Mo^{99} activity or its mass 99 predecessors occurred under the given experimental conditions.

The work was repeated on slugs which were first bombarded cold and heated only after sufficient time had elapsed so that essentially all of the chain of mass 99 had decayed to Mo^{99} . No loss of Mo^{99} activity was observed. In succeeding work on the diffusion of other fission products, the amount of Mo^{99} was taken as a measure of the absolute number of fissions in the sample.

b. Loss of As, Br, Sr, Zr, Ru, Rh, Pd, Ag, Cd, Sn, Sb, Te, I, Cs, Ba, Ce, Nd, and Eu.

Diffusion rates for various fission products were measured in the following way. A group of five to eight slugs were irradiated together in a thermal neutron flux. Sufficient decay time was then allowed to permit parent isotopes to decay quantitatively to the isotope of interest. These slugs were then heated either for variable times at a given temperature (usually 30, 60, 120, and 240 seconds) or for 30 seconds at varying temperatures.

The data obtained from these measurements are presented in Tables II and III and are plotted in Figs. 3 and 4. The values for apparent diffusion constants and for c_i 's as defined in equation (1) are presented for some of these fission products in Table IV. Some activation energies for the diffusion constants which apply to the rapid part of the diffusion process, calculated from the usual temperature dependence,

$$D = D_0 e^{-\frac{E}{RT}}$$

are presented in Table V.

TABLE II

Percent Retention of Fission Products in Graphite
at 2400°C as a Function of Time (Los Alamos)

Element	30 sec.	60 sec.	120 sec.	240 sec.
As ⁷⁷	80	44	48	---
Br ⁸³	13	2.2	1.0	---
Sr ⁸⁹	2.8	0.18	0.03	---
Ru ¹⁰³	---	---	---	70
Rh ¹⁰⁵	---	---	---	92
Pd ^{109, 112}	42	31	34	23
Ag ¹¹¹	<< 0.1	---	---	---
Cd ¹¹⁵	<< 0.1	---	---	---
Sb ¹²⁷	20	---	11	6.2
Sn ^{125, 121}	---	---	---	0.1
Te ¹³²	16	5.2	4.8	3.4
I ^{131, 133}	5	3.3	3.0	1.4
Cs ¹³⁷	---	---	---	0.26
Ba ¹⁴⁰	<< 0.1	---	---	---
Ce ¹⁴³	---	---	---	27
Nd ¹⁴⁷	---	---	---	19
Eu ^{156, 157}	---	---	---	74

TABLE III

Percent Fission Product Activity Remaining in Graphite
as a Function of Temperature for 30 sec. Heating
(Los Alamos)

Element	1600°C	1800°C	2000°C	2200°C	2400°C	2600°C
As ⁷⁷	---	97	86	71	66	31.4
Br ⁸³	---	66.3	56.6	13.9	8.5	5.4
Pd ^{109, 112}	85.4	84	76	71	---	64
Cd ¹¹⁵	47	31	6.1	0.35	---	0.044
Sb ¹²⁷	---	98	83	58	---	17
Te ¹³²	---	98	73	65	---	35
I ^{131, 133}	93	91	78	55	6	3.8
Ba ¹⁴⁰	---	--	98	91	---	50

TABLE IV

Some Fission Product Diffusion Rates at 2400°C

Isotope	$D_1(\text{sec}^{-1})$	$D_2(\text{sec}^{-1})$	c_1	$(1-c_1)$
As ⁷⁷	0.026	0.0014	0.50	0.50
Br ⁸³	0.076	0.005	0.985	0.015
Sr ⁸⁹	0.119	---	1.0	---
Zr ⁹⁷	0	0	---	---
Ru ¹⁰³	---	0.0012	---	1.0
Rh ¹⁰⁵	---	0.00034	---	1.0
Pd ^{109, 112}	0.074	0.0019	0.62	0.38
Ag ¹¹¹	> 0.2	---	1.0	---
Cd ¹¹⁵	~ 0.12	---	~ 1.0	---
Sb ¹²⁷	0.107	0.0045	0.81	0.19
Te ¹³²	0.075	0.0023	0.94	0.06
I ^{131, 133}	0.139	0.0048	0.96	0.04
Ce ¹⁴³	---	~ 0.005	---	1.0
Nd ¹⁴⁷	---	~ 0.008	---	1.0
Eu ^{156, 157}	---	~ 0.001	---	1.0

TABLE V

Some Apparent Activation Energies for
the "Fast" Diffusion Process

Isotope	Apparent E_{act} (kcal.)
As ⁷⁷	32
Br ⁸³	27
Pd ^{109, 112}	9
Cd ¹¹⁵	30
Sb ¹²⁷	50
Te ¹³²	18
I ^{131, 133}	42

c. Loss of Gamma Activity

Two weighed graphite slugs of known uranium content were irradiated for five minutes in a flowing helium atmosphere, one at room temperature and the other at 2600°C. They were then measured with a vibrating reed electrometer which measures gross gamma activity. The ratio of the hot slug gamma activity to the cold slug activity is presented as a function of time after bombardment in Table VI. The average gamma energy in both slugs is ~ 1.3 Mev up to six hours after bombardment.

These data indicate that 70-80% of the gamma-emitting fission products are lost in the course of a five minute bombardment at 2600°C.

TABLE VI

Retention of Gamma Activity in Graphite after
Five Minute Heating at 2600°C

Time after bombardment (mins.)	35	75	120	140	180	240	300	360
Fraction remaining	0.305	0.267	0.234	0.211	0.224	0.196	0.197	0.182

d. Loss of Delayed Neutron Emitters

The ratio of delayed neutron activity in heated and unheated slugs was measured as a function of heating and bombardment time and as a function of temperature.

Start of neutron exposure was controlled by raising the cadmium curtain in the south thermal neutron column of the Water Boiler reactor. The rise time of the neutron flux was approximately 500 milliseconds. The bombardment was terminated by firing the slug into the neutron counter well where it was caught in a bottle of water. Transit time of the slug to the counter was approximately 200 milliseconds.

Data obtained from these measurements are presented in Tables VII and VIII and are plotted in Figs. 5 and 6.

TABLE VII

Delayed Neutrons Remaining after Simultaneous Heating
and Bombardment of U loaded Graphite at 2400°C

Bombardment time (secs.)	5.5	9.5	18.5	28.5	58.5	150	300
Fraction remaining	0.84	0.765	0.675	0.62	0.60	0.60	0.60

TABLE VIII

Delayed Neutron Retention in Graphite and Magnesium Oxide
after Heating (Bombardment time: 240 seconds)

Matrix Material	Temperature					
	1600°C	1800°C	2000°C	2100°C	2250°C	2400°C
Graphite	0.921	0.919	0.810	0.790	0.635	0.60
MgO	0.900	0.900	0.760	---	---	---

Fig. 3 - Some typical diffusion losses at 2400°C in graphite.

Fig. 4 - Temperature dependence of diffusion losses in graphite.

Fig. 5 - Calculated and observed loss of delayed neutron activity from graphite (2400°C).

Because the most probable delayed neutron precursors are bromine and iodine isotopes, a predicted curve for the diffusion loss of delayed neutron precursors has been constructed on the following basis:

The delayed neutron periods and their abundances, as reported by Keepin, et. al.,¹³ are:

1. 54.5 sec., 3.8% abundant	4. 2.2 sec., 40.7% abundant
2. 21.8 sec., 21.3% abundant	5. 0.50 sec., 12.8% abundant
3. 6.0 sec., 18.8% abundant	6. 0.18 sec., 2.6% abundant

The first period is assumed to be 100% bromine. Various assumptions were made for the other periods, ranging from 50% to 100% iodine and the remainder bromine in period 2, from 33% to 100% bromine and the remainder iodine in period 3, from 55% to 100% iodine in period 4, and from 100% iodine to 100% bromine in periods 5 and 6. For most combinations of these assumptions, the measured loss rate fell within 1% of the averaged calculated curve obtained in the following way:

For the first period the ratio, r_1 , of the delayed neutron intensity in the hot slug to the delayed neutron intensity in the cold slug is

$$r_1 = \frac{c_1 \lambda_1}{\Lambda_{11}} \left(\frac{1 - e^{-\Lambda_{11}t}}{1 - e^{-\lambda_1 t}} \right) + \frac{\lambda_1}{\Lambda_{12}} (1 - c_1) \left(\frac{1 - e^{-\Lambda_{12}t}}{1 - e^{-\lambda_1 t}} \right) \quad (4)$$

At 2400°C:

$$\begin{aligned} \lambda_1 &= 0.013/\text{sec.} \\ \Lambda_{11} &= 0.076 + 0.013 = 0.089/\text{sec.} \\ \Lambda_{12} &= 0.005 + 0.013 = 0.018/\text{sec.} \\ c_1 &= 0.985 \\ 1 - c_1 &= 0.015 \end{aligned}$$

From these values, r_1 is calculated at various times and plotted. Similar curves are drawn for the other periods. A sum of these curves, each weighted by its relative abundance, is plotted in Fig. 5.

The calculated curve coincides with the observed data. However, satisfactory agreement can be obtained by substituting considerable proportions of Sb and Te precursors for the halogens. The substitution of As or other hypothetical precursors with lower diffusion rates leads to poor agreement with observation.

Similarly, the measured temperature dependences of bromine and iodine diffusion rates were used to calculate a temperature dependence curve for loss of the delayed neutron activity. The calculated and measured saturation retention of gross delayed neutron activity for 240 second bombardment time are plotted in Fig. 6 as a function of temperature. The agreement is consistent with the hypothesis that most of the delayed neutron emitter precursors are bromine and iodine but does not exclude the possibility of significant admixtures of other elements.

e. Analysis of Volatilized Delayed Neutron Precursors

Measurements were made on gases evolved from graphite elements that were simultaneously heated to 2400°C and bombarded for ~ 2 seconds. The evolved gas was swept through an evacuated line containing an activated

charcoal trap loaded with silver nitrate. The trap was heated to $\sim 80^{\circ}\text{C}$. The gas finally swept into an evacuated collector bottle. Transit time to the bottle was ~ 500 milliseconds.

The ratio of β - γ activity, charcoal trap to collector bottle, was two to one. The delayed neutron activity, however, was quantitatively in the charcoal trap with no detectable activity in the collector bottle. The trap counted as high as 4100 neutrons in 15 seconds. The collector bottle counted zero above a background of 30 per minute.

The conclusion drawn from these measurements was that no appreciable fraction of the delayed neutron activity was due to rare gas precursors or to daughter elements of rare gas precursors. The data are presented in Table IX.

TABLE IX
Results of Gas Analysis from Graphite Elements
(Bombardment time ~ 2 seconds before sweeping.)

~ Temp.	Maximum β - γ Level		Neutron counts in 15 seconds	
	Trap	Collector bottle	Trap	Collector bottle
2000 $^{\circ}\text{C}$	> 20 mr at 5 inches	~ 10 mr at 5 inches	~ 2100	non-detectable
2100 $^{\circ}\text{C}$	> 30 mr at 5 inches	~ 15 mr at 5 inches	~ 3100	non-detectable
2300 $^{\circ}\text{C}$	> 40 mr at 5 inches	~ 20 mr at 5 inches	~ 4100	non-detectable

2. Magnesium Oxide Matrix

a. Loss of As, Sr, Zr, Mo, Pd, Ag, Cd, Sb, Te, I, and Ba

The fractional retention of eleven fission products at 1950°C is plotted as a function of time in Fig. 7. It is noteworthy that molybdenum and arsenic are somewhat more volatile in MgO than in graphite. Palladium, tellurium, iodine, and barium behave very much as they do in graphite. Cadmium, silver, tin, and probably strontium, are very much less volatile in MgO than they are in graphite.

b. Loss of Delayed Neutron Emitters

The retention of gross delayed neutron activity in MgO up to 1000°C is plotted in Fig. 6. Slightly larger losses of the delayed neutron activity from MgO are consistent with the slightly larger diffusion constant measured for iodine in magnesium oxide. By inference, the diffusion constant for bromine is also somewhat larger in magnesium oxide although it was not measured.

c. Identification of Short-Lived Fission Products by Volatilization Techniques

A simple experiment may be devised for determination of the half-life of Pd^{115} on the assumption that the half-life of Rh^{115} is a very few seconds or less, a reasonable assumption since Rh^{115} is twelve neutrons

Fig. 6 - Percent gross delayed neutron activity retained after saturation (240 sec.). Bombardment and heating.

Fig. 7 - Some typical diffusion rates at 1950°C in MgO.

Fig. 8 - Measurement of half-life of Pd^{115} .

removed from stable Rh^{103} . If a series of graphite slugs are bombarded for sixty seconds and then, variously, heated to 2500°C immediately after bombardment for twenty seconds or allowed to decay in the cold for different lengths of time and then heated for twenty seconds, it can be calculated from the known diffusion constants for Rh, Pd, Ag, and Cd that during heating $\sim 2\%$ of the Rh is lost, $\sim 20\%$ of the Pd is lost, $\sim 99.999\%$ of the Ag is lost, and $\sim 99.9\%$ of the Cd is lost. If, during bombardment, essentially all of the Rh has decayed to Pd, then the loss of the 115 chain up to and including Pd^{115} is a constant fraction of the Pd^{115} present, which is in turn related by the simple exponential decay law for Pd^{115} to the length of time between bombardment and heating. All Pd^{115} which decays to Ag^{115} is quantitatively volatilized as is the Cd^{115} growing in from Ag^{115} . The analogy to a chemical "milking" experiment is very close.

This experiment was performed and the amount of mass 115 determined by analysis for Cd^{115} . The results of two runs are plotted in Fig. 8. The half-life obtained for Pd^{115} is 47 ± 3 seconds. The half-life of this isotope has also been reported by Alexander, Schindewolf, and Coryell¹⁴ as 45 seconds.

This technique or modifications of it is applicable to a large number of problems concerning fission yields where chemical separations require too much time for investigation of very short-lived early members of chains. It is being further developed at Los Alamos and promises to be useful.

VI. DISCUSSION OF RESULTS

The data presented here indicate that fission products diffuse very rapidly from refractory fuel element matrices in the temperature region above 1500°C . The most noticeable consequence of rapid diffusion is that a large fraction of the excess reactivity provided by delayed neutrons is lost to the reactor at temperatures in excess of 2000°C .

The delayed neutron precursor elements diffuse out of graphite at rates which are consistent with the measured rates for bromine and iodine. Gas trapping experiments exclude significant contributions from rare gas precursors and daughter products of rare gas precursors. Therefore, the present data are consistent with the hypothesis that most, perhaps 80% or more, of the delayed neutron activity arises from bromine and iodine precursors.

Although diffusion from the refractory matrices studied here is a complex process, the rate can be treated as a sum of a fast and slow simple exponential decay of each fission product. The technique of high temperature volatilization can be used to identify some very short-lived fission products which might require too long a processing time to identify chemically.

There are no simple correlations of diffusion rates of the fission products with any single property, such as the melting point or boiling point of the metal or carbide or diameter of the atom or probable ion. However, special explanations may be offered for most observations. Where very stable carbides are known to be formed, the fission products (Zr and Mo) do not diffuse from graphite. Some fission products which may very well be present as metal atoms in graphite and whose metals have rather low melting and boiling points (e.g., Cd and Sn) diffuse rapidly. The very rapid diffusion of silver from graphite is probably related to the low melting point of the metal plus its small

atomic radius. In the MgO matrix, fission products with volatile oxides diffused more rapidly than they did in graphite (Mo, As). Fission products with refractory oxides generally diffused less rapidly than in graphite.

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