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Diffusion of Uranium in H-451 Graphite at 900 to 1400°C

O. K. Tallent
R. P. Wichner
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ABSTRACT

In this study, the diffusion of uranium (as a stand-in for plutonium) was investigated under conditions approximating those of the primary coolant loop in a High Temperature Gas-Cooled Reactor (HTGR). Profiles were obtained for uranium penetration in H-451 graphite (from the Great Lakes Carbon Company) at temperatures ranging from 900 to 1400°C. Profile data for given temperatures were considered in terms of the following expression:

$$\left[\operatorname{erf}^{-1}\left(1 - \frac{C}{C_0}\right)\right]^2 = \frac{x^2}{4Dt} ,$$

where C is the concentration of uranium at time, t, for a distance, x, into the pellet; C_0 is a constant representing the uranium concentration at $x = 0$ for all t, and D is the diffusion coefficient.

Diffusion coefficients for uranium dicarbide at 1000 and 1400°C were found to be defined by:

$$D_{UC_2} = 3.5 \times 10^{-3}\ e^{-4.8 \times 10^4/RT}\ \mathrm{cm^2/s} .$$

For uranium dioxide at 900, 1000, and 1400°C, diffusion coefficients are defined by:

$$D_{UO_2} = 2.34 \times 10^{-6}\ e^{-3.2 \times 10^4/RT}\ \mathrm{cm^2/s} ,$$

where R is the gas constant, and T is the temperature in degrees Kelvin.

1. INTRODUCTION

Knowledge of the diffusion of plutonium in graphite is required for realistic estimation of plutonium contamination in the primary cooling system of High Temperature Gas-Cooled Reactors (HTGRs). Estimates of plutonium contamination levels in the primary cooling system of HTGRs are presently based on the conservative assumption of zero hold-up in the core graphite, which leads to unrealistically high estimates. One way to deal with this problem is to improve fuel fabrication techniques in order to reduce uranium contamination levels (thus also reduce transmuted plutonium contamination levels) outside the fuel particle. However, this added expense may be unnecessary. A more realistic method for calculating plutonium contamination of the primary coolant loop might demonstrate that such added fuel fabrication expense is not needed.

To determine the rate or extent of transport of plutonium to the coolant gas, it is necessary to know, in addition to the plutonium diffusion rate, the vapor pressure of plutonium which has diffused or otherwise been transported to a surface passage exposed to the gas. Our ultimate objective is to study the diffusion and vapor pressure of the adsorbed plutonium. The use of uranium as a stand-in for plutonium in this initial part of the study has allowed development work to be conducted and equipment to be tested without the constraints of alpha glovebox containment. Furthermore, the results of the uranium study are also of interest because rates of uranium and plutonium diffusion in the system are expected to be comparable and because uranium is an element in the transport pathway of the plutonium to the cooling system's passages.

Specific problems that were addressed include: the preparation of H-451 graphite (Great Lakes Carbon Company) surfaces with adsorbed or embedded uranium dioxide (UO_2); analyses to determine the amounts and distribution of the adsorbed uranium; and measurement of uranium diffusion rates at 900, 1000, and 1400°C in a helium atmosphere. The diffusion rate of uranium dicarbide (UC_2) was also investigated, since uranium oxides at sufficiently high temperatures (1000 to 1400°C) can react with graphite to form carbides under conditions where carbon monoxide, a reaction product, is swept out of the system.[1] In tests where UO_2 was the initial diffusing species, a partial conversion to the carbide likely occurred.

Diffusion experiments are frequently designed to provide either a constant source of the diffusing material or a source with instantaneous transfer of a finite amount of material and a concentration which decreases as the diffusion proceeds. The experiments for this study were designed to provide a constant source of uranium. The data obtained have been tested on mathematical models for both types of boundary conditions.

2. EXPERIMENTAL PROCEDURES

2.1 GENERAL APPROACH

A pellet transfer method was used in which uranium was diffused from a source pellet into a second, clean, initially uranium-free test pellet. Data were collected to determine the penetration profiles of the uranium into the test pellets as a function of time and temperature. The uranium concentrations in the source pellets are considered to have remained essentially constant, since these concentrations were large compared to the concentrations diffused to the test pellets. The transfer of uranium across the plane of contact between pellets to provide a constant uranium source on the test pellet outer surface is less certain. Data were collected, and distribution coefficients calculated, using both the instantaneous transfer (decreasing source) method and the constant-source method. It appears certain that the real distribution coefficient values fall within the range of values calculated using these two methods.

2.2 PREPARATION OF URANIUM SOURCE PELLETS

The H-451 graphite was in the form of 0.80-cm-diam and 1.25-cm-long pellets with machine-smoothed end surfaces. The graphite had a bulk density of 1.75 g/cm^3, a BET surface area of 0.63 m^2/g, and an open porosity of 17.2 vol %. The procedure used to adsorb or embed uranium (or UO_2) on the pellet surfaces involved soaking the pellets in ethanol containing 0.1 to 1.0 $\underline{M}$ $UO_2(NO_3)_2$ for 24 h at ~23°C; drying the pellets in air for

24 h at 130°C; and then calcining them under helium gas for 24 h at ~1000°C. The purpose of the calcination was to denitrate the adsorbed $UO_2(NO_3)_2$. The uranium adsorbed on the surface of the graphite pellets was widely and evenly distributed, as indicated by the typical x-ray map shown in Fig. 1 and the scanning electron micrographs in Fig. 2. Each dot on the x-ray map indicates the detection of a uranium atom. Similar maps (not shown) of pellets with ground surfaces indicated ~90% of the adsorbed uranium to be within 0.025 mm of the surface. The porous nature of the graphite can be seen in the scanning electron micrographs in Fig. 2, where the white color indicates adsorbed uranium. Uranium adsorbed at a depth of more than a few micrometers inside the pores, for example, is not detectable in the micrographs. The amounts of uranium adsorbed (as determined by delayed neutron activation analyses) are defined with a correlation coefficient of 0.99 by the adsorption equation:

$$U_{ads} = 9.87U_{sol} + 0.01 \ , \qquad (1)$$

where U_{ads} denotes amount of adsorbed uranium in mg per cm^2 of geometric surface area, and U_{sol} denotes the initial molar concentration of uranium in the ethanol solution. The total amounts of uranium in the prepared source pellets ranged from ~630 to 2390 μg. A uranium carbide specimen, rather than a source pellet, was used for the uranium carbide diffusion tests.

2.3 DIFFUSION TESTS

The source pellets, as previously indicated, were placed in contact with the test pellets for the diffusion tests. Both pellets were inserted into a tightly fitting cylindrical opening in a 2- by 2- by 1.25-cm graphite block, such that the plane of contact between the pellets was approximately in the center of the block, which was then placed in a 2.54-cm-diam alumina tube in a horizontal tube furnace and heated. Heating periods ranged from 2 to 21 d at 900, 1000, or 1400°C. The graphite block served to hold the pellets in contact, minimize their

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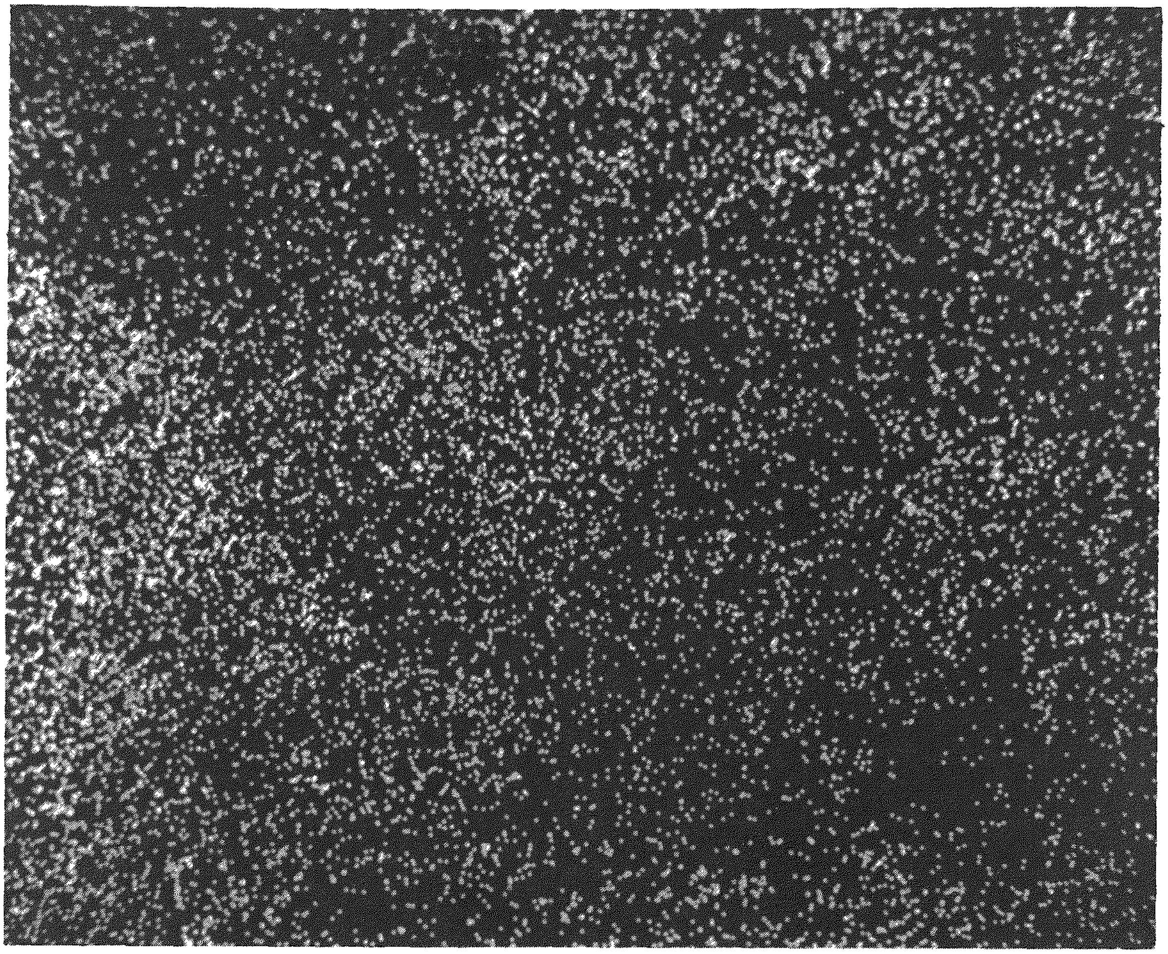

Fig. 1. X-ray map of uranium adsorbed on H-451 graphite (magnification: 2000X).

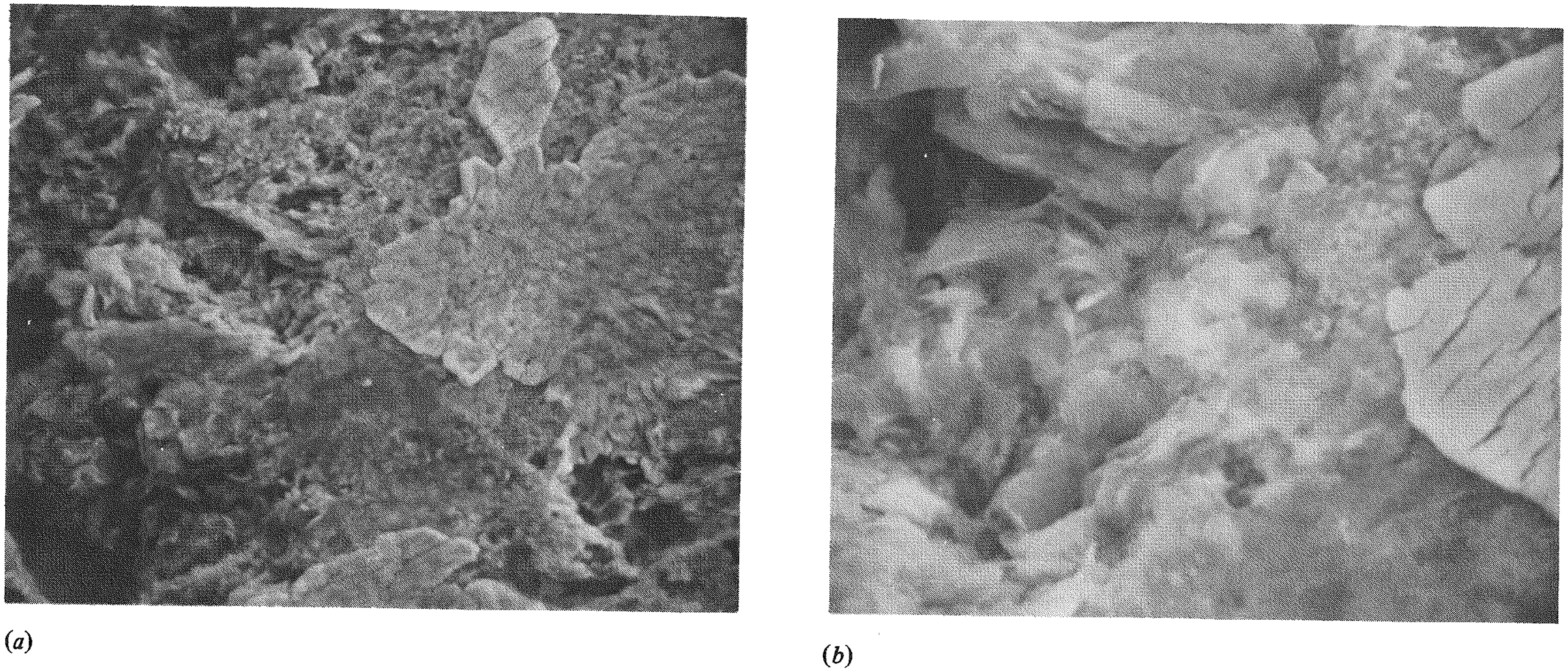

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Fig. 2. Scanning electron micrographs of uranium adsorbed on H-451 graphite; (a) magnification: 1000X; (b) magnification: 5000X.

corrosion, and minimize the escape of carbon monoxide gas from the pellet surfaces, thus helping to stabilize the uranium as UO_2. A 90 cm^3/m (STP) flow of helium gas (purified over titanium sponge at 400°C) was passed through the tube during the heating period. The pellets were then cooled to room temperature and analyzed by delayed neutron activation analysis for total uranium.

The method used to investigate diffusion of UC_2 was essentially the same as that used for UO_2, except that a uranium carbide specimen was substituted for the UO_2 source pellet. The specimen composition was 75—95% UC plus 2—10% UC_2; however, the experimental conditions[2] were such that the diffusing species was UC_2.

2.4 PELLET URANIUM ANALYSES

For purposes of analysis, the test pellets were irradiated in the Oak Ridge Research Reactor (ORR) for 5 min if they contained ~0.1 to 1 ppm uranium or in the High Flux Isotope Reactor (HFIR) for ~1 h if they contained <0.1 ppm uranium. A TP-5000 gamma-ray spectrometer was used to determine the amount of ^{239}Np in the irradiated pellets, after which multiple layers, each ~0.0025-cm thick, were ground from the pellet ends that had been in contact with a source pellet. The widths of the ground-off layers were determined to an accuracy of ±0.00025 cm using a Bausch and Lomb DR-25-B optical gauge. The amounts of ^{239}Np in the powder from each ground-off layer and in the remaining pellet were determined using the gamma-ray spectrometer. The concentration of uranium in each layer was calculated as the product of the total uranium in the pellet (as determined by delayed neutron activation analysis) and the ratio of the ^{239}Np concentration in the layer to the initial ^{239}Np concentration in the whole pellet.

3. EXPERIMENTAL RESULTS

3.1 TOTAL URANIUM TRANSFER

The total amounts of uranium (initially as UO_2) transferred from source to test pellets in 15 individual tests are listed in Table 1. The

Table 1. Total uranium transfer from source to test pellets during contact for various times at 900 to 1400°C

		Source pellet[a]			
Temperature (°C)	Time (d)	Total uranium[b] (µg)	Contact area between pellets[c] (µg)	Amount of uranium transferred[d] (µg)	Percentage transfer[e]
1400	7	753	91	2.060	0.23
1400	7	642	78	0.106	0.14
1400	7	607	74	0.642	0.87
1000	7	1235	150	0.133	0.09
1000	7	736	89	0.203	0.23
1000	11	1227	149	0.150	0.10
1000	11	1102	133	0.380	0.28
1000	14	1146	139	0.117	0.08
1000	14	753	91	0.140	0.15
1000	21	2150	260	0.170	0.06
1000	21	1450	176	0.180	0.10
1000	21	900	109	0.130	0.12
1000	21	851	103	0.120	0.12
900	7	641	78	0.103	0.13
900	7	627	76	0.133	0.17
900	21	N.D.[f]	N.D.	0.033	N.D.

[a] Each pellet weighed ~1.08 g.

[b] Uranium initially present as UO_2.

[c] The geometric contact areas were 0.505 cm^2, or approximately 12.5% of the total surface area of each pellet.

[d] This is the total amount of uranium transferred from the source pellet to the test pellet during an individual test.

[e] This is the percentage of available uranium that was transferred from the source pellet to the test pellet during an individual test.

[f] N.D. = not determined.

mass of uranium initially present on the source pellets ranged from 625 to 2150 μg (Table 1), while the mass transferred to the test pellets ranged from 0.1030 to 2.060 μg. The ratio of the geometric contact surface area (0.505 cm^2) between the two pellets to the source-pellet geometric surface area (4.17 cm^2) was 0.121. The product of the surface area ratio and the total mass of uranium on an individual source pellet is the approximate amount of uranium available for transfer to the test pellet (Table 1), since the loaded uranium was primarily on the surface. The fractional percentage of available uranium transferred to the test pellets was small, in most instances $<0.3\%$ (Table 1), indicating that the source-pellet uranium concentrations remained relatively constant during the tests. There was no obvious correlation between the amounts of uranium transferred and the time and temperature of the tests.

3.2 DIFFUSION COEFFICIENT ESTIMATION

The penetration data used for estimation of the diffusion coefficients for uranium initially present as UO_2 are shown in Table 2, and the data for uranium present as UC_2 are in Table 3. These data are derived solely from the test pellets, since the source-pellet data are not required for diffusion coefficient calculations.

While the experiments were designed to provide a constant uranium source, in actuality the uranium source concentration at the surface of the test pellets may have varied, as discussed in Sect. 2.1. The distribution coefficients calculated using the assumption of a constant source concentration (Sect. 3.2.1) have therefore been tested against a second mathematical model (Sect. 3.2.2) to determine sensitivity with respect to the assumed model. The alternate model assumes instantaneous transfer of fixed amounts of uranium to the test pellets.

3.2.1 Diffusion coefficients based on a constant source concentration

The diffusion data were first considered in terms of the equation[3,4] shown below, which is a solution of Fick's second law assuming a model with a constant source of supply of the diffusing substance:

Table 2. Effects of time and temperature on the penetration of uranium as UO_2 in H-451 graphite

Temperature (°C)	Time (d)	Penetration distance (cm)	Uranium concentration [(g/cm^3) × 10^6]
900	21	0.0014	8.09
		0.0049	2.61
		0.0090	0.02
1000	7	0.0019	10.50
		0.0051	0.07
1000	11	0.0011	54.7
		0.0033	15.8
1000	11	0.0014	15.0
		0.0038	2.5
1000	21	0.0013	17.6
		0.0035	7.6
1000	21	0.0009	1.6
		0.0047	0.3
1400	7	0.0014	1600
		0.0047	1350
		0.0094	950
		0.0137	540
		0.0175	408

Table 3. Effects of time and temperature on the penetration of uranium as UC_2 in H-451 graphite

Temperature (°C)	Time (d)	Penetration distance (cm)	Uranium concentration [(g/cm^3) × 10^4]
1000	12	0.0014	58.00
		0.0032	21.00
		0.0082	4.97
		0.0117	1.95
1400	4	0.0092	10.50
		0.0144	8.01
		0.0206	6.72

$$\left[\mathrm{erf}^{-1}\left(1 - \frac{C}{C_0}\right)\right]^2 = \frac{x^2}{4Dt} \; . \qquad (2)$$

In this equation, C is the concentration of uranium at time, t, at a distance, x, into the pellet; C_0 is a constant representing the uranium concentration at x = 0 for all t; and D is the diffusion coefficient. Most solid-state diffusion data give essentially linear plots for log C vs x^2. This type of plot (not shown) was used to extrapolate to x = 0 for effective uranium concentrations at the surface of the test pellets; these values are listed in Table 4. Plots of the left side of Eq. (2) vs x^2 have an intercept at the origin and a slope of 1/4Dt, allowing evaluation of D. Examples of such plots using Eq. (2) are shown in Fig. 3 for UC_2 at 1000°C and in Fig. 4 for UO_2 (initial) at 1400°C.

For the method using a constant uranium source, the diffusion coefficients of uranium initially present as UO_2 were evaluated for tests at 900, 1000, and 1400°C, respectively, to be 5.43×10^{-12}, 3.80×10^{-12}, and 1.55×10^{-10} cm^2/s. The value for 1000°C is an average of 5 separate test values ranging from 3.30×10^{-12} to 4.30×10^{-12} cm^2/s. The coefficients for uranium present as UC_2 were evaluated for tests at 1000 and 1400°C, respectively, to be 1.86×10^{-11} and 1.80×10^{-9} cm^2/s (Table 5).

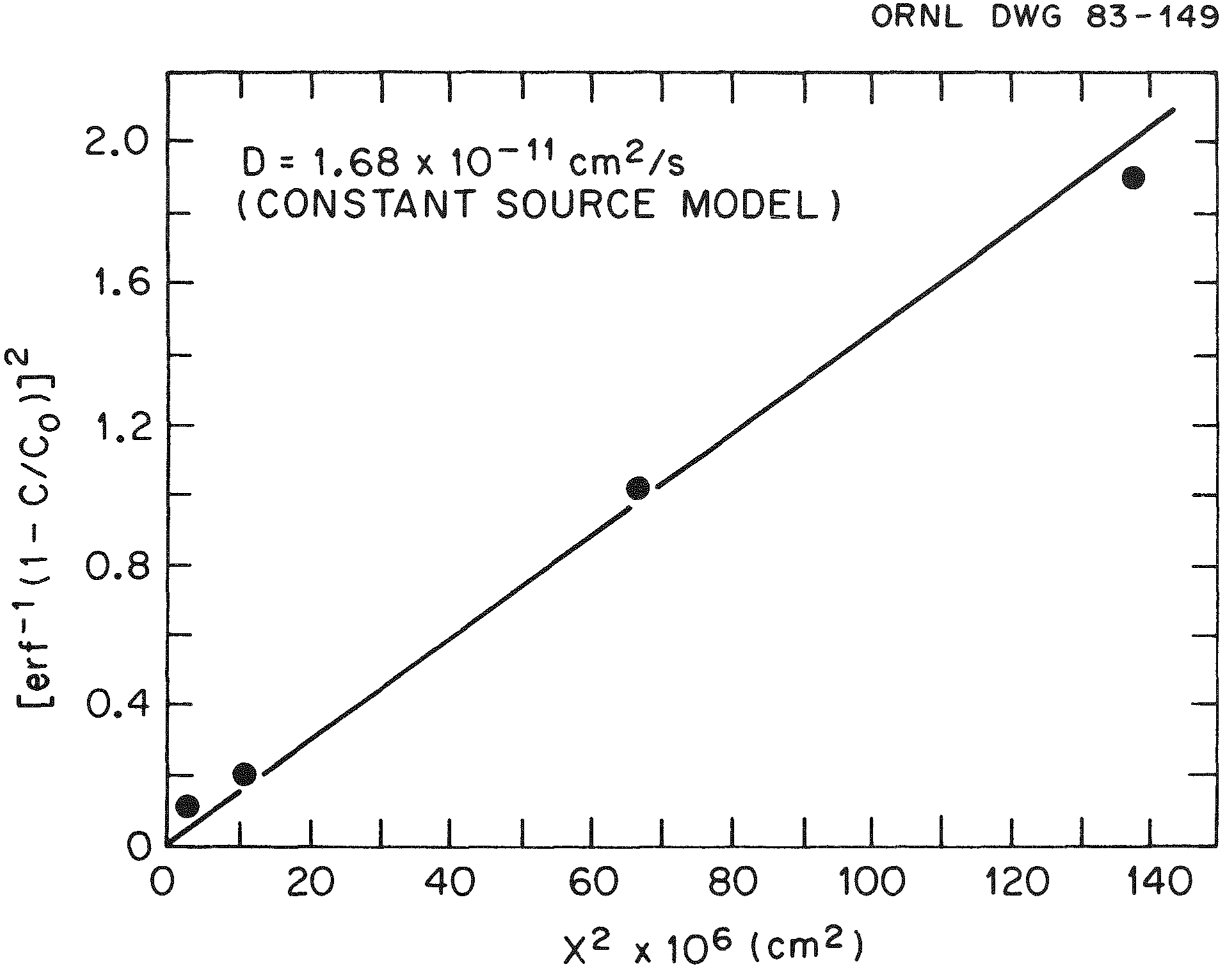

Fig. 3. Penetration profile for the diffusion of uranium as UC_2 in H-451 graphite at 1000°C for 12 d.

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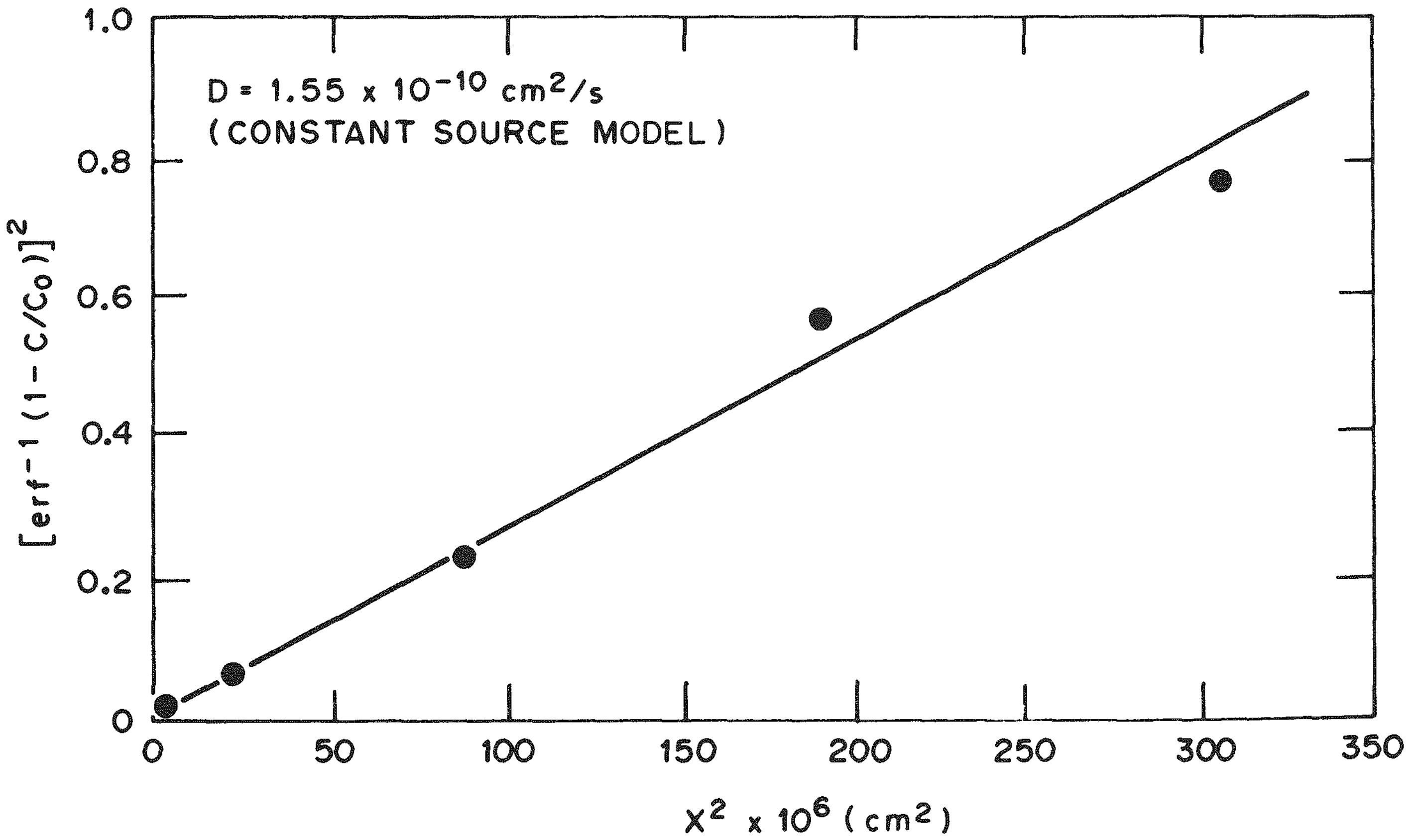

Fig. 4. Penetration profile for the diffusion of uranium initially present as UO_2 in H-451 graphite at 1400°C for 7 d (constant source model).

Table 4. Estimated test pellet uranium concentrations at zero penetration distance

Temperature (°C)	Time (d)	Initial form of uranium	Uranium concentration[a] at zero penetration distance, C_0 [(g/cm^3) × 10^5]
900	21	UO_2	1.26
1000	7	UO_2	2.37
1000	11	UO_2	6.42
1000	11	UO_2	2.01
1000	21	UO_2	2.04
1000	21	UO_2	0.17
1400	7	UO_2	145.00
1000	12	UC	400.00
1400	4	UC	105.00

[a] These estimated concentrations are based on extrapolations to $x = 0$ of log C vs x^2 penetration curves, where C is the uranium concentration at a penetration distance x in the graphite.

Table 5. Diffusion coefficients for uranium in H-451 graphite at 900 to 1400°C

Initial uranium source	Temperature (°C)	Diffusion coeffients Assuming decreasing uranium source (cm^2/s)	Diffusion coeffients Assuming constant uranium source (cm^2/s)
UO_2	900	1.73×10^{-12}	3.54×10^{-12}
UO_2	1000	1.85×10^{-12}[a]	3.80×10^{-12}[b]
UO_2	1400	9.09×10^{-11}	1.55×10^{-10}
UC	1000	1.06×10^{-11}	1.68×10^{-11}
UC	1400	5.80×10^{-10}	1.80×10^{-9}

[a] The 1.85×10^{-12} cm^2/s value for 1000°C is an average of 5 test values ranging from 1.72×10^{-12} to 2.05×10^{-12} cm^2/s.

[b] The 3.80×10^{-12} cm^2/s value for 1000°C is an average of 5 test values ranging from 3.3×10^{-12} to 4.3×10^{-12} cm^2/s.

3.2.2 Diffusion coefficients based on instantaneous fixed transfer

In this discussion, the data are considered in terms of the following equation, which is a solution of Fick's second law assuming a decreasing uranium source, or the instantaneous transfer of fixed amounts of uranium to the test pellets:[5]

$$\log C = \log C_0 - \frac{x^2}{9.2Dt} \tag{3}$$

In this equation, C, C_0, X, D, and t have the same designations as were used for Eq. (2), except that C_0 decreases with time (C_0 is not used in the calculations of D in this model). Plots of log C vs x^2 are expected to be linear and to have a slope of 1/9.2Dt, allowing evaluation of D. Examples of such plots are shown in Fig. 5 for UC_2 at 1000°C and in Fig. 6 for UO_2 (initial) at 1400°C. The diffusion coefficients for uranium initially present as UO_2 using this method were evaluated for tests at 900, 1000, and 1400°C to be, respectively, 1.73×10^{-12}, 1.85×10^{-12}, and 9.09×10^{-11} cm^2/s (Table 5). The 1.85×10^{-12} cm^2/s value is an average of 5 values ranging from 1.72×10^{-12} to 2.05×10^{-12} cm^2/s. The coefficients for uranium present as UC_2 were evaluated for tests at 1000 and 1400°C to be 1.06×10^{-11} and 5.80×10^{-10} cm^2/s, respectively (Table 5).

4. DISCUSSION

The diffusion coefficients evaluated using each of the mathematical models that have been discussed here are listed in Table 5. The coefficients for the two models are fractionally different by ~20 to 50%. Based on the order of magnitude of the numerical values, the differences may be considered insignificant. It was expected that differences in the diffusion coefficients evaluated using the two models would be minimal, since the total amounts of uranium transferred were small. The Arrhenius equations obtained using the constant uranium surface concentration model for the diffusion coefficients for UC_2 and UO_2 (initial), respectively, are:

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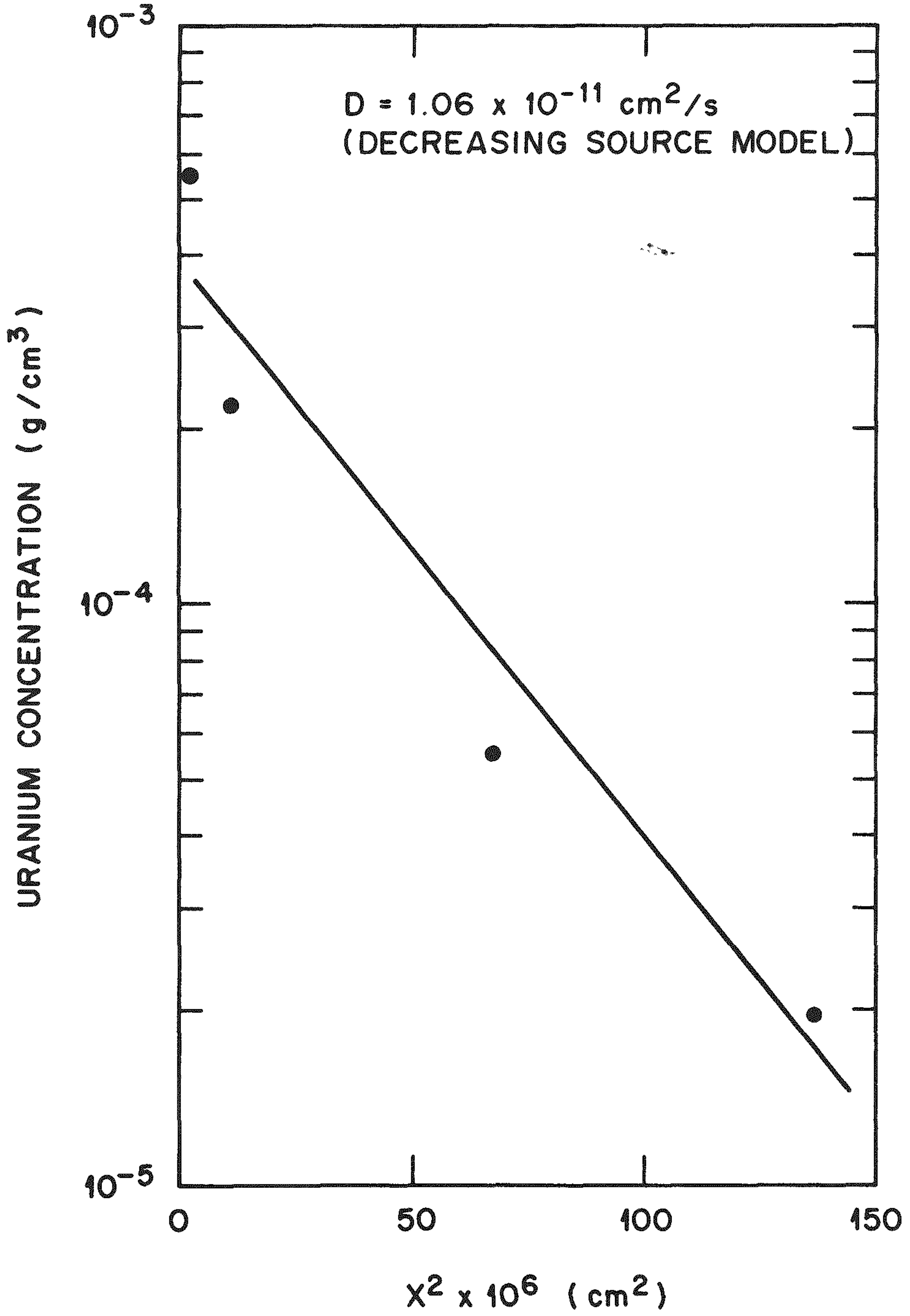

Fig. 5. Penetration profile for the diffusion of uranium as UC_2 in H-451 graphite at 1000°C for 12 d.

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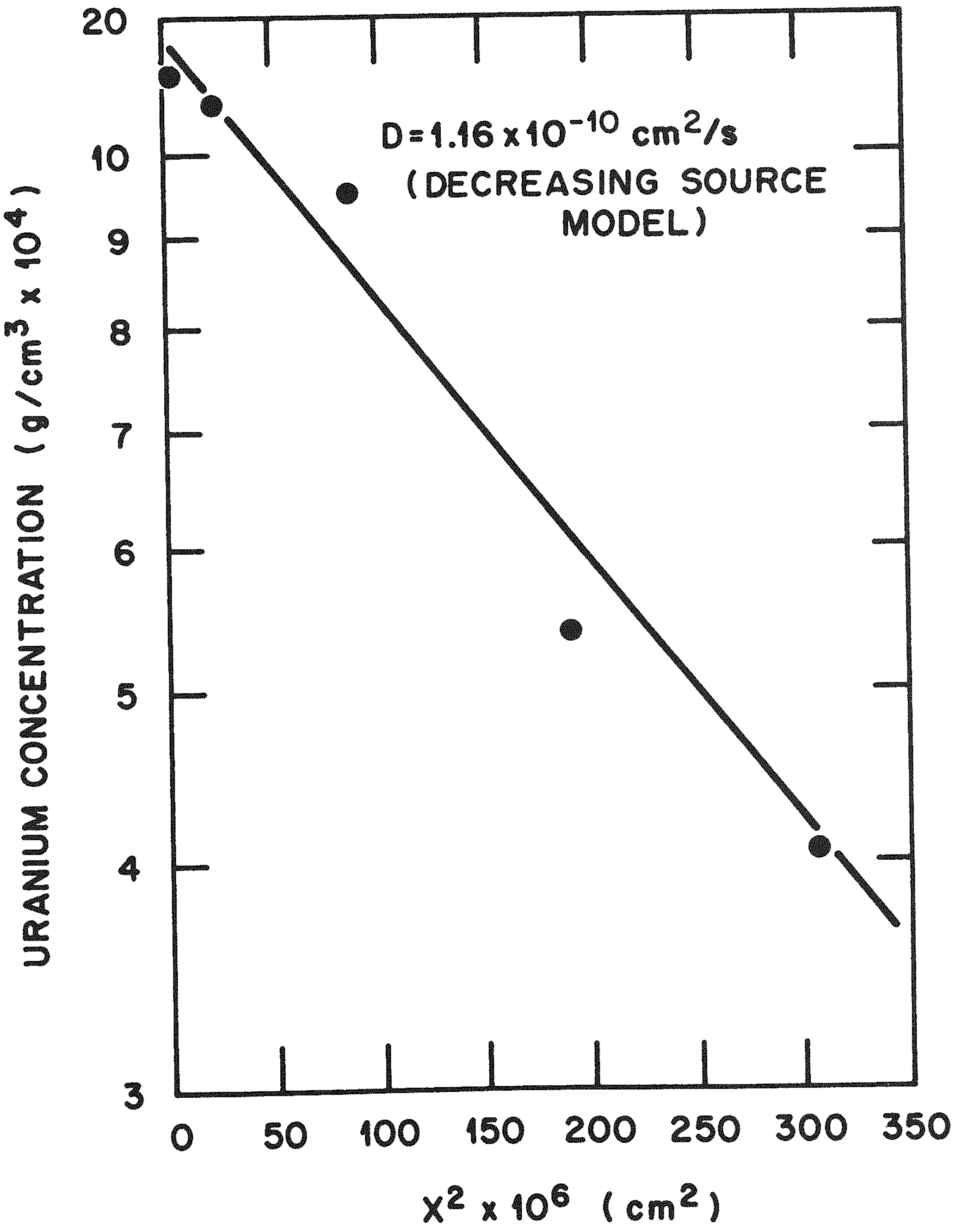

Fig. 6. Penetration profile for the diffusion of uranium initially present as UO_2 in H-451 graphite at 1400°C for 7 d (decreasing source model).

$$D_{UC_2} = 3.5 \times 10^{-3}\ e^{-4.8 \times 10^4/RT}\ \mathrm{cm^2/s}\ , \qquad (4)$$

and

$$D_{UO_2} = 2.34 \times 10^{-6}\ e^{-3.2 \times 10^4/RT}\ \mathrm{cm^2/s}\ . \qquad (5)$$

The activation energies for UC_2 and UO_2 diffusion [based on Eqs. (4) and (5)] are 201 and 136 kJ/mol, respectively. Arrhenius plots of the data are shown in Fig. 7, and from the data shown in these plots and in Table 5, it can be seen that the coefficients for the carbide are ~5 to 12 times greater in the temperature range investigated than those for the oxide. The coefficients for the carbide are also more temperature dependent. Use of the equations to calculate diffusion coefficients for UO_2 and UC_2 probably should be limited to the temperature range of 800 to 1500°C, since Arrhenius plots for the diffusion of a number of heavy metals have been shown to be nonlinear.[6]

Assuming that the diffusion behavior of plutonium is similar to that of uranium, the results of this study indicate that the present assumption of zero plutonium hold-up in the core graphite of HTGRs is unrealistic. The diffusion data presented here should aid in making more exact estimates of plutonium contamination of the HTGR primary cooling system.

We expect to use the techniques and equipment developed for this study to continue our investigation of the diffusion of plutonium.

5. ACKNOWLEDGMENTS

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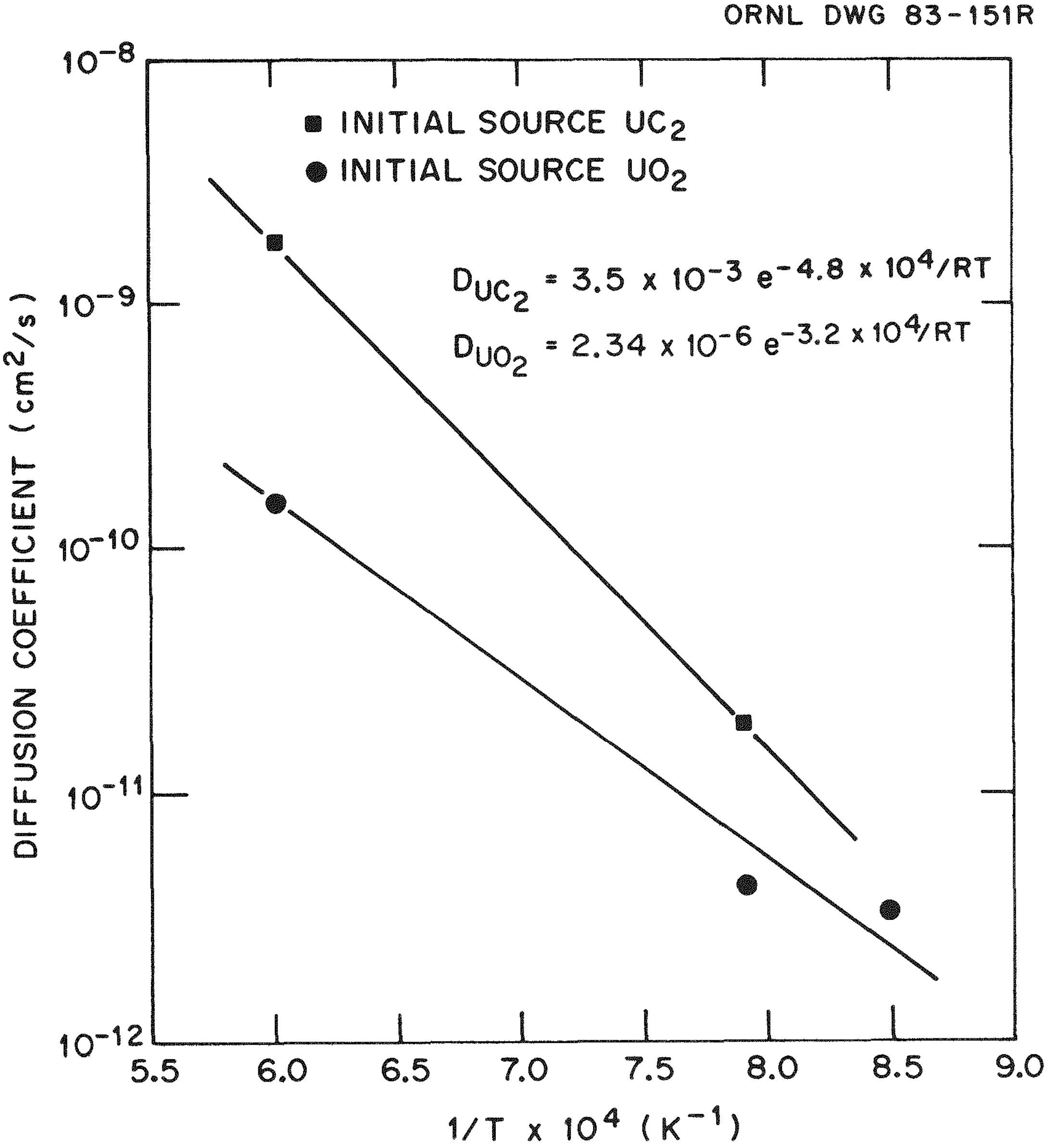

Fig. 7. Arrhenius plots of uranium diffusion coefficients in H-451 graphite (constant surface uranium concentration method).

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