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CESIUM DIFFUSION COEFFICIENTS AND OTHER RESULTS
FROM THE ANALYSIS OF PEACH BOTTOM D13-05
FUEL ELEMENT ACTIVITY PROFILES

by

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ABSTRACT

Fission product activity profiles of Sr-89, Sr-90, Cs-134, and Cs-137 from radial sections taken from the spine and sleeve of Peach Bottom fuel element D13-05 have been analyzed. The Sr-89 and Sr-90 activity profiles appear to have resulted from the decay of their gaseous precursors, Kr-89 and Kr-90. Thus, no strontium metal diffusion data were obtained.

It was determined that gaseous precursor concentration profiles in the sleeve may be similar in shape to that of the daughter metal owing to strong counter-flowing convective currents. Because of the uncertainty of the xenon precursor contribution to the total activity profile, cesium metal diffusion coefficients were not clearly established for the sleeve graphite.

For the spine region of the fuel element, each profile component (gaseous precursor and directly released metal) was fitted quite closely by curves calculated using the FIPER code. This correlation instills confidence in the FIPER code and lends support to the concept that both metal and gaseous precursor follow a transport behavior which closely approximates that expected from simple classical diffusion, in the absence of strong convective currents. Analysis of the cesium metal diffusion profiles gives a diffusion coefficient and an activation energy of D_{Cs} (1000°C) = $7 \times 10^{-9} \text{ cm}^2/\text{sec}$ and $Q = 27 \text{ kcal/mole}$, respectively.

NOMENCLATURE

s	= distance along pore
t	= time
x	= distance
y	= $(V/D_g) \xi L$
C	= concentration
C_L	= boundary concentration at $x = L$
C_0	= boundary concentration at $x = 0$
D_{eff}	= reduced gaseous diffusion coefficient resulting from pores occupying only a fraction of total volume
D_g	= diffusion coefficient of gas in a pore
J	= current of the fission product in the pore
K	= mass transport coefficient
L	= thickness of a slab
Q	= activation energy
T	= temperature
V	= average velocity of helium flow in a pore
λ	= radionuclide decay constant
μ	= viscosity of gas
ξ	= tortuosity of the pore
ϕ	= sorption ratio

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INTRODUCTION

Information on metallic fission product behavior obtained from High-Temperature Gas-Cooled Reactor (HTGR) fuel materials which have operated in-pile or as components of actual fuel elements is most valuable because the availability of such data is quite limited. Diffusion data obtained from laboratory experiments are sometimes difficult to interpret owing to the difficulty of simulating in-reactor conditions such as the very pronounced effect of concentration on the diffusion coefficient of metals in graphite (Ref. 1). The early work of Bromley, et al. (Refs. 2,3), on the diffusion of cesium in four different nuclear-grade graphites, for example, yielded a very high diffusion coefficient at activation energies of $Q = 16.4$ to 19.2 kcal/mole. However, these were for cesium concentrations far greater than might be found in an HTGR.

Fission product activity profiles obtained in connection with post-irradiation examinations of Peach Bottom HTGR fuel elements provide additional needed data on fission product behavior and permit determination of diffusion coefficients from metallic fission product profiles measured in the fuel elements. Accordingly, the work reported here comprises an analysis of cesium (Cs-134 and Cs-137) and strontium (Sr-89 and Sr-90) concentration profiles found in the spine and sleeve graphites of Peach Bottom fuel element D13-05. The FIPER code (Ref. 4) was used to compute theoretical profiles which were fitted to the actual data in the process of analysis. The goodness-of-fit then gave an indication of the accuracy of FIPER in representing actual fission product transport behavior.

SAMPLING PROCEDURE AND ANALYSIS

Fuel element D13-05 was one of 804 fuel elements of Core 1 of the 115-MW(t) Peach Bottom HTGR which operated for 452 effective full power days before shutdown in October 1969. D13-05 was returned to Gulf General Atomic for postirradiation examination, which included sectioning of the spine and sleeve and analysis for the concentration of long-lived cesium and strontium activities.

The sectioning was accomplished by turning the sleeve and spine graphite on a lathe, uniformly removing a layer of graphite for each section. This technique is believed superior to that of taking corings of a sleeve or spine, whereby only localized concentrations are measured. By turning the specimen on a lathe, a truly average sample is obtained and any circumferential variations caused by graphite inhomogeneity are averaged. The columns in Table 1 designated "Distance from Centerline" indicate the average concentration about that radius.

Weighed samples of the sections were gamma-analyzed using a Ge(Li) crystal detector and a 4096-channel pulse height analyzer. Radiochemical analysis involving chemical separation and beta counting was performed to determine Sr-89 and Sr-90 concentrations. Table 1 includes all the cesium and strontium concentration profile data. These data are for radial sections of the spine and sleeve taken at certain fuel compact positions. Figure 1 illustrates the fuel element and indicates the location of the 3-in.-long fuel compacts. The first fuel compact (No. 1) is 26 in. above the bottom of the fuel element. Thirty compacts are stacked in the sleeve around the spine to give a fueled region 90 in. long in the 144-in.-long fuel element.

TABLE 1A
SPINE CONCENTRATION PROFILE DATA FOR CESIUM

Distance from Centerline (cm)	Cs-137 Concentration ($\mu\text{moles/cm}^3$)	Cs-134 Concentration ($\mu\text{moles/cm}^3$)	Distance from Centerline (cm)	Cs-137 Concentration ($\mu\text{moles/cm}^3$)	Cs-134 Concentration ($\mu\text{moles/cm}^3$)
Compact Position No. 2			Compact Position No. 5		
0.41	1.5-6 ^(a)	7.2-8	0.42	2.6-6	8.3-8
0.65	2.1-6	6.4-8	0.69	4.0-6	9.6-8
0.90	3.7-6	7.6-8	0.94	6.0-6	9.0-8
1.16	5.8-6	7.4-8	1.19	1.0-5	9.2-8
1.41	1.0-5	7.0-8	1.44	1.8-5	1.0-7
1.66	2.0-5	7.2-8	1.70	2.4-5	2.1-7
1.89	3.3-5	8.2-8	1.93	4.7-5	1.1-6
2.04	3.7-5	1.5-7	2.08	1.2-4	5.4-6
2.12	7.5-4	8.3-7	2.15	1.7-3	8.2-5
2.16	1.4-3	3.6-5	2.19	4.7-3	2.1-4
Compact Position No. 9			Compact Position No. 13		
0.42	7.9-6	2.9-7	0.44	2.5-5	1.8-6
0.69	1.1-5	3.7-7	0.69	3.7-5	2.9-6
0.94	1.8-5	8.2-7	0.94	7.5-5	6.4-6
1.19	4.7-5	2.9-6	1.19	1.8-4	1.5-5
1.44	1.8-4	1.2-5	1.44	5.4-4	4.6-5
1.70	6.1-4	4.4-5	1.70	1.7-3	1.3-4
1.93	2.0-3	1.4-4	1.93	3.8-3	3.3-4
2.08	5.1-3	3.2-4	2.08	8.5-3	6.8-4
2.15	1.1-2	7.6-4	2.15	1.1-2	1.3-3
2.19	7.5-2	4.7-3	2.19	6.1-2	4.2-3
Compact Position No. 18			Compact Position No. 22		
0.44	3.7-4	3.3-5	0.44	7.6-4	6.1-5
0.69	5.8-4	5.1-5	0.69	9.7-4	7.9-5
0.94	1.0-3	9.0-5	0.94	1.4-3	1.1-4
1.19	1.2-3	1.5-4	1.19	2.6-3	1.9-4
1.44	5.1-3	4.2-4	1.44	4.7-3	3.6-4
1.70	1.2-2	9.3-4	1.70	9.0-3	6.7-4
1.93	2.5-2	1.8-3	1.93	2.0-2	1.3-3
2.08	4.4-2	3.2-3	2.08	3.3-2	2.2-3
2.15	7.6-2	5.6-3	2.15	4.7-2	3.2-3
2.19	2.0-1	1.3-2	2.19	1.1-1	7.1-3
Compact Position No. 29					
0.42	3.9-5	1.7-6			
0.69	5.1-5	2.2-6			
0.94	9.2-5	3.9-6			
1.19	1.5-4	6.4-6			
1.44	3.5-4	1.4-5			
1.70	8.2-4	3.2-5			
1.97	1.7-3	6.4-5			
2.08	2.9-3	1.1-4			
2.15	3.2-3	1.5-4			
2.19	1.9-2	5.7-4			

(a) 1.5-6 means 1.5×10^{-6}

TABLE 1B
SLEEVE CONCENTRATION PROFILE DATA FOR CESIUM

Distance from Centerline (cm)	Cs-137 Concentration ($\mu\text{moles}/\text{cm}^3$)	Cs-134 Concentration ($\mu\text{moles}/\text{cm}^3$)	Distance from Centerline (cm)	Cs-137 Concentration ($\mu\text{moles}/\text{cm}^3$)	Cs-134 Concentration ($\mu\text{moles}/\text{cm}^3$)
Compact Position No. 2			Compact Position No. 5		
3.54	1.3-3 ^(a)	4.6-5	3.50	1.4-2	8.1-4
3.61	6.1-5	2.4-6	3.54	9.7-4	5.3-5
3.69	1.1-5	6.5-7	3.61	5.1-4	3.1-5
3.81	6.8-6	3.5-7	3.69	1.8-4	1.1-5
3.94	2.9-6	--	3.80	5.3-5	3.1-6
4.05	3.6-6	1.8-7	3.90	1.5-5	7.1-7
4.14	3.3-6	1.9-7	4.00	7.4-6	7.9-7
4.20	3.2-6	1.8-7	4.10	4.4-6	4.2-7
4.40	4.6-4	3.5-5	4.20	3.6-6	4.2-7
Compact Position No. 9			Compact Position No. 29		
3.50	3.1-2	3.1-3	3.66	1.3-3	5.0-5
3.54	8.6-3	6.3-4	3.79	5.6-4	2.4-5
3.61	6.3-3	4.7-4	3.92	2.5-4	1.1-5
3.69	3.9-3	2.5-4	4.04	1.2-4	5.1-6
3.80	1.7-3	1.3-4	4.14	6.1-5	2.8-6
3.90	8.9-4	7.1-5	4.20	4.3-5	1.8-6
4.00	4.6-4	3.7-5	4.26	2.5-5	1.1-6
4.10	2.5-4	1.9-5	4.33	1.1-5	6.1-7
4.20	1.2-4	9.9-6	4.39	7.1-6	5.7-7

(a) 1.3-3 means 1.3×10^{-3} .

TABLE 1C
SPINE CONCENTRATION PROFILE DATA FOR STRONTIUM

Distance from Centerline (cm)	Sr-90 Concentration ($\mu\text{moles/cm}^3$)	Sr-89 Concentration ($\mu\text{moles/cm}^3$)	Distance From Centerline (cm)	Sr-90 Concentration ($\mu\text{moles/cm}^3$)	Sr-89 Concentration ($\mu\text{moles/cm}^3$)	Distance From Centerline (cm)	Sr-90 Concentration ($\mu\text{moles/cm}^3$)	Sr-89 Concentration ($\mu\text{moles/cm}^3$)
Compact Position No. 2			Compact Position No. 9			Compact Position No. 18		
0.41	3.9-7 ^(a)	2.6-6	0.44	9.3-8	3.5-6	0.44	7.5-6	2.8-6
0.65	2.1-8	2.7-6	0.69	2.5-7	2.6-6	0.69	5.1-6	3.7-6
0.90	1.6-7	9.5-7	0.94	1.0-5	3.2-6	0.94	4.6-6	1.9-6
1.41	2.6-6	1.2-6	1.44	1.1-5	6.7-6	1.44	2.5-5	2.7-6
1.89	4.7-5	5.6-6	1.93	7.7-5	2.6-5	1.93	7.3-5	5.4-6

(a) 3.9-7 means 3.9×10^{-7} .

TABLE 1D
SLEEVE CONCENTRATION PROFILE DATA FOR STRONTIUM

Distance from Centerline (cm)	Sr-90 Concentration ($\mu\text{moles/cm}^3$)	Sr-89 Concentration ($\mu\text{moles/cm}^3$)	Distance from Centerline (cm)	Sr-90 Concentration ($\mu\text{moles/cm}^3$)	Sr-89 Concentration ($\mu\text{moles/cm}^3$)
Compact Position No. 2			Compact Position No. 9		
3.6	2.8-5 ^(a)	3.6-5	3.6	6.6-6	2.4-5
3.8	4.1-6	1.2-5	3.8	1.4-6	1.6-5
4.0	8.8-7	1.7-5	4.0	1.4-6	6.0-6
4.2	5.8-7	2.4-5	4.2	1.3-6	5.1-5

(a) 2.8-5 means 2.8×10^{-5} .

Fig. 1. Peach Bottom fuel element

A cross-sectional view of the fuel region of the fuel element is given in Fig. 2, along with the "as-built" dimensions. A sleeve of low-permeability graphite contains the fuel compacts. This sleeve is made from Great Lakes Carbon Company HLM-85 graphite (referred to as H-253 graphite by purchase order number). The sleeve was impregnated several times to achieve a helium permeability of approximately 10^{-5} cm²/sec at reactor operating conditions, or 10^{-3} cm²/sec at 1 atm. The density of the sleeve is 1.92 to 1.96 g/cm³. In the fuel compact at the fuel-sleeve interface are purge grooves which assist in sweeping out fission products. The fuel compacts have a fuel binder matrix material density of 1.71 g/cm³. The fuel compact surrounds a graphite spine made from Speer Carbon Company 710G base stock. After a single pitch impregnation, the graphite (designated 711 GSXY) had a density of 1.84 g/cm³ and a permeability of 0.11 cm²/sec at 1 atm.

The graphite used for the Fort St. Vrain fuel elements is also a nuclear-grade, extruded needle-coke graphite (Great Lakes Carbon Company H-327). This graphite has a bulk density of 1.72 g/cm³ and a helium permeability of about 1.0 cm²/sec at 1 atm.

SPINE O.D. 1.732 ± 0.003 IN.

FUEL COMPACT O.D. $2.743 \begin{smallmatrix} +0.003 \\ -0.005 \end{smallmatrix}$ IN.

FUEL COMPACT I.D. 1.75 ± 0.005 IN.

SLEEVE O.D. 3.485 ± 0.005 IN.
I.D. 2.752 ± 0.002 IN.

PURGE GROOVES $0.108 \begin{smallmatrix} +0.010 \\ -0.000 \end{smallmatrix}$ IN. WIDTH

$0.054 \begin{smallmatrix} +0.010 \\ -0.000 \end{smallmatrix}$ IN. DEPTH

Fig. 2. Cross section of fuel element fuel region

THERMAL ANALYSIS

The temperature profiles for each axial (fuel compact) position are given in Table 2. The temperatures were calculated using the TAC 2D code (Ref. 5). The conditions for this analysis were confirmed by reviewing Peach Bottom reactor operation data. The results are shown in Table 3.

Figure 3, which gives power factors, shows element D13-05 and its surrounding positions in the core, one of which is an empty guide tube. In calculating the temperatures, symmetry was assumed about the cluster of elements as indicated by the horizontal line in Fig. 3.

The principal uncertainty in temperature calculations arises from uncertainty in the data on irradiation-induced dimensional change in the sleeve and fuel compact. Fuel temperatures are sensitive to the size of the clearance between the fuel compact and sleeve. The manufacturing, experimental, and computational uncertainties in calculating maximum fuel temperature yield a standard deviation of 105°C for the as-built fuel element (Ref. 6). The uncertainty in the temperature calculation assuming zero gap between the fuel compact and sleeve has a standard deviation of 78°C. A review of reactor operating conditions indicated that the power distribution about the element was approximately constant with time. Likewise, when the reactor was operating, it normally operated at approximately 95% design power and 105% design coolant flow conditions.

TABLE 2
D13-05 TEMPERATURE PROFILES (°C)

Reference Angle θ (degrees)	Purge Gap Thickness = 0.0						Purge Gap Thickness = 0.0055 in.					
	Spine Center- line	Spine Surface	Fuel Compact ID	Fuel Compact OD	Sleeve ID	Sleeve OD	Spine Center- line	Spine Surface	Fuel Compact ID	Fuel Compact OD	Sleeve ID	Sleeve OD
Compact 2												
0	553	546	543	505	493	453	620	614	611	573	491	452
30	553	547	544	506	494	454	620	615	612	574	492	453
60	553	550	547	512	501	470	621	617	615	579	500	469
90	553	551	550	514	502	462	621	619	617	581	500	461
120	553	553	552	517	506	474	621	620	620	584	504	473
150	553	553	552	515	503	462	621	621	620	584	500	460
180	553	553	552	515	502	461	621	622	622	584	500	459
Compact 5												
0	703	691	685	623	603	538	800	788	783	721	599	535
30	703	693	688	626	606	541	800	790	785	724	602	538
60	704	697	694	637	618	567	801	794	790	733	616	566
90	704	701	699	641	621	557	801	798	795	738	619	555
120	705	704	703	647	628	577	801	801	799	743	626	576
150	705	705	704	644	624	558	802	802	801	742	620	555
180	705	705	704	644	623	557	802	803	802	742	619	554
Compact 9												
0	835	817	808	726	700	617	946	929	921	840	695	613
30	836	820	812	731	706	621	946	931	925	845	700	617
60	837	827	823	750	726	662	948	938	934	860	724	660
90	837	834	832	760	735	656	948	944	942	870	733	654
120	838	839	838	769	745	681	948	949	948	877	743	680
150	838	841	840	766	740	657	949	952	950	877	735	654
180	838	842	841	766	740	656	949	952	951	877	735	653

TABLE 2 (Continued)

Reference Angle θ (degrees)	Purge Gap Thickness = 0.0						Purge Gap Thickness = 0.0055 in.					
	Spine Center- line	Spine Surface	Fuel Compact ID	Fuel Compact OD	Sleeve ID	Sleeve OD	Spine Center- line	Spine Surface	Fuel Compact ID	Fuel Compact OD	Sleeve ID	Sleeve OD
Compact 13												
0	938	917	906	815	786	672	1052	1032	1022	932	779	688
30	939	920	911	821	792	698	1052	1035	1026	938	785	693
60	940	929	925	844	818	747	1055	1043	1038	956	815	745
90	941	938	936	858	830	743	1054	1051	1048	969	828	741
120	942	944	943	867	841	771	1055	1056	1055	978	840	770
150	942	946	945	864	835	744	1056	1059	1058	977	830	741
180	942	947	946	863	835	743	1056	1060	1059	977	829	739
Compact 18												
0	1008	981	969	876	847	755	1112	1088	1077	986	840	749
30	1008	986	976	884	855	761	1113	1092	1083	994	848	756
60	1010	998	993	914	889	820	1115	1102	1097	1018	886	817
90	1011	1009	1008	935	909	827	1115	1113	1111	1036	908	826
120	1012	1018	1018	946	922	855	1116	1120	1120	1047	921	854
150	1013	1021	1020	943	916	828	1117	1124	1123	1046	911	825
180	1013	1021	1021	943	915	827	1117	1125	1125	1047	911	824
Compact 22												
0	975	948	936	855	831	752	1062	1038	1027	949	823	747
30	976	953	942	864	839	758	1063	1042	1033	957	832	754
60	977	965	961	895	874	815	1065	1053	1048	983	872	816
90	978	978	978	920	899	834	1065	1065	1063	1004	899	833
120	980	986	988	931	911	857	1067	1072	1073	1015	911	857
150	980	990	990	928	906	835	1067	1075	1076	1014	903	833
180	981	991	991	928	906	835	1067	1077	1077	1015	902	832

TABLE 2 (Continued)

Reference Angle θ (degrees)	Purge Gap Thickness = 0.0						Purge Gap Thickness = 0.0055 in.					
	Spine Center- line	Spine Surface	Fuel Compact ID	Fuel Compact OD	Sleeve ID	Sleeve OD	Spine Center- line	Spine Surface	Fuel Compact ID	Fuel Compact OD	Sleeve ID	Sleeve OD
Compact 29												
0	916	888	876	817	800	745	973	949	938	882	792	740
30	916	893	883	825	808	751	974	953	945	890	801	746
60	918	907	903	860	847	810	976	965	962	918	845	808
90	919	921	923	890	878	841	976	978	978	944	880	842
120	921	931	934	902	890	857	979	986	989	955	891	858
150	922	934	937	901	887	843	979	990	992	955	836	843
180	922	936	938	901	887	843	979	991	991	956	885	842

TABLE 3
DIFFUSION COEFFICIENTS FROM PEACH BOTTOM D13-05 SPINE CONCENTRATION PROFILES

Compact Position	Spine Temperature Range (°C)		Cs Precursor, D_g (cm ² /sec)	Cs Metal, D (cm ² /sec)	Cs Conc. Spine Cs Conc. Fuel Matrix	Sr Precursor, D_g (cm ² /sec)
	5-mil Purge Gap	No Gap				
2	614 - 622	546 - 553	3.70×10^{-4}	1.0×10^{-10} 6.0×10^{-12}	1.0×10^{-5} 1.0	4.2×10^{-4}
5	788 - 803	691 - 705	5.0×10^{-4}	8.0×10^{-10} 6.0×10^{-11}	4.1×10^{-6} 5.5×10^{-4}	8.5×10^{-4}
9	929 - 952	817 - 842	6.22×10^{-4}	$2.4 \times 10^{-9(a)}$	1.0×10^{-3}	
13	1032 - 1060	917 - 947	7.33×10^{-4}	3.5×10^{-9}	1.3×10^{-3}	
18	1088 - 1125	981 - 1021	8.07×10^{-4}	3.5×10^{-9}	7.5×10^{-3}	9.6×10^{-4}
22	1038 - 1077	948 - 991	7.70×10^{-4}	3.4×10^{-9}	6.5×10^{-3}	
29	949 - 991	888 - 936	7.05×10^{-4}	4.0×10^{-9}	7.4×10^{-4}	

(a) The diffusion coefficient for Cs-134 metal profile is 2.5×10^{-9} cm²/sec.

Fig. 3. Power factors

ANALYSIS OF SPINE CONCENTRATION PROFILES

CESIUM

Concentration profiles were obtained from Peach Bottom Core 1 fuel element D13-05 for seven axial positions by the procedure already described. Figures 4 through 10 are the cesium concentration profiles for the spine section. The solid lines represent the contribution of the metal's gaseous precursor to the total concentration profiles, and the dashed lines represent the metal component contributions. Diffusion coefficients for the gaseous precursor and directly released metal component were adjusted as input into the FIPER code to give a best fit of the data as indicated by the dashed and solid lines. Table 3 gives the values of diffusion coefficients that were determined.

Figure 4 indicates that for the relatively cool temperature near the coolant inlet to the core, the measured cesium concentration profile can be attributed almost completely to diffusion and decay of gaseous precursors. The gaseous precursor of Cs-137 is Xe-137, which has a 3.82-min half-life. The Cs-134 profile results from the decay of Xe-133 to stable Cs-133, which is activated to Cs-134. The half-life of Xe-133 is 5.27 days. The half-lives of Cs-134 and Cs-137 are 2.06 and 30.2 yr, respectively. Since the half-life of Xe-133 is long compared with the time required for xenon to diffuse through the spine, the precursor portion of the Cs-134 profile is flat. The Xe-137 nuclide has a much shorter half-life which results in a sloping profile. The fit of the Xe-137 precursor profile was made with the FIPER Q code, a variation of FIPER-X (Ref. 4), and was verified by comparing the results with an analytical solution.

The metal contribution to the concentration profile is shown most clearly by those data points nearest the fuel compact. There is only

Fig. 4. FIPER code fit to cesium concentration profile data of Peach Bottom Core 1, D13-05 fuel element, spine position 2

one data point in which the Cs-137 metal contribution clearly dominates the precursor contribution. Three Cs-134 data points are assumed to result from the diffusion of Cs-134 metal from the fuel compact. The precursor contribution to the concentration profile, as defined by the flat portion of the profile, is subtracted from the three points to give exclusively metal profile data. Because the limited amount of data for this metal profile can cause a large error in the determined value of the diffusion coefficient, two separate FIPER Q code analyses were fitted to the three data points to give an upper and a lower value of diffusion coefficient.

The analysis technique of Fig. 5 is the same as that of Fig. 4. The scatter in the Cs-137 precursor data causes the contribution of the metal component to be uncertain. The precursor contribution to the Cs-134 profile is very small near the fuel compact. The six Cs-134 data points are used to obtain two cesium diffusion coefficients.

Figure 6 indicates that the diffusion coefficients for Cs-137 and Cs-134 are approximately the same (2.4×10^{-9} versus 2.5×10^{-9}). The higher temperature of compact position 9 causes the cesium metal to have a higher diffusion coefficient and thus make a significant contribution to the whole profile. However, the precursor contribution is still appreciable. Using the precursor diffusion coefficient obtained from compact 2, the precursor diffusion coefficient for compact position 9 was obtained by the formula

$$\frac{D_{\text{eff}}(9)}{D_{\text{eff}}(2)} = \left(\frac{T_9}{T_2} \right)^{1.8} .$$

The 1.8 power corresponds to soft shell collision theory for gaseous diffusion. The precursor diffusion coefficient was used as input into the FIPER Q code. The resulting profile was fitted to the second data point from the spine center line. The precursor profile was then subtracted from the experimental data to give the contribution of the directly formed metal to the concentration profile.

Fig. 5. FIPER code fit to cesium concentration profile data of Peach Bottom Core 1, D13-05 fuel element, spine position 5

Fig. 6. FIPER code fit to cesium concentration profile data of Peach Bottom Core 1, D13-05 fuel element, spine position 9

Analysis of Figs. 7 through 10 proceeds in the same way. Note the accuracy with which the classical diffusion theory of the FIPER Q code fits the cesium metal concentration profile, as well as the precursor profile. In some instances, the concentration nearest the fuel compact is unusually high, possibly due to contamination of the spine surface resulting from disassembly of the fuel element.

The FIPER code fit of the experimental data was done by a trial and error procedure. The absolute magnitude of the profile was fixed by varying the sorption ratio, ϕ (ϕ is the ratio of the concentration at the outer surface of spine to the concentration at the inner radius of the fuel compact matrix). The values of the sorption ratios are given in Table 3. Only one sorption ratio and one diffusion coefficient fit the experimental data. To demonstrate the sensitivity of the calculated profiles, the cesium metal diffusion coefficient in the graphite determined by the FIPER code fit for spine position 29 has been doubled and halved and the resulting profiles have been plotted in Fig. 10.

The extraordinarily accurate fit of the FIPER Q calculated profile to the experimentally determined cesium metal profile suggests the validity of the FIPER Q code calculational techniques. The diffusion coefficients that are determined by this analysis of reactor in-pile cesium concentration profiles in nuclear graphite permit these techniques to be confidently applied to reactor design.

STRONTIUM

Figures 11, 12, and 13 are plots of the strontium data and the FIPER code fit to the data. There is a much smaller amount of data for strontium than for cesium because strontium concentration measurements depend upon beta particle detection. The cesium concentrations were measured using gamma ray spectroscopy techniques, which are much easier and subject to less error.

Fig. 7. FIPER code fit to cesium concentration profile data of Peach Bottom Core 1, D13-05 fuel element, spine position 13

Fig. 8. FIPER code fit to cesium concentration profile data of Peach Bottom Core 1, D13-05 fuel element, spine position 18

Fig. 9. FIPER code fit to cesium concentration profile data of Peach Bottom Core 1, D13-05 fuel element, spine position 22

Fig. 10. FIPER code fit to cesium concentration profile data of Peach Bottom Core 1, D13-05 fuel element, spine position 29

Fig. 11. FIPER code fit to strontium concentration profile data of Peach Bottom Core 1, D13-05 fuel element, spine position 2

Fig. 12. FIPER code fit to strontium concentration profile data of Peach Bottom Core 1, D13-05 fuel element, spine position 9.

Fig. 13. FIPER code fit to strontium concentration profile data of Peach Bottom Core 1, D13-05 fuel element, spine position 18

Two strontium metal concentration profiles were measured: Sr-89 and Sr-90. The gaseous precursor of Sr-89 is Kr-89, which decays to Rb-89 with a 3.2-min half-life, which decays to Sr-89 with a 15-min half-life. The gaseous precursor of Sr-90 is Kr-90, which decays to Rb-90 with a 33-sec half-life, which decays to Sr-90 with a 2.7-min half-life. The half-lives of Sr-89 and 90 are 50.8 days and 28.9 yr, respectively.

Figures 11, 12, and 13 show a FIPER code fit to the gaseous precursor profile. It is clear that these strontium data points result from the decay of the gaseous precursor. After a FIPER code fit of one of the concentration profiles has been made and the diffusion coefficient determined, a fit of the other profile data can be obtained merely by changing the decay constant of the precursor component. The diffusion coefficients for the strontium precursors (krypton isotopes) are similar to the diffusion coefficients of the cesium precursors (xenon isotopes).

No strontium metal profiles were detected. This seems to confirm other measurements which indicate that strontium has a lower diffusion coefficient than cesium in nuclear graphite. The problem of measuring low diffusion coefficients is illustrated by the difficulty with which the cesium metal diffusion coefficients were determined in spine positions 2 and 5.

Table 3 summarizes the temperatures and diffusion coefficients determined for spine graphite by this study. The Arrhenius plot of the cesium metal diffusion coefficient (D_{Cs}) and a comparison of these data with D_{Cs} values obtained by other investigators are presented below following a discussion of sleeve data.

ANALYSIS OF SLEEVE CONCENTRATION PROFILES

CESIUM

Figures 14 through 19 give the FIPER code fit to the experimental data in the sleeves. The similarity in the shapes of the Cs-137 and Cs-134 profiles suggests that the cesium isotopes diffuse as cesium metal atoms. Table 4 summarizes the results of this analysis. However, it may be argued that sleeve concentration profiles are due to decay of the gaseous precursor.

By comparing the temperatures for the spine and sleeve given in Tables 3 and 4, it is seen that the temperatures in sleeve positions 2, 5, and 9 are very similar to the temperatures in spine positions 2 and 5. The concentration profiles for the spines in these three positions are made up almost entirely by the precursor component. Therefore, it is expected that the concentration profiles in sleeve positions 2, 5, and 9 will also be made up almost entirely by the precursor component. The temperature in sleeve position 29 is similar to the temperature in spine position 9. The precursor contribution to spine position 9 also makes a significant contribution to the profile. Thus, because of the relative low temperatures of the sleeves, the precursor component might be expected to dominate the metal profile component. If the concentration profiles are due to the precursors, the slopes of the Cs-137 and Cs-134 profiles might be expected to be different, as they were in spine positions 2 and 5. This difference in slopes would result from the large differences in precursor half-lives.

Possibly a more important effect than differences in half-lives, however, is the influence of back-flowing helium. There is a pressure difference between the helium coolant channel and purge grooves which

Fig. 14. FIPER code fit to cesium concentration profile data of Peach Bottom Core 1, D13-05 fuel element, sleeve position 2

Fig. 15. FIPER code fit to cesium concentration profile data of Peach Bottom Core 1, D13-05 fuel element, sleeve position 5

Fig. 16. FIPER code fit to cesium concentration profile data of Peach Bottom Core 1, D13-05 fuel element, sleeve position 9

Fig. 17. FIPER code fit to cesium concentration profile data of Peach Bottom Core 1, D13-05 fuel element, sleeve position 29

Fig. 18. Temperature dependence resulting from a fit of the sleeve profile

Fig. 19. FIPER code fit to strontium concentration profile data of Peach Bottom Core 1, D13-05 fuel element, sleeve position 2

TABLE 4
DIFFUSION DATA RESULTING FROM FIPER CODE ANALYSES FOR PEACH BOTTOM D13-05
FUEL ELEMENT SLEEVE^(a)

Compact No.	Sleeve Temp. (°C)	D_{Cs} (cm ² /sec)	$\frac{\text{Cs Conc. Fuel Matrix}}{\text{Cs Conc. Sleeve}}$	Sr Precursor, D_g (cm ² /sec)
2	453 - 502	8.0×10^{-11}	3750	3.0×10^{-4}
5	538 - 623	6.0×10^{-10}	7800	
9	617 - 740	1.4×10^{-9}	1000	8.5×10^{-4}
29	745 - 887	1.6×10^{-9}	2500	

(a) Data obtained from FIPER fit to concentration profiles.

causes helium to flow from the coolant through the sleeve into the purge stream. Therefore, the gaseous precursor must diffuse against a flowing stream. The FIPER Q code is not capable of handling this transport process because it was not formulated to treat diffusion with convective flow. However, this process can be discussed in accordance with the treatment given in the Appendix for the diffusion of gaseous precursors in the case of back-flow of helium in pores of the sleeve graphite.

The significant result, as developed in the Appendix, is that for the isotopes of interest the precursor profiles will, for all practical purposes, be independent of their half-lives and their concentration will decrease exponentially in accordance with the equation $C = C_0 \exp[-(V/D_g) \xi x]$. Thus, there is no foundation for expecting Cs-134 and Cs-137 concentration profiles resulting from their precursor to have different slopes due to differences in precursor half-lives, as discussed above.

Table 5 gives the results of a calculation of $V\xi/D_g$ corresponding to an approximate fitting of the above equation to the cesium profiles given in Figs. 14 through 17 which provided the basis for the FIPER calculations. These correspond to fuel compact positions 2, 5, 9, and 29, respectively.

The values of $V\xi/D_g$ in Table 5 are plotted versus $10^4/T$ in Fig. 18, which also includes a line corresponding to $V\xi/D_g \propto (1/T)^{2.5}$, the temperature dependence as derived in the Appendix. It is seen that the points follow the temperature dependence fairly well, except in the case of position 2. Also given in Table 5 are the values of $(V/D_g) \xi$ obtained from experimental data. The procedure for arriving at these values is described in the Appendix, where the values are substituted into Eq. 8. The resulting profiles are plotted in Figs. 14 through 17 and give an indication of the expected shape of the precursor contribution. (These lines are not meant to be a fit to the data.)

Thus, there is some support for the idea that the cesium profiles of the sleeve graphite may result from the decay of the Xe-137 and Xe-134 precursors when the effect of helium back-flow is taken into account.

TABLE 5
RESULTS OF FITTING CESIUM PROFILES TO $C = C_o \exp \left(- \frac{V}{D_g} \xi x \right)$

Position	Temp. Range (°C)	$\frac{10^4}{T_{avg}}$ (°K)	$\left(\frac{V}{D_g} \xi \right)_{fit}$	$\left(\frac{V}{D_g} \xi \right)_{exp.}$
2	453 - 502	13.3	30.4	7.35
5	538 - 623	11.7	11.5	5.39
9	617 - 740	10.5	6.6	4.10
29	745 - 887	9.2	6.3	2.92

Whether the sleeve concentration profiles are due to flow and decay of a precursor or to metal diffusion, Cs-134 and Cs-137 concentration profiles would have similar shapes.

The profiles for position 2 (Fig. 14) and position 5 (Fig. 15), however, show tails which are not explained by "back-flow" theory.

One is left with some uncertainty as to whether the sleeve concentration profiles are best interpreted as being due to gaseous precursors with helium back-flow or to cesium metal diffusion with diffusion coefficients as given in Table 4.

Because the gaseous precursor contribution could not be subtracted from the profiles to give an exclusively metal profile, the apparent cesium metal diffusion coefficients listed in Table 4 are in error. No such problem exists for the spine data, so the cesium metal diffusion coefficients resulting from this study are those determined from the spine data.

It is to be noted, as shown in Fig. 20, that the diffusion coefficients obtained for positions 2 and 5 are over an order of magnitude greater than that based on data by Bryant, et al. (Ref. 7).

STRONTIUM

The data in Fig. 19 on strontium concentration profiles for sleeve position 2 do not show parallel behavior for the Sr-89 and Sr-90 profiles and are difficult to interpret (at least in the case of Sr-89). On the other hand, the data in Fig. 21 for sleeve position 9 show that the Sr-89 and Sr-90 profiles are parallel, as would be expected if the profiles were the result of gaseous precursor diffusion with helium back-flow. It appears very likely that, as in the case of the spine graphite, the strontium profiles result from gaseous precursor transport.

Fig. 20. Arrhenius plot of cesium diffusion coefficients for Peach Bottom D13-05 sleeve

Fig. 21. FIPER code fit to strontium concentration profile data of Peach Bottom Core 1, D13-05 fuel element, sleeve position 9

ARRHENIUS PLOT OF CESIUM METAL DIFFUSION COEFFICIENTS IN THE SPINE

The spine graphite is more nearly like the graphite used in large HTGRs than the sleeve graphite. Furthermore, analysis of the spine profiles did not involve the complex convective flow conditions that existed in the sleeve graphite. Cesium metal diffusion coefficients were clearly identified in the case of the spine graphite. Therefore, the cesium metal diffusion coefficients, D_{Cs} , determined by analyzing spine data are values to be considered along with previous data ("reference D_{Cs} ") for use in future computations.

Figures 22 and 23 are Arrhenius plots ($\ln D_{Cs}$ versus $10^4/T$) of the cesium spine diffusion coefficients. The dashed lines represent a fit to the data. The solid lines represent the current reference data used in calculations. The temperatures used in Fig. 22 correspond to the case where there is a 5-mil gap between the fuel compact and the sleeve. This represents the "as-built" fuel. As the reactor was operated at power, the fuel compacts swelled, closing this gap. Figure 23 is an Arrhenius plot with temperatures corresponding to this no-gap case.

It should be noted that determination of the gap between the fuel compacts and sleeve was omitted in the end-of-life experimental measurements. However, when the fuel element was disassembled, fuel compact 2 slid out, whereas the other fuel had to be forced out. It accordingly would seem that the no-gap case (Fig. 23) would be the best approximation to the actual situation. The no-gap case gives higher values for D_{Cs} at a given temperature and thus represents conservative values for a computation of cesium release.

In view of the above, the best values for D_{Cs} of the spine graphite are the fit to the experimental data shown in Fig. 23. This line corresponds to an activation energy of 27 kcal/mole and a diffusion coefficient

Fig. 22. Arrhenius plot of cesium diffusion coefficients for Peach Bottom D13-05 spine (5-mil gap between fuel compact and sleeve)

Fig. 23. Arrhenius plot of cesium diffusion coefficients for Peach Bottom D13-05 spine (zero mil gap between fuel compact and sleeve)

of $7 \times 10^{-9} \text{ cm}^2/\text{sec}$ at 1000°C . This is much higher than the very low activation energy ($Q = 3 \text{ kcal/mole}$) obtained by Vanslager (Ref. 8) from a FIPER analysis of the Peach Bottom D06-01 fuel element cesium profile data. It is believed that the low activation energy resulted from temperature uncertainties and the small range of temperature covered by the profiles. The analysis of Peach Bottom fuel element D13-05 is an improvement over the D06-01 analysis because the contribution of the gaseous precursor profile component was subtracted from the total profile to give the cesium metal profile. When this procedure is used for the low temperatures of spine compact 5 of D06-01, and the diffusion coefficient is compared with that of the other three compacts (each at essentially the same temperature), an activation energy of 25 kcal/mole is calculated. Thus, there is good agreement between the two analyses.

COMPARISON OF D_{Cs} FOR PEACH BOTTOM SPINE GRAPHITE WITH OTHER CESIUM
DIFFUSION COEFFICIENT DATA

Several measurements of cesium diffusion coefficients have been obtained in the laboratory and from postirradiation analysis of concentration profiles. Frequently, there are discrepancies between the two methods of determining diffusion coefficients because of the difficulty in duplicating in-pile conditions in the laboratory. In a paper that summarizes the British experimental results (Ref. 9), Flowers, et al., report diffusion coefficients measured in the laboratory to be orders of magnitude larger than in-pile determinations. This anomaly is attributed to gradual diffusion into graphite grains during long-term irradiations which occurs simultaneously with relatively rapid diffusion between grains. Diffusion into grains does not significantly occur in short-term laboratory experiments.

A variation of cesium diffusion coefficient with concentration has been reported by Flowers (Ref. 10). At concentration levels of $\sim 1 \mu\text{g/g}$, a line

$$\log_{10} D_{Cs} = 0.19 - \frac{10390}{T}$$

approximates the data. In the region from 100 to 1000 $\mu\text{g cesium/g graphite}$, data on AGL 9 graphite gave a line

$$\log_{10} D_{Cs} = -1.158 - \frac{3940}{T} \quad .$$

These two lines represent a cesium diffusion coefficient which varies by more than three orders of magnitude. The line representing diffusion at low concentration in a Dragon fuel tube agrees well with the data obtained from the Peach Bottom D13-05 spine analysis. At concentration levels

representative of the D13-05 analysis, cesium diffusion coefficient dependence on concentration is not thought to be significant. Bromley, et al. (Refs. 2,3), and Riedinger, et al. (Ref. 11), have also reported cesium diffusion coefficients at high concentrations.

The value of cesium diffusion coefficients is also dependent on the type of graphite. It has been reported that the cesium diffusion coefficient for graphite matrix material of fuel compacts and rods is an order of magnitude higher than that for nuclear structural graphite (Ref. 9).

The results of the D13-05 fuel element analysis are only compared directly with other in-pile analysis of nuclear graphite. It is maintained that such analysis gives a more accurate indication of the cesium diffusion in fuel elements than laboratory analyses because the difficulty of reproducing in-pile conditions in the laboratory is avoided. The current reference value of cesium diffusion coefficients was obtained from the work of Bryant, et al. (Ref. 7). Bryant's out-of-pile experiments were conducted with graphite and at temperatures which are not representative of reactor conditions.

The British have performed many postirradiation examinations of Dragon fuel tubes (Refs. 12-19). Data summarizing their results (Ref. 9) are plotted in Fig. 24. The analysis of the Peach Bottom Core 1 D13-05 fuel element represents the best analysis of in-pile cesium concentration profile performed at Gulf General Atomic. The results of the D13-05 spine analysis are also plotted in Fig. 24 (average "no gap" temperatures are used).

Also included in Fig. 24 are some unpublished values of D_{Cs} calculated by H. J. de Nordwall and L. R. Zumwalt in May 1968 from cesium diffusion profiles found in the postirradiation chemical examination of the GAIL IV fuel element by Bell, et al. (Ref. 20). The movement of control rods caused large axial variation in temperature over much of the GAIL IV fuel element. The temperature uncertainty is large for coring numbers 1, 3, 4, and 13. Therefore, these data are not plotted in Fig. 24. The D_{Cs} values obtained from the GAIL IV cesium profiles are given in Table 6.

Fig. 24. Cesium diffusion coefficients determined from fuel element concentration profile analyses

TABLE 6
DIFFUSION COEFFICIENTS OF CESIUM IN SPEER 780-S GRAPHITE (D_{Cs}) BASED ON
GAIL IV FUEL ELEMENT CESIUM PROFILES

Isotope	Coring Number	Temp Est. ($^{\circ}$ C)	C_o (mg Cs/g C)	D_{Cs}
137	1	640	6.2×10^{-6}	2.8×10^{-10}
137	1	640	5.6×10^{-6}	1.10×10^{-9}
137	3	640	6.8×10^{-4}	3.2×10^{-10}
137	3	640	5.9×10^{-4}	8.4×10^{-10}
134	3	640	2.6×10^{-6}	4.2×10^{-10}
137	4	770	3.5×10^{-4}	3.6×10^{-10}
134	5	910	5.0×10^{-5}	1.08×10^{-9}
137	5	910	4.5×10^{-5}	1.18×10^{-9}
134	7	910	--	1.19×10^{-9}
137	7	910	--	1.31×10^{-9}
137	9	950	6.3×10^{-3}	1.12×10^{-9}
134	9	950	1.05×10^{-4}	1.38×10^{-9}
134	11	950	3.4×10^{-6}	2.4×10^{-9}
137	11	950	4.6×10^{-4}	2.4×10^{-9}
134	13	870	2.9×10^{-5}	5.8×10^{-9}
137	13	870	2.5×10^{-3}	1.7×10^{-9}

A least-squares fit was performed for data obtained from this study and from the literature and is represented by the solid line in Fig. 24. The dotted line represents a 95% confidence that the mean lies below this confidence level. The current reference values of the cesium diffusion coefficient are also plotted as a dashed line. The activation energy obtained from the slope of the least-squares line is 29.2 kcal/mole. This activation energy is very near that determined from the D13-05 data alone, i.e., 27 kcal/mole.

CONCLUSIONS

1. Experimentally determined cesium and strontium concentration profiles in graphite were fitted quite closely to curves predicted by the FIPER code for the D13-05 fuel element spine. This correlation instills confidence in the capability of the FIPER code calculations to determine fission product release from HTGR graphite for those conditions where the effect of convective flow may be neglected.
2. This analysis of the Peach Bottom Core 1 D13-05 fuel element spine yielded a cesium metal diffusion coefficient of $7 \times 10^{-9} \text{ cm}^2/\text{sec}$ at 1000°C and an activation energy of 27 kcal/mole. Cesium metal diffusion coefficients were not determined for the sleeve because the gaseous precursor component contribution to the concentration profiles could not be clearly established.
3. No change in reference cesium metal diffusion coefficient is recommended, since the current reference values lie approximately on the upper 95% confidence limit line of a compilation of all available in-pile cesium diffusion data.

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APPENDIX

DISCUSSION OF GASEOUS PRECURSORS IN THE CASE OF BACK-FLOW OF HELIUM IN PORES OF SLEEVE GRAPHITE

Consider an idealized pore of constant radius which penetrates a graphite slab of thickness L . Let x = distance, perpendicular to slab face, and s = distance along pore. Then $s = \xi x$, where ξ = tortuosity of the pore.

An idealized uniform circular pore with the applicable parameters and boundary conditions is shown in Fig. 25.

$x = 0$ corresponds to the side of the fuel element sleeve adjacent to the fuel element purge stream and fuel compacts, while $x = L$ corresponds to the outside of the sleeve which is in contact with helium coolant. The helium pressure is many orders of magnitude greater than the partial pressure of fission product gaseous precursors. Thus, gas flow is directly proportional to the helium pressure gradient.

The Peach Bottom HTGR was designed so there would be a pressure gradient of helium decreasing from $x = L$ to $x = 0$. Thus, at the steady state in a uniform pore, there will be a velocity V of helium in a direction opposite to the flow of fission product gases which are at a higher concentration $C = C_0$ at $x = 0$ than at $x = L$ ($C = C_L$), since their source is the fuel compacts. Test and activity levels actually found in the Peach Bottom HTGR coolant stream verified that helium back-flow is highly effective in reducing krypton and xenon diffusion through the fuel element sleeve (Refs. 21,22).

The equation for diffusion of gaseous precursors in a given pore is

$$-\text{div } J - \lambda C = \frac{\partial C}{\partial t} \quad , \quad (1)$$

Fig. 25. Idealized uniform circular pore

where the current of fission product in the pore is

$$J = -D_g \frac{\partial C}{\partial s} - VC \quad . \quad (2)$$

The quantities s , V , C are as defined above and D_g is the diffusion coefficient of the gas in the pore. It is assumed that gas phase diffusion is not limited by the diameter of the pore and that flow through the pore is laminar.

Substituting Eq. 2 into Eq. 1 yields the diffusion equation

$$D_g \frac{\partial^2 C}{\partial s^2} + V \frac{\partial C}{\partial s} - \lambda C = \frac{\partial C}{\partial t} \quad .$$

For the steady state,

$$D_g \frac{d^2 C}{ds^2} + V \frac{dC}{ds} - \lambda C = 0 \quad . \quad (3)$$

To solve this, one finds the roots of the auxiliary equation

$$D_g p^2 + Vp - \lambda = 0 \quad , \quad (4)$$

which are

$$p_1 = - \left[\frac{V}{2D_g} + \frac{1}{2} \sqrt{\left(\frac{V}{D_g} \right)^2 + 4 \frac{\lambda}{D_g}} \right] \quad , \quad (5a)$$

$$p_2 = - \left[\frac{V}{2D_g} - \frac{1}{2} \sqrt{\left(\frac{V}{D_g} \right)^2 + 4 \frac{\lambda}{D_g}} \right] \quad . \quad (5b)$$

The solution is

$$C = K_1 \exp(p_1 s) + K_2 \exp(p_2 s) \quad , \quad (6)$$

where K_1 and K_2 are constants determined by the boundary conditions. For the gaseous precursors of interest in Peach Bottom fuel element sleeve graphite, the parameters are such that

$$\frac{\lambda}{D_g} \ll \left(\frac{V}{D_g} \right)^2 \quad . \quad (7)$$

For this case, $P_1 \approx -V/D_g$ and $p_2 \approx 0$. After defining $C = C_o$ at $x = 0$ and $C = C_L$ at $x = L$, the concentration profile for a gaseous precursor is given by

$$\frac{C}{C_o} = e^{-(V/D_g)\xi x} + \left[\frac{C_L}{C_o} - e^{-(V/D_g)\xi L} \right] \left[\frac{1 - e^{-(V/D_g)\xi x}}{1 - e^{-(V/D_g)\xi L}} \right] \quad (8)$$

Figure 26 gives concentration profiles in dimensionless units (C/C_o versus x/L). It may be seen from Eq. 8 that, if $C_L \ll C_o$ and $(V/D_g) \xi L \geq 3$, the concentration profile is given to a good approximation by

$$C/C_o = e^{-(V/D_g)\xi x} \quad . \quad (9)$$

This equation applies to the profiles discussed in the text. However, it is to be noted that in the sleeve graphite there will be a distribution of pore sizes. Thus, an observed profile, if due to the decay of precursors, will be a superposition of profiles from pores of various size.

To justify the statement made above that

$$\frac{\lambda}{D_g} \ll \left(\frac{V}{D_g} \right)^2 \quad , \quad (7)$$

the value of V/D_g must be estimated from the data given in Refs. 21 and 22. It is assumed that the idealized uniform circular pore model of Fig. 25 applies.

Fig. 26. Concentration profile of gaseous precursor as a function of back-flow velocity

Differentiating and substituting Eq. 8 into Eq. 2 gives

$$J = \frac{C_o V}{\exp\left(\frac{V}{D_g}\right) \xi L - 1} \quad , \quad (10)$$

when $C_L \ll C_o$. Equation 10 represents the current of diffusing precursor gas atoms through a pore when there is back-flow of helium gas in the pore.

The transport coefficient for the diffusing gas molecules for these conditions is defined as

$$K = \frac{J \xi L}{C_o} = \frac{V \xi L}{\exp\left(\frac{V}{D_g}\right) \xi L - 1} \quad . \quad (11)$$

(This transport coefficient is called the "back diffusion" coefficient by Riedinger (Ref. 21)). The limit as $V \rightarrow 0$ in Eq. 11 is $K = D_g$. The current ratio is therefore

$$\frac{J(V)}{J(0)} = \frac{K}{D_g} = \frac{\frac{V}{D_g} \xi L}{\exp\left(\frac{V}{D_g}\right) \xi L - 1} \quad . \quad (12)$$

In a porous material, the K and D_g are reduced since the pores occupy only a fraction of the total area. However, the reduction in K and D_g is in the same proportion to give K_{eff} and D_{eff} . The variables measured in the laboratory are K_{eff} and D_{eff} . Thus,

$$\frac{K_{eff}}{D_{eff}} = \frac{K}{D_g} = \frac{y}{e^y - 1} \quad , \quad (12')$$

where $y = (V/D_g) \xi L$.

Next calculate y , taking $K_{\text{eff}} = 7 \times 10^{-7} \text{ cm}^2/\text{sec}$ as given in Ref. 22 for xenon release into the Peach Bottom primary coolant. The value of D_{eff} is $1.6 \times 10^{-5} \text{ cm}^2/\text{sec}$ at 300°K and was obtained from Fig. 9 of Ref. 21. For xenon at Peach Bottom operating conditions of 23.8 atm of helium pressure and a typical sleeve temperature of 1000°K ,

$$D_{\text{eff}} = \frac{1.6 \times 10^{-5}}{23.8} \frac{1000}{300} = 5.8 \times 10^{-6} \quad .$$

Thus, at reactor conditions

$$\frac{K_{\text{eff}}}{D_{\text{eff}}} = \frac{7 \times 10^{-7}}{5.8 \times 10^{-6}} = \frac{1}{8.35} \quad .$$

Substituting into Eq. 12' yields

$$\frac{e^y - 1}{y} = 8.35$$

and

$$y = \frac{V}{D_g} \xi L = 3.37 \quad .$$

The thickness of the Peach Bottom sleeve is $L = 0.93 \text{ cm}$. The tortuosity of the sleeve graphite is assumed to be 3; i.e., $\xi = 3$. Sometimes higher tortuosities, $\xi = 10$, are estimated, but these include the effect of blind-ended pores and have questionable physical reality. For a bed of pack spheres, $\xi = \sqrt{2}$. Substituting these values gives

$$\frac{V}{D_g} = \frac{3.37}{3 (0.93)} = 1.21 \quad .$$

Now compare the value of V/D_g to λ/D_g . The decay constant for Xe-137 is $\lambda = 4.1 \times 10^{-3} \text{ sec}^{-1}$. For xenon diffusing in helium at 1000°K and 350

psia,* $D_g = 0.175 \text{ cm}^2/\text{sec}$. The decay constant of Xe-133, the precursor of Cs-134, is even smaller. Thus, $\lambda/D_g = 0.0234$ and $\lambda/D_g \ll V/D_g$. It is concluded that Eq. 8 is an excellent approximation to the solution of the steady-state transport equation (Eq. 5) and the concentration profile is independent of half-life.

For a given pressure gradient,

$$V \propto \frac{1}{\mu} \quad , \quad (13)$$

where μ is the helium viscosity.

Now (Ref. 23),

$$\mu \propto T^{0.66} \approx T^{0.7} \quad , \quad (14)$$

and approximately

$$D_g \propto T^{1.8} \quad . \quad (15)$$

Thus,

$$\frac{V}{D_g} \propto \frac{1}{T}^{2.5} \quad . \quad (16)$$

Accordingly, in a graphite sleeve where $C/C_0 \exp[-(V/D_g) \xi x]$, the slope of $\log C/C_0$ versus x should vary approximately inversely as the 2.5 power of absolute temperature, assuming the helium pressure drop across the sleeve remains constant.

*Calculated from formula 2.37, p. 25, Ref. 23.

Substituting into Eq. 8 gives the following cesium concentration profile resulting from the xenon precursor at 1000°K:

$$\frac{C}{C_o} = 1.036 e^{-3.62x} - 0.036 ,$$

where x is thickness (in centimeters) and

$$\frac{V}{D_g} \xi L = 3.37 \quad \text{or} \quad \frac{V}{D_g} \xi = 3.62 \text{ cm}^{-1} .$$

For the average temperature of sleeve position 2, T = 753°K,

$$\frac{V}{D_g} \xi = 3.62 \left(\frac{1000}{753} \right)^{2.5} = 7.35 ,$$

$$\frac{V}{D_g} \xi L = 6.84 .$$

Then the concentration profile due to the xenon precursor is

$$\frac{C}{C_o} = (1.0011) e^{-7.39x} - 0.0011 ,$$

$$\frac{C}{C_o} \approx e^{-7.35x} .$$

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