



Measurement of cesium diffusion coefficients in graphite IG-110



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HIGHLIGHTS

- A method was developed for real time analysis of fission product diffusion in graphite by ICP-MS.
- The design simulates HTGR conditions.
- Diffusion coefficients for cesium in IG-110 graphite were obtained.

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ABSTRACT

An understanding of the transport of fission products in High Temperature Gas-Cooled Reactors (HTGRs) is needed for operational safety as well as source term estimations. We have measured diffusion coefficients of Cs in IG-110 by using the release method, wherein we infused small graphite spheres with Cs and measured the release rates using ICP-MS. Diffusion behavior was investigated in the temperature range of 1100–1300 K. We have obtained: $D_{Cs} = (1.0 \times 10^{-7} \text{ m}^2/\text{s}) \exp\left(\frac{-1.1 \times 10^5 \text{ J/mol}}{RT}\right)$ and, compared our results with those available in the literature.

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1. Introduction

High Temperature Gas-Cooled Reactors (HTGRs) have five barriers to fission product (FP) release (the TRISO fuel coating, the fuel elements, core graphite, primary coolant system, and the reactor building). While very substantial understandings and data already exist [1–3], there is a need for new data and computational tools as new types of nuclear graphite are being used, or will be used than in the past. Our purpose in this paper is to describe a method we have developed to obtain diffusion coefficients of Cs in IG-110 graphite at HTGR temperatures. The method uses inductively coupled plasma-mass spectrometry (ICP-MS) to measure Cs release rates from graphite spheres infused with Cs. The method is general, and can be applied to measurements of diffusion coefficients of other substances or mixtures of substances as well.

We note that the release and profile methods were used previously for measurements of diffusion coefficients of Cs and other FP diffusants in graphite [4–11]. In these works graphite samples

were impregnated with the FP material, and then annealed at specified temperatures and for certain time periods to effect release of FP. These techniques involved radioanalytical measurements of initial and final FP concentration in the graphite sample (release method), or the concentration profile by sectioning of the sample (profile method). The diffusion coefficients were then extracted from the data by comparison with theoretical results as given by the diffusion equation. Additionally, in one case [7] the release rates of Cs-134 from a sphere were measured using gamma spectrometry, and the diffusion coefficient was extracted by comparing this rate to the theoretical expression for the release rate. Results for H-451, matrix graphite A-3, HS-1-1, and IG-110 that have been reported in the literature are summarized in Table 1.

2. Materials and methods

2.1. The release model and method

One considers a sample impregnated with a FP or any other volatile. As the sample is heated to a higher temperature, the FP is released from the sample at a time dependent rate. This release rate is dependent on many parameters as the sample may be porous, fractured, contain trapping contaminants, and the initial

Abbreviations: HTGR, High Temperature Gas-Cooled Reactor; ICP-MS, inductively coupled plasma-mass spectrometry; INAA, instrumental neutron activation analysis; FP, fission product.

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Table 1
Selected previous measurements of Cs diffusion in various graphites.

Investigators	Graphite	Method	$D = D_0 \exp(-E/RT)$		D ($\text{m}^2 \text{s}^{-1}$)
			D_0 ($\text{m}^2 \text{s}^{-1}$)	E (J mole $^{-1}$)	
Meyers and Hill [6]	H-451, cylindrical samples	Used sectioning technique. Diffusion trapping included; data obtained between ≈ 850 to 950 °C			2.20×10^{-10} to 1.31×10^{-9}
Leyers [7]	Matrix A3, spherical samples	Helium flow over 1 cm sphere infused with Cs-134; gamma activity measurement for release rate	2.01×10^{-4}	1.98×10^5	
Hoinkis [8]	Matrix A3-3	Doping with Cs-137 and release	1.99×10^{-4}	1.81×10^5	
Evans et al. [9]	HS-1-1	Sectioning technique	4.44×10^{-2}	1.27×10^5	
Hayashi and Fukuda [10]	IG-110 graphite	Sectioning of diffusion couple acceptor specimen	1.2×10^{-4}	1.12×10^5	
			(Series A) 1.7×10^{-4}	9.5×10^4	
			(Series B) 9.0×10^{-6}	1.57×10^5	
Hayashi et al. [11]	Irradiated IG-110 graphite	Sectioning of IG-110 specimen from OGL-1 fuel irradiation experiments			

distribution of the FP itself may not be uniform or precisely known. The sample shape may deviate from an ideal geometry, and because of flow of helium or some other gas over the sample, surface mass transfer may not be easily estimated. While all these factors need to be eventually considered, ideally, one assumes that the release rate is dominated by the diffusion equation, and in this instance the effect of other factors is small. If a mathematical expression for the release rate, (dependent on the diffusion coefficient and known sample dimensions) and hence the cumulative release over a certain time period, are obtained, then by comparing the expressions for rate or cumulative release with the data, one can obtain the diffusion coefficient. The case of the spherical geometry is especially simple, where:

$$\frac{\partial C(r,t)}{\partial t} = \frac{1}{r^2} \frac{\partial}{\partial r} \left(D r^2 \frac{\partial C(r,t)}{\partial r} \right) \quad (1)$$

With the initial and boundary conditions:

$$C(r, 0) = C_0 \quad (2)$$

$$C(0, t) = \text{finite} \quad (3)$$

$$C(R, t) = 0 \quad (4)$$

Here, $C(r,t)$ is the concentration of the FP (g/m^3), D is the diffusion coefficient (m^2/s), r is the radial coordinate (m), R is the radius of the sphere (m), and t is the time (s). C_0 is a constant; the initial uniform concentration.

This is a well-known equation of mass transfer, and can be solved by series expansion or transform techniques [12]. In particular, the cumulative fractional release (defined as the ratio of total mass release of FP from the sphere at time t to the initial total mass of FP in the sphere), can be expressed as:

$$F(t) = 1 - \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} e^{-\left(\frac{n\pi R}{R}\right)^2 Dt} \quad (5)$$

This series converges slowly, especially for short times. For extraction of the diffusion coefficient, a short time solution (obtained through the Laplace Transformation) is more convenient. This solution is known to be:

$$F(t) = 6 \sqrt{\frac{Dt}{\pi R^2}} - 3 \frac{Dt}{R^2} \quad (6)$$

This solution is considered to be valid for a fairly long range of time (up to and exceeding 90% release), and generally for short times (up to 30% release) one can use just the first term only [12]. The diffusion coefficient is calculated by fitting solutions of the fractional release equation above to fractional release data. We can also use the theoretical rate equation, which is the product

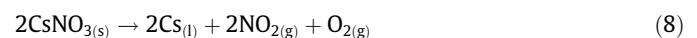
of the derivative of the above and the initial FP mass, for comparisons with the corresponding measurements. The short time theoretical release rate $Z_R(t)$ (g/s) is:

$$Z_R(t) = 3m_0 \left(\sqrt{\frac{D}{\pi R^2 t}} - \frac{D}{R^2} \right) \quad (7)$$

where m_0 is the total initial mass (g) of FP in the sphere. We have used both the cumulative release and the release rate data and expressions for calculation of diffusion coefficients and have observed agreement among the obtained values.

2.2. Materials and sample preparation

The graphite used in this study was IG-110 manufactured by Toyo Tanso. It is produced using an isostatic rubber press process and is semi-isotropic. Spherical samples were milled to a radius of 0.2 cm. The spheres were infused (impregnated) with cesium using a modified procedure that was adapted from Hayashi and Fukuda [10]. Ten graphite spheres were loaded into a quartz vial (0.6 cm diameter, 4 cm length) with 600 μg of Cs in the form of CsNO_3 . The vial was then sealed under vacuum with a measured pressure of 40 m Torr. The sealed quartz vial was heated to 500 °C to convert the nitrate salt to elemental Cs by the following reaction:



After 1 h, the temperature was increased to 1100 °C and maintained for 99 additional hours to uniformly distribute cesium within the graphite spheres. The temperature was reduced at a rate of 1 °C/min until the oven temperature was 200 °C. The spheres were removed from the vial and reduced to a final radius of 0.15 cm using SiC sandpaper. The purpose of reducing the radius from 0.2 cm to 0.15 cm was to remove any Cs which may have condensed onto the spheres during the cooling period in the oven.

The initial mass of Cs in each sphere was measured using instrumental neutron activation analysis (INAA). Comparator standards were prepared from a certified solution of CsNO_3 purchased from High Purity standards. A 50 μL aliquot of the standard solution was pipetted onto filter placed paper in a high density polyethylene vial with a volume of 200 μL . The standards were dried and capped with friction fit caps. The graphite spheres and comparator Cs standards were irradiated in the row 2 pneumatic tube irradiation position for 30 s in a neutron flux of 5.0×10^{13} n/cm 2 /s. The 127.5 keV gamma ray from decay of $^{134\text{m}}\text{Cs}$ produced by the reaction $^{133}\text{Cs}(n,\gamma)^{134\text{m}}\text{Cs}$ was measured by counting the sample 2.5 cm from the face of a HPGe detector. The samples were counted

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