EXPERIMENTS ON THE CONCENTRATION DEPENDENCE OF STRONTIUM DIFFUSION IN GRAPHITE

L. R. Zumwalt and P. T. Williams
North Carolina State University

A number of studies indicate volatile metals such as cesium, barium and strontium show concentration dependent diffusion in graphite (1, 2, 3). In connection with a study of the effect of strontium concentration on cesium sorption in H-451 nuclear grade graphite, the diffusion of Sr-85 traces was studied in samples preloaded with ordinary stable strontium at several selected concentrations. The experimental technique involved the determination of tracer activity profiles in rod-shaped samples after they had been annealed in contact with a source disk of the same diameter. The method is similar to that of the classical study of "self-diffusion" in metals where the diffusion is at a constant chemical potential.

The source disk and sample rods, of H-451 graphite, were of diameter, 0.794 cm, and of lengths $L = 0.1$ cm and $S = 1.9$ cm respectively. Strontium was originally added as the nitrate dissolved in a water-acetone mixture. After a series of processes of impregnation, evaporation, baking and finally annealing at 1400°C in a graphite tube (King) furnace uniform impregnations of stable strontium at nominal concentrations of 10,100,300 and 1000 μg Sr/g C were obtained. The source disks were then further impregnated with strontium-85 (and annealed) which added 18 ± 4 μg Sr-85 to the source disk depending on the experiment. The manipulations involving the impregnated and annealed strontium-graphite samples were carried out in a dry, inert gas (argon) atmosphere. After adding the Sr-85, the source disk and rod sample faces, which had been lapped to a good fit before the impregnations, were put together and placed inside a molybdenum container provided with a tantalum spring which pressed the graphite source-rod couple together in an axial direction through pressure applied by the screw-top closure of the container. This assembly was transferred to the King Furnace and then diffusion runs were carried out at 1300°C for two hours in an argon atmosphere.

After the diffusion runs the source disk was counted to determine the amount (using a calibrated reference source) of strontium-85 remaining in the disk, C_s. Successive axial sections of the graphite rod were taken using the lathe and a vacuum collection rig whereby the graphite cuttings of each section were collected on a 9 mm diameter filter paper. These papers were folded, weighed and counted to determine the amount of Sr-85 and mass of graphite in each section. In this manner strontium-85 concentration profiles were obtained. These were integrated to find the average concentration $\bar{C}$. From a plot of Sr-85 concentration $C(x)$ versus distance along the rod axis (x), the extrapolated boundary concentration $C(0)$ was obtained from a curve fit so as to avoid any perturbations, which occurred in some cases, of concentrations for sections taken very near the surface $(x \approx 0)$.

Plots of $C(x)/C(0)$ vs x, i.e. normalized concentration profiles, all for 2.0 hour diffusion anneals at $1300 \pm 10^\circ$C but for different initial preloaded strontium concentrations are given in Figure 1. The profile curves which are the result of an "eyeball fit" of the data points show the data to have little scatter and indicate a monatonically increasing rate of diffusion with increasing strontium concentration.

To obtain a first approximation of the diffusion coefficients it was assumed that the first portion of the curves would approximately correspond to the diffusion equation solution for constant Sr-85 concentration at $x = 0$. Then at time t at the end of the anneal

$$C(x)/C(0) = \text{erfc} \; (x/2\sqrt{Dt}) \tag{1}$$

Figure 2 is a plot of the diffusion coefficients, D_{Sr} (obtained from a curve fit using Equation 1) versus the approximately uniform total strontium concentration of the graphite-rods.

The plot shows the diffusion coefficient to vary approximately as the square-root of concentration. The empirical equation corresponding to the straight line fit is,

$$D \left(\frac{cm^2}{sec}\right) = 1.3 \times 10^{-6} \left[C \left(\frac{\mu gSr}{gc}\right)\right]^{0.47}$$

This relationship is substantially different than the variation of D with strontium concentration reported by Sandalls and Walford (1) for AGL-9 graphite at 1000°C. They give

$$D \left(\frac{cm^2}{sec}\right) = 4.42 \times 10^{-16} \left[C \left(\frac{\mu gSr}{g}\right)\right]^{3.15}$$

where C must exceed 62 μg/g.

Once the necessary computer programing is complete it is planned to revaluate the diffusion coefficients obtained in our experiments by a more precise analysis which takes account of the depletion of the source and employs a boundary condition of the third kind which takes into account the resistance to mass transfer due to the small gap at the source disk - graphite rod interface.

The solution, obtained using the method of the integral transform (4) is

$$C(x)/C_{so} = \frac{L}{S+L} + H_1 \sum_{m=1}^{\infty} K\left(\beta m,0\right) K\left(\beta m,x\right) \frac{S}{S+L} \frac{\exp(-kt)}{\beta m^2 - k/D}$$
$$- H_1 \sum_{m=1}^{\infty} K\left(\beta m,0\right) K\left(\beta m,x\right) \exp\left(-\beta m^2 Dt\right) \left[\frac{L}{(S+L)\beta m^2} + \frac{S}{(S+L)(\beta m^2 - k/D)} \right] \tag{2}$$

where

$$K\left(\beta m,x\right) = \sqrt{2} \left[\frac{\beta m^2 + H_1^2}{S(\beta m^2 + H_1^2) + H_1} \right] \cos \beta m(S-x) \tag{3}$$

βm are roots of $\beta \tan (\beta S) = H_1$ \hfill (4)

$$H_1 = \frac{-\left(\frac{dC}{dx}\right)_{x=0}}{C_s - C(0)} \tag{5}$$

The concentration of its source disk at time t
is,

$$C_s = C_{so} \left[\frac{S}{S+L}\right] \exp\left(-kt\right) + C_{so} \left[\frac{L}{S+L}\right], \qquad (6)$$

where

C_{so} = the initial source concentration.

All boundaries, excepting the source-graphite inter-
face, are taken as of the second kind - i.e.
$dC/dx = 0$.

References

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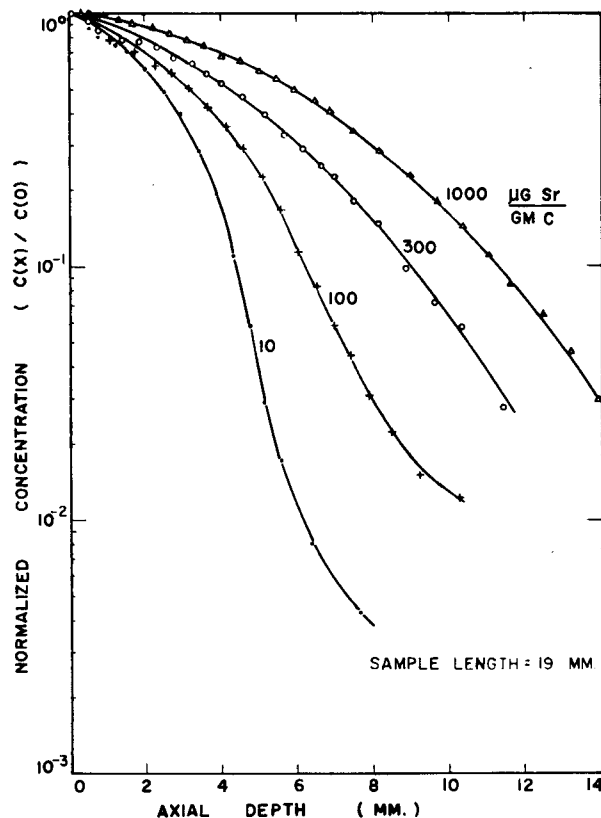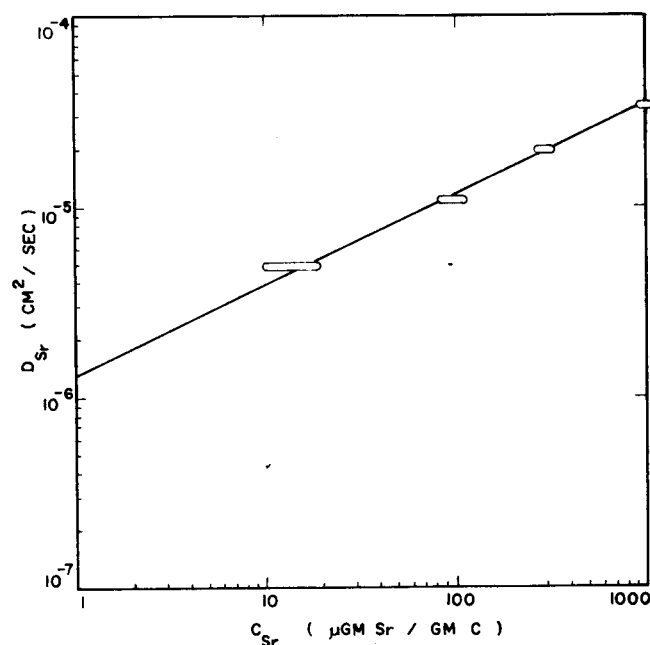   ton, Pa. (1968).

Figure 1. Normalized Concentration Profiles

Figure 2. Diffusion Coefficient vs. Concentration

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