Thermochimica Acta, 180 (1991) 139–145
Elsevier Science Publishers B.V., Amsterdam

THE EFFECT OF TEMPERATURE ON DIFFUSION OF IODINE THROUGH TOPOPAH SPRING TUFF.

Part II

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(Received 21 August 1990)

ABSTRACT

The effective diffusivity of iodine through the lower non-lithophysal zone of the Topopah Spring Tuff (outcrop sample of the proposed nuclear repository site layer) was determined using an unsteady state method. For the non-lithophysal zone of the Topopah Spring Tuff, results showed that the effective diffusivity of iodine through non-lithophysal zone tuff increased with temperature. The following correlation was obtained to estimate the effective diffusivity of iodine as a function of temperature

$D_e = 1.8821 \times 10^{-3} - 1.2918 \times 10^{-5} T + 2.1039 \times 10^{-8} T^2$

LIST OF SYMBOLS

C_{I_2}	concentration of iodine in the helium and iodine gas mixture (g mol cm^{-3})
D_e	effective diffusivity of iodine (cm^2 s^{-1})
Δr	pellet thickness (cm)
T	absolute temperature (K)
t	time (s)

Greek letters

μ_1	first moment, residence time (s)
ϵ	porosity of tuff samples
ρ_b	bulk density of tuff samples (g cm^{-3})
ρ_s	skeletal or true density of tuff samples (g cm^{-3})

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INTRODUCTION

Spent fuel from power reactors is currently considered to be the most significant commercial high-level nuclear waste-form in the U.S.A. The long-lived radionuclides that could enter the vapor phase at the spent-fuel storage temperatures are ^{14}C (in CO_2, CO, or CH_4) and ^{129}I. Both ^{14}C and ^{129}I have long half-lives. The half-life of ^{14}C is 5730 years [1] and the half-life of ^{129}I is 16 million years [2].

The Topopah Spring Member of the Paintbrush Tuff in Yucca Mountain, Neveda is a prime candidate for the site of the first national high-level radioactive waste repository. Spent fuel from power reactors is currently considered to be the most significant commercial high-level nuclear waste-form. According to Van Konynenburg et al. [3], radionuclides with long half-lives will have significant remaining activities after a 300–1000 year containment period. The long-lived radionuclides that could enter the vapor phase at spent-fuel storage temperatures are ^{14}C and ^{129}I. According to the annotated outline for site characterization plans [4], the diffusion of radionuclides into the rock matrix, including diffusion into pore spaces, must also be addressed.

Scientists have been conducting research on various aspects and conditions for design and operation of a safe, high-level waste repository. One of the questions to be answered in characterizing the site is the extent of the diffusion of radioactive gases, such as carbon dioxide and iodine, to the accessible environment. The objectives of this study were to measure the effect of temperature on the effective diffusivity of iodine through Topopah Spring Tuff. There are no data available on the effect of temperature on iodine gas diffusivity through volcanic rock tuff, except that of Bardakci et al. [5] on “G” Tunnel tuff. Bardakci et al. [5] found that the effective diffusivity of iodine through “G” Tunnel tuff is 0.0011 at 480 K and this increased when the temperature increased.

The samples from the same outcrop of the lower non-lithophysal zone of the Topopah Spring Tuff described in Part I of this publication, were used in this study.

EXPERIMENTAL

In this method, a porous solid was brought into contact with an adsorbable gas and the unsteady rate of desorption was measured. Wang and Smith [6], Dogu and Smith [7], and Dogu and Ercan [8] have used a single dynamic pulse-response technique to measure diffusivities of gases though porous solids. When the unsteady state method is used, indirect calculations are required to determine the effective diffusivity of gases. This method was chosen over the steady state method to minimize the amount of iodine gas to be handled. Iodine is a solid element at ambient conditions with a melting point of 386.65 K and a boiling point of 457.50 K. Upon heating at ambient pressure, solid iodine sublimes to the vapor phase. Since we planned to measure the effective diffusivity of iodine gas, and we did not want any deposition of the iodine gas, the experiments were carried out between 473 and 573 K. But the system can be operated between 298 and 1173 K for the other gas systems. The stainless steel lines that the iodine contacted were heated using heating tape to avoid sublimation of iodine gas. Both of the exit streams were extended into the hood. In the unsteady state method, a porous solid is brought into contact with an absorbable gas and the unsteady rate of desorption is measured. When the unsteady state method is used, indirect calculations are required to determine the effective diffusivity of gases. The equipment used in the steady state technique which was described in Part I of this publication, was modified to include thermal conductivity cells to give a continuous analysis of each gas stream, as shown in Fig. 1. This modified system was used to measure the effective diffusivity of iodine gas through the non-lithophysal zone tuff. The pellet was mounted in the Bardakci diffusion cell as described in Part I, and helium gas was passed radially across each side of the pellet. The flow rates were kept at about 700 $\mathrm{cm}^3\ \mathrm{min}^{-1}$. Pure helium gas, at the same flow rate, was passed through the reference cell of each thermal conductivity cell. Iodine gas was produced by heating the solid iodine crystals contained in a beaker heated in a hot water bath, as shown in Fig. 2. A pulse of pure iodine vapor was obtained by injecting about 5 ml of iodine gas into the helium gas inlet stream on one side of the diffusion cell. The thermal conductivity cells were used to measure the concentration of iodine gas as a function of time in the opposite exit stream. According to Smith [9], the effective diffusivity of iodine was then calculated from the equation

$$D_\mathrm{e} = \Delta r^2 \epsilon / (6\mu_1) \tag{1}$$

Here μ_1 is the first moment (retention time) of the diffusing component, iodine, in the pellet. The first moment of the iodine is obtained from the measured response pulse of iodine in the helium gas stream at the opposite side of the diffusion cell

$$\mu_1 = \frac{\int_0^\infty C_{\mathrm{I}_2}(t)t\,\mathrm{d}t}{\int_0^\infty C_{\mathrm{I}_2}(t)\,\mathrm{d}t} \tag{2}$$

The first moment should be corrected by subtracting the retention time of iodine in its injection side and in the opposite side of the pellet, travelling in helium. In addition, the first moment of the injection time should also be subtracted.

The bulk density of the samples were determined from the volume and

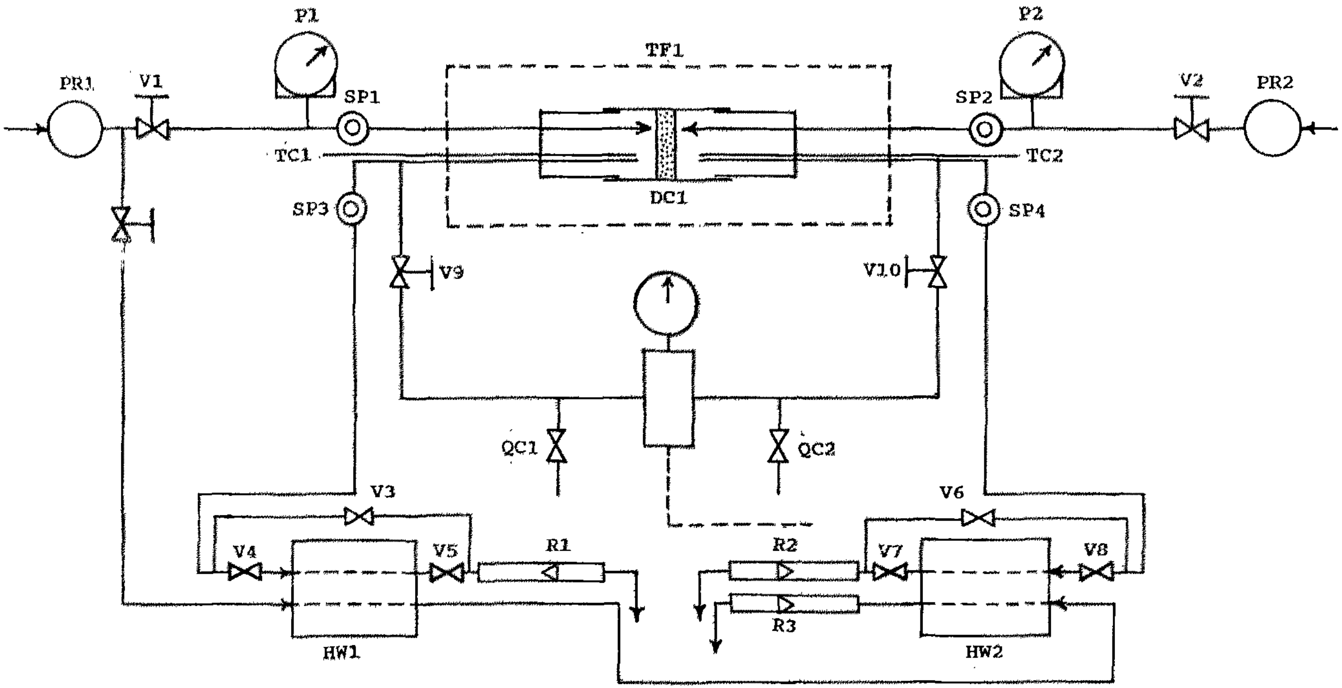

Fig. 1. Flowchart for the unsteady state diffusion measurement system: PR, pressure regulator; V, hand valve; TC, thermocouple; TF, tubular furnace; DC, diffusion cell; SP, sampling ports (septums); QC, quick connects; R, rotameter; DP, differential pressure cell and indicator; P1 and P2, pressure gauges; HW, hot wire detector, thermal conductivity cell.

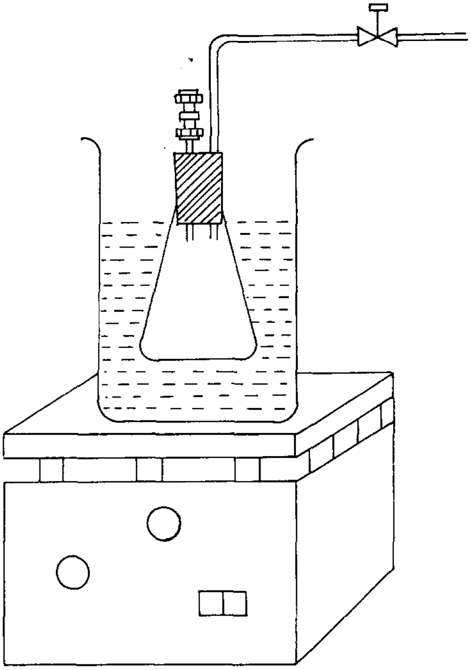

Fig. 2. Iodine gas production unit placed on a hot plate.

the weight of the dry samples. Since the pellets were cylindrical, it was easy to obtain an accurate volume. The true or skeletal density of the volcanic rock was obtained using a Multypycnometer manufactured by Quantachrome Corporation, NY. Then eqn. (3) was used to calculate the porosity

$$\epsilon = 1.0 - \rho_b/\rho_s \tag{3}$$

This porosity value, the corrected first moment of desorption, and the thickness of the pellet was used in eqn. (1) to calculate the effective diffusivity of iodine through the outcrop sample of the lower non-lithophysal zone of the Topopah Spring Tuff.

RESULTS AND DISCUSSION

The effective diffusivity of iodine through the outcrop sample of the lower non-lithophysal zone of the Topopah Spring Tuff was determined as a function of temperature as shown in Fig. 3. The experiments were carried out with two separate sample pellets. The results are very close to each other. For the dry non-lithophysal zone samples, the following correlation was obtained to estimate the effective diffusivity of carbon dioxide as a function of temperature

$$D_e = 1.8821 \times 10^{-3} - 1.2918 \times 10^{-5}T + 2.1039 \times 10^{-8}T^2 \tag{4}$$

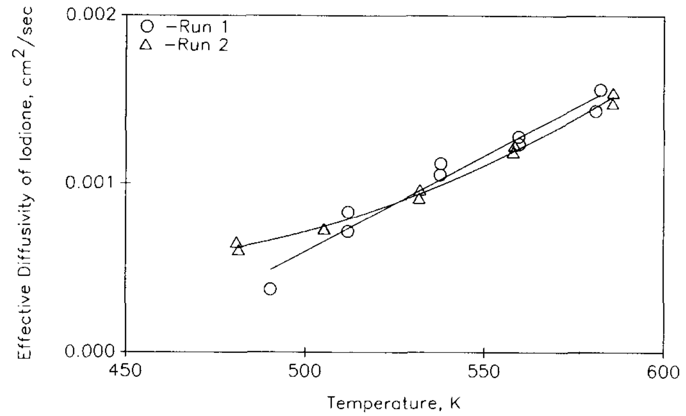

Fig. 3. Effective diffusivity of iodine through the lower non-lithophysal zone of the Topopah Spring Tuff as a function of temperature.

The bulk densities for the samples 1 and 2 were 2.3306 and 2.3375 g cm^{-3} and the skeletal densities of the samples were 2.7805 and 2.7804 g cm^{-3} respectively. Then, using eqn. (3), the porosities of the samples 1 and 2 were found to be 0.162 and 0.159, respectively. These porosities were utilized in eqn. (1) in calculations of the effective diffusivities of iodine. According to Evans et al. [10], the ratio of the effective diffusivity of the radioactive iodine (with iodine-129) to effective diffusivity of iodine (with iodine-127) is equal to the square root of the ratio of the molecular weight of iodine gas (with iodine-127) to the molecular weight of iodine gas (with iodine-129).

CONCLUSIONS

The following conclusions can be drawn from this study.

1. The unsteady state diffusion system can be used to determine the effective diffusivity of iodine through tuff samples.
2. The effective diffusivity of iodine through all the tuff samples investigated increased with temperature.
3. For the dry non-lithophysal zone of the Topopah Spring samples, the following correlation was obtained to estimate the effective diffusivity of iodine as a function of temperature

$$D_e = 1.8821 \times 10^{-3} - 1.2918 \times 10^{-5} T + 2.1039 \times 10^{-8} T^2$$

ACKNOWLEDGMENT

The author gratefully acknowledges the financial support for this work received from the Oak Ridge Associated Universities, U.S. Department of

Energy, Office of Radioactive Waste Management (Contract Number 3C-56702). We would like to thank Ronald J. Sternfels, Virginia M. Oversby and Billie Stooksbury for their support of the project. The author thanks Ajeet Singh for his help in carrying out the experiments. The author would also like to thank Mr. Chris Lewis of Yucca Mountain Project Office, U.S. DOE, Nevada Operations Office for providing the samples from the lower non-lithophysal zone of the Topopah Spring Tuff. Partial support was provided by the National Science Foundation (Grant Number RII-8804227).

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