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Temperature and saturation effects on diffusion of carbon dioxide through "G" tunnel and Yucca Mountain tuffs

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Abstract

The effective diffusivity of carbon dioxide through various tuff samples is reviewed. The effective diffusivities of carbon dioxide through "G" tunnel tuff, the lower non-lithophysal zone of the Topopah Spring tuff (outcrop sample of proposed nuclear repository site layer), USW H-6 test well tuff, the lower lithophysal zone of the Topopah Spring tuff (outcrop sample of the layer right above the proposed repository) and Tiva Canyon tuff samples (proposed repository layer) have been evaluated. Empirical correlations were reported for each tuff to estimate the effective diffusivity of carbon dioxide through them. The effective diffusivity of carbon dioxide through all the tuff samples increased with temperature and decreased as the moisture content increased.

INTRODUCTION

The United States has many nuclear power plants in operation today, and the problem of high-level radioactive waste disposal is of national concern. The operation of a geologic repository for the permanent disposal of radioactive waste is presently under serious consideration by the United States Government in accordance with the 1982 Nuclear Waste Policy Act. Three candidates for the site of the first geologic repository included burial sites in basalt, salt, and tuff rock formations [1]. However, when the United States Congress passed the budget reconciliation act of December 1987, all site-specific research on the design and development of a high-level nuclear waste repository was restricted to the Yucca Mountain tuff site. The repository is expected to be built sometime between 1998 and 2003 if a construction authorization is given by the Nuclear Regulatory Commission.

The Topopah Spring member of the Paintbrush tuff in Yucca Mountain, Nevada, 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 form of commercial high-level nuclear waste. According to Van Konynenburg et al. [2], radionuclides with long half-lives will have significant remaining activities after a 300 to 1000 year

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containment period. The long-lived radionuclides that could enter the vapor phase at spent fuel storage temperatures are ^{14}C and ^{129}I.

Carbon-14 is produced in the primary cooling water of nuclear power plants. Part of the carbon-14 is released to the atmosphere, primarily as CH_4 and CO_2. Thomas and Brown [3] have calculated the health effects that would occur if all of the carbon-14 produced in a 400-GW(e) fuel cycle operating for 100 years were to be released to the atmosphere. They concluded that an average of six additional deaths per year would occur because of the carbon-14 release to the atmosphere.

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 diffusion of radioactive gases, such as carbon dioxide and iodine, to the accessible environment. The objective of this paper is to review all the work done on the effects of temperature and water content on the effective diffusivity of carbon dioxide through "G" tunnel and the Yucca Mountain tuffs. This study provides data which may be used to estimate the amount of carbon dioxide that will diffuse through various tuff layers. Since it would be very hard, if not impossible, to sample a mountain statistically, these data might not provide the best values for gas diffusivities through tuff layers.

REVIEW OF CARBON DIOXIDE DIFFUSION THROUGH TUFF

A steady-state diffusion method was used to determine the effective diffusivity of carbon dioxide gas through both types of tuff. The steady-state method used for the counter diffusion of gases in porous solids was developed by Wicke and Kallenbach [4] and modified by Bardakci and Gasner [5]. The use of a Bardakci diffusion cell in the steady-state diffusion method to determine the effective diffusivity of carbon dioxide is explained by Bardakci et al. [6,7] and Bardakci [8].

The tuff samples from the following five sources were investigated: (a) "G" tunnel tuff, (b) the lower non-lithophysal layer of the Topopah Spring tuff, (c) the Topopah Spring tuff from the proposed repository site, (d) the lower lithophysal zone of the Topopah Spring tuff and (e) Tiva Canyon tuff. The data reported on the respective layers will be summarized.

"G" tunnel tuff

The effects of temperature and saturation on diffusion of carbon dioxide through "G" tunnel tuff have been investigated by Bardakci et al. [6,7]. At the start, since tuff samples from the proposed repository site were not made available, first investigations were done with "G" tunnel tuff. The rock sample was obtained from a nearby location called Rainier Mesa Member Ash Flow from "G" tunnel. The effective diffusivity of carbon dioxide

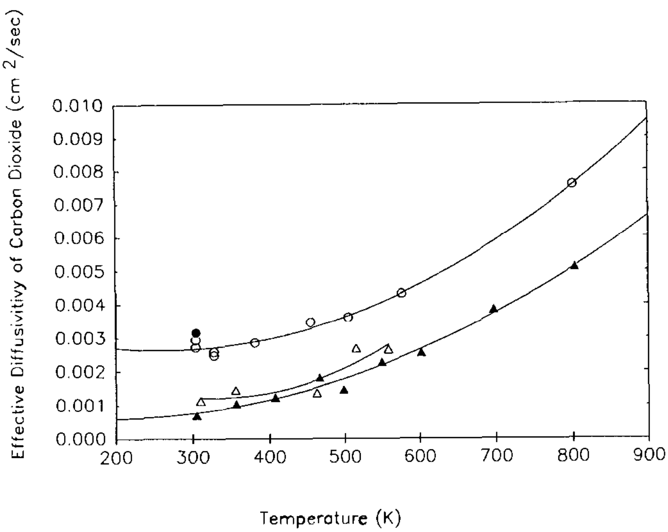

Fig. 1. Effective diffusivity of carbon dioxide through Rainier Mesa Member of the Timber Mountain tuff as a function of temperature.

through "G" tunnel tuff is given in Fig. 1. The following empirical equation was obtained using average values of the effective diffusivities at each temperature:

$$D_e = 0.003 - 1.032 \times 10^{-5}\, T + 1.768 \times 10^{-8}\, T^2 \tag{1}$$

where D_e is the effective diffusivity of carbon dioxide in $cm^2\ s^{-1}$ and T is the absolute temperature in K. The effective diffusivity of carbon dioxide through "G" tunnel tuff was also determined as a function of the average percent saturation, as shown in Fig. 2. The average percent saturation is 100 times the ratio of the average moisture content during the experiment to the

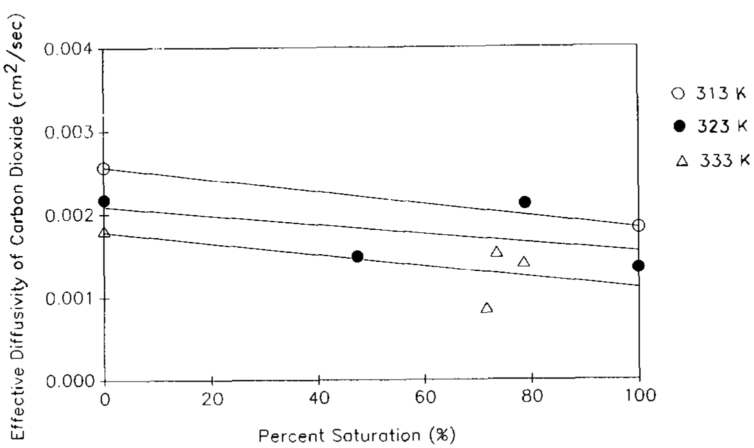

Fig. 2. Effective diffusivity of carbon dioxide through Rainier Mesa Member of the Timber Mountain tuff as a function of percent saturation.

maximum moisture content of that specific sample. One sample was used over the entire saturation range and at a fixed temperature to illustrate the effects of sample variability and of the average percent saturation. The average percent saturation was the average of percentage saturation before and after each experiment, and varied by $\pm 15\%$. When the average slope for all the samples was combined with eqn. (1), the following equation was obtained to estimate the effective diffusivity of carbon dioxide through "G" tunnel tuff as a function of temperature and percent saturation

$$D_e = 0.003 - 1.032 \times 10^{-5}\, T + 1.768 \times 10^{-8}\, T^2 - 7.107 \times 10^{-6} \quad \text{(percent saturation)} \tag{2}$$

Lower non-lithophysal layer of the Topopah Spring tuff

The samples studied by Bardakci [8] were cored from a 60 kg large rock sample of the lower non-lithophysal zone of the Topopah Spring tuff. This rock was an outcrop sample taken from the extension of the proposed repository site layer. This rock was from Busted Butte location, which is 4 miles away from the repository site and located at the south of the Yucca Mountain. As expected, the effective diffusivity of carbon dioxide through the non-lithophysal zone tuff increased with temperature as shown in Fig. 3. In Fig. 3, each run represents the experiments done with separate pellets cut from the same rock sample. Sample 1 had a porosity of 0.144. The second sample had a porosity of 0.140. The mean pore radius values of 51.1 nm and 20.1 nm were obtained for the samples 1 and 2 respectively using an Autoscan60 mercury porosimeter supplied by the Quantachrome Corporation (New York). Since sample 1 had the higher porosity and larger mean pore radius, the effective diffusivity values for that sample are slightly

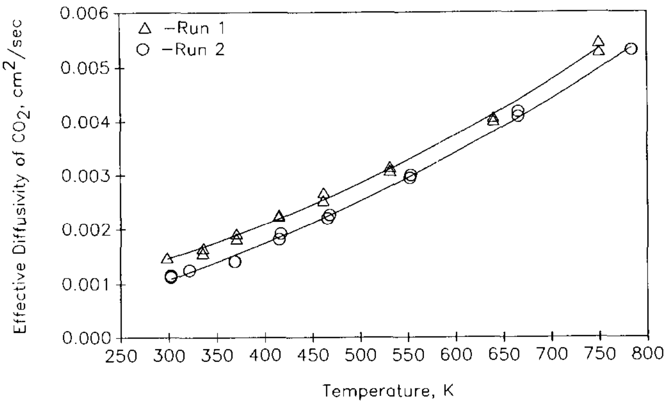

Fig. 3. Effective diffusivity of carbon dioxide through the lower non-lithophysal zone of the Topopah Spring tuff as a function of temperature.

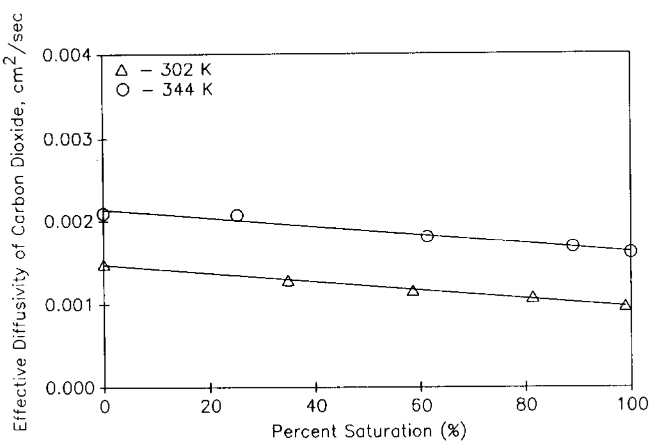

Fig. 4. Effective diffusivity of carbon dioxide through the lower non-lithophysal zone of the Topopah Spring tuff as a function of percent saturation.

higher. The specific surface areas of samples 1 and 2 were 0.87 and 0.83 m^2 g^{-1} respectively. For the dry non-lithophysal zone samples, the following empirical correlation was obtained to estimate the effective diffusivity of carbon dioxide as a function of temperature and the percent saturation.

$$D_e = -1.1578 \times 10^{-4} + 2.9885 \times 10^{-6}\, T + 5.3104 \times 10^{-9}\, T^2 \tag{3}$$

The experiments were carried out with two separate samples as a function of the average percent saturation. The effective diffusivity of carbon dioxide decreased as the moisture content of the lower non-lithophysal zone tuff increased, as shown in Fig. 4, in which only one sample was used during the run carried out at 302 K, and another sample was used during the runs at 344 K. The slopes of the lines for the first and the second samples were -5.0370×10^{-6} and -5.1108×10^{-6} $cm^2/(s \times$ percent saturation) respectively. The average slope of the two lines in Fig. 4 was -5.0739×10^{-6} $cm^2/(s \times$ percent saturation). When the saturation data were combined with eqn. (3), the following equation was obtained to estimate the effective diffusivity of carbon dioxide as a function of temperature and the percent saturation

$$D_e = -1.1578 \times 10^{-4} + 2.9885 \times 10^{-6}\, T + 5.3104 \times 10^{-9}\, T^2 - 5.0739 \times 10^{-6} \text{ (percent saturation)} \tag{4}$$

Topopah Spring tuff

Only one single Topopah Spring tuff sample of the proposed repository site layer, which had a diameter of 2.54 cm and a thickness of 2.22 mm, could be obtained [7]. The sample was taken from the USW H-6 test well between 338.08 and 338.24 m below the earth's surface.

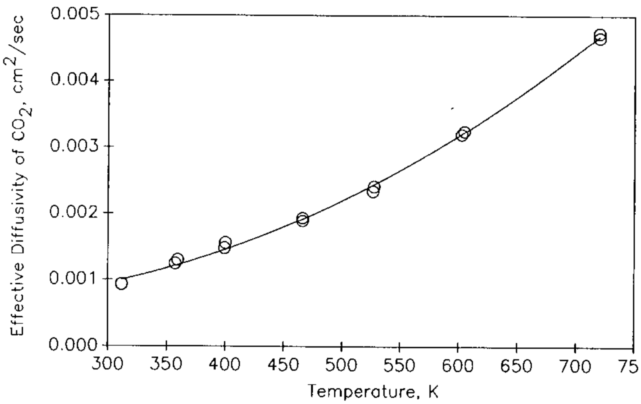

Fig. 5. Effective diffusivity of carbon dioxide through the USW H-6 test well sample of the Topopah Spring tuff as a function of temperature.

The effective diffusivity of carbon dioxide through the USW H-6 test well sample of this Topopah Spring tuff also increased with temperature, as shown in Fig. 5. For the repository layer the effective diffusivity of carbon dioxide was 0.00093 $cm^2\ s^{-1}$ at 312 K. The porosity of this sample was 0.137, which is lower than that of the non-lithophysal zone tuff. The specific surface area was higher (0.91 $m^2\ g^{-1}$) and the mean pore radius was lower (4.4 nm). The following empirical correlation was obtained to estimate the effective diffusivity of carbon dioxide through the proposed repository layer as a function of temperature. Since the effective diffusivities of outcrop samples are close to those of the USW H-6 test well samples, one can assume that the slopes of the effective diffusivity of carbon dioxide versus percent saturation lines are the same for both non-lithophysal zone tuff and USW H-6 test well tuff samples. Therefore the following empirical correlation was obtained to estimate the effective diffusivity of carbon dioxide through the proposed repository layer as a function of temperature and percent saturation

$$D_e = 9.1242 \times 10^{-4} - 3.5180 \times 10^{-6}\ T + 1.2168 \times 10^{-8}\ T^2 - 5.07390 \times 10^{-6}\ \text{(percent saturation)} \qquad (5)$$

Lower lithophysal zone of the Topopah Spring tuff

The effective diffusivity of carbon dioxide through the lower lithophysal zone of the Topopah Spring tuff (outcrop sample of the layer above the proposed nuclear repository site layer) also increased with temperature, as shown in Fig. 6. This sample is from the same location as the lower non-lithophysal zone of the Topopah Spring tuff. The porosities of samples 1 and 2 were 0.17 and 0.15 respectively. The corresponding specific surface

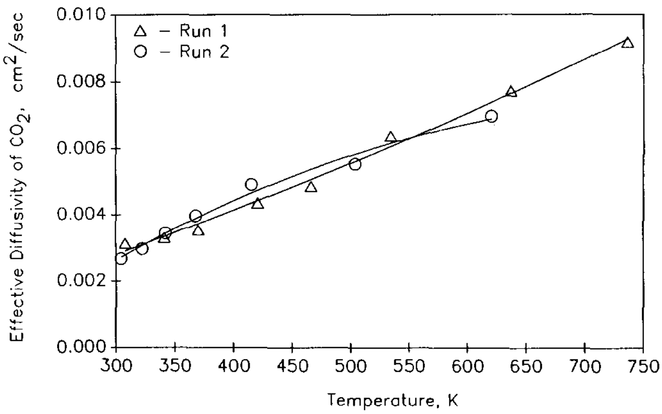

Fig. 6. Effective diffusivity of carbon dioxide through the lower lithophysal zone of the Topopah Spring tuff as a function of temperature.

areas were 1.520 and 1.477 $m^2\ g^{-1}$. For this layer the effective diffusivity of carbon dioxide was around 0.0028 $cm^2\ s^{-1}$ at 300 K. The following correlation was obtained to estimate the effective diffusivity of carbon dioxide through this lower lithophysal zone of the Topopah Spring tuff

$$D_e = -1.1119 \times 10^{-3} + 1.2512 \times 10^{-5}\, T + 1.8288 \times 10^{-9}\, T^2 \qquad (6)$$

Tiva Canyon

For the Tiva Canyon tuff, results show that the effective diffusivity of carbon dioxide is around 0.01 $cm^2\ s^{-1}$ at 300 K. As expected, the effective diffusivity through the non-lithophysal zone tuff increased with temperature, as shown in Fig. 7. The Tiva Canyon sample was taken directly above the proposed repository block called "Upper Cliff", which is a densely welded

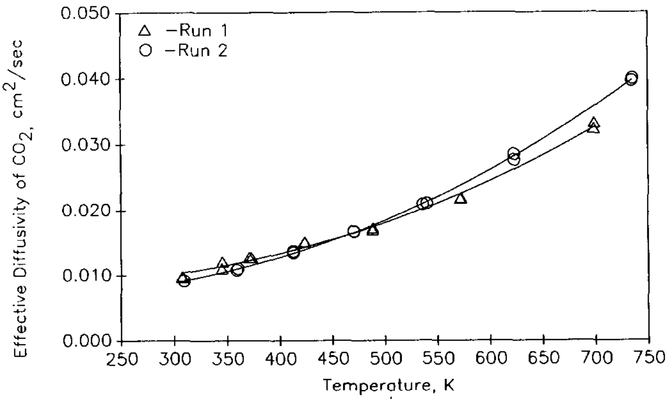

Fig. 7. Effective diffusivity of carbon dioxide through Tiva Canyon tuff as a function of temperature.

tuff. In Fig. 7, each run represents the experiments done with separate pellets cut from the same rock sample. For the Tiva Canyon tuff samples, the following correlation was obtained to estimate the effective diffusivity of carbon dioxide as a function of temperature

$$D_e = 1.2168 \times 10^{-2} - 3.7713 \times 10^{-5}\, T + 9.9510 \times 10^{-8}\, T^2 \qquad (7)$$

CONCLUSIONS

This information on the diffusion of gases through tuff may provide data to determine whether the Nuclear Regulatory Commission and Environmental Protection Agency regulations can be met. Since it is very hard, if not impossible, to sample a mountain statistically, our data may not provide the best value for diffusivity through tuff. Although it might be inappropriate to use these data to design a repository in the Topopah Spring tuff layer, the reviewed data do give an estimate of the magnitude of the carbon dioxide diffusion and how the effective diffusivity changes with temperature in the Yucca Mountain tuffs.

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