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RADON GAS DIFFUSION COEFFICIENT IN MOISTURIZED SOIL SAMPLES

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Abstract- The diffusion of Radon gas to the atmosphere has extensively been studied for earthquake prediction, active fault determination, and discovery of uranium mines. Radon concentration under the ground surface and its diffusion through the soil texture covering the fault zones are crucial for assessing Radon potential as earthquake precursor. The Radon diffusion coefficient through various moisturized soil samples using steady-state and transient methods has been studied to evaluate the effect of soil physical properties on Radon concentration in atmosphere. Undisturbed soil samples with several different moisture contents were examined. The diffusion coefficient decreased by increasing the moisture content of the soil samples with porosity of 37%. The results indicate the soil properties and its moisture content are of the essential factors that could significantly affect the anomalies of Radon gas when regarded as one of the earthquake-precursors. In addition to, the obtained diffusion coefficients were in accordance with the former studies by other researchers.

Keywords: Radon, Soil, Diffusion, Moisture, Earthquake Precursor.

I. INTRODUCTION

Radon is the second-most common cause of lung cancer after smoking, among world people. Also, Radon concentration changes is used as one of earthquake precursors. So, it is important to investigate Radon diffusion. Radon gas moves from the soil to the atmosphere by diffusion process that follows the Fick's law in porous media [1]. The major factor of the law is its diffusion coefficient that greatly depends on various factors such as temperature, moisture, porosity and effective permeability. Among them, moisture has an important effect on diffusion coefficient [2].

Two general methods have been used to determine the Radon diffusion coefficient: Steady state and transient methods. Ouffni measured the diffusion coefficients of bole, lime, and compacted sand (4.3-1.26) $\times 10^{-6}$ m^2s^{-1}, by steady state method [3]. The diffusion coefficient for sandy rocks with 0.1 $\times 10^{-2}$ m thickness was calculated to be 2.2×10^{-6} m^2s^{-1} [4].

For a type of dry soil, it has been found to be 3.2×10^{-6} m^2s^{-1} where the effect of soil compaction on the coefficient has also been studied [5]. For soils with 0.05-0.34 moisture it was estimated to be between 1.5×10^{-6} m^2s^{-1} and 3.2×10^{-6} m^2s^{-1} [2]. Fernandez et al. determined diffusion coefficient from 1.5×10^{-6} m^2s^{-1} to 3.2×10^{-6} m^2s^{-1} as a function of thickness for Radon impermeable membrane [6]. Rogers and Nielson showed that this coefficient is a function of moisture and decreases when moisture increases. It was also been theoretically found out as an exponential function in terms of moisture and porosity coefficient [7].

Tsapalov et al. determined the Radon diffusion coefficient of soil and other building materials by non-steady state method. They estimated the Radon diffusion coefficient about 4.5×10^{-9}-1.5×10^{-6} m^2s^{-1} for soil samples 6.35 cm thickness [8].

Kumar and Chauhan presented two active and passive methods to measure Radon diffusion coefficient through building materials like cement, soil, sand, etc. The diffusion coefficient from these materials varied from $(0.9\pm 0.5)\times 10^{-7}$ to $(22.9\pm 13.2)\times 10^{-6}$ m^2s^{-1} [9].

Ryzhakova suggested a mathematical method for estimating Radon diffusion. He measured Radon activity in soil at two different depths and received characteristics of soil effect on diffusion process. [10]

This study is aimed at investigating the effect of covered soil of the fault zones, in the raining seasons, on the Radon exhalation rate to the atmosphere. This objective is to be achieved through the study of the Radon diffusion coefficient in soil samples of the known fault zone using steady state and transient methods and finding out its relation with the soil moisture and permeability.

II. MATERIAL AND METHOD

A. Measuring System

An apparatus including two Teflon cylindrical chambers with volumes V_1 (small chamber) and V_2 (large chamber) and soil sample container with height d and a surface area of S enclosed between them is considered. The Radon is introduced into the small chamber and diffuses from soil sample to the large chamber.

The small chamber is connected to Radon source and the large chamber is connected to RAD7 detector that shows Radon concentration, temperature and moisture percentage simultaneously. A moisture retention material is used to prevent moisture increase in the detector, as it is moisture sensitive.

Prior to doing the measurements, the containers and gas taps were washed with dish soap, as oil and other kinds of grease solve Radon atoms and could produce important errors in the measurements. In order to determine the leakage of the large chamber, a Teflon cylinder of the same size of the sample holder was fitted between the two chambers and the small chamber was connected to the Radon source

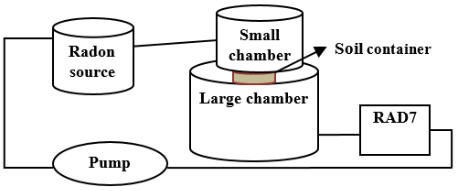

Figure 1. Schematic of Radon diffusion measurement system [16]

B. Theory

The Radon concentration in chamber 2 is decreased through two processes, radioactive decay (λ_d) and leakage. The total removal constant (λ_t) is defined as $\lambda_t = \lambda_l + \lambda_d$. The leakage constant (λ_l) is experimentally determined by comparison of the time variation of total Radon concentration and radioactive decay behavior of Radon in chamber 2 [11].

To find the diffusion coefficient of Radon through the sample container, the time-dependant one-dimensional diffusion equation for defined experimental condition must be solved [11].

$$D\frac{\partial^2 I(z,t)}{\partial z^2} - \lambda_l VI(z,t) - \lambda_t VI(z,t) = V\frac{\partial I(z,t)}{\partial t} \quad (1)$$

With boundary conditions of the experimental set-up, the spatial variations of Radon concentration in the samples could be ignored. Thus, the Equation (1) is reduced to

$$V_2\frac{dI_2(t)}{dt} = F - \lambda_d V_2 I_2(t) - \lambda_l V_2 I_2(t) \quad (2)$$

where, F is diffused Radon flux to the large chamber and is defined as Equation (3)

$$F = -SD\frac{\Delta I}{\Delta z} = -SD\frac{I_2(t) - I_1(t)}{d} \quad (3)$$

Combining Equations (2) and (3) we get

$$\frac{dI_2(t)}{dt} + \left(\frac{SD}{V_2 d} + \lambda_t\right) I_2(t) = -\frac{SD}{V_2 d} I_1(t) \quad (4)$$

In steady state method, Radon concentration does not change by time [3]

$$dI_2(t)/dt = 0 \quad (5)$$

Thus, the diffusion coefficient is obtained to be [16] as

$$D = \lambda_t V_2 d / S(I_1 / I_2 - 1) \quad (6)$$

In transient method, Equation (4) should be solved [12] as

$$I_2(t_1) = I_2(t_2)\left(1 - e^{-\left(\left(\frac{SD}{V_2 d} + \lambda_t\right)\Delta t\right)}\right) \quad (7)$$

In this method, Radon concentrations of the large chamber at two various times are measured before it reaches its constant value. Thus, diffusion coefficient in this method is given by Equation (8) [12].

$$D = -\frac{V_2 d}{S\,\Delta t}\ln\left[\left(1 - \frac{I_2(t_1)}{I_2(t_2)}\right) e^{\lambda_t \Delta t}\right] \quad (8)$$

Another factor in Radon transportation is soil effective permeability (k) that is dependent on soil properties and moisture. It is given by Equation (9) [13].

$$k = \left(\frac{\varepsilon}{110}\right)^2 d_a^{(4/3)} \exp(-12m^4) \quad (9)$$

Rogers and Nilson have also obtained the Equation (10)

$$D = D_a \exp(-6m - 6m^{14\varepsilon}) \quad (10)$$

That indicates the dependence of the Radon diffusion coefficient to the moisture and porosity of the soil sample. D_a: Radon diffusion coefficient in air is equal to 1.2×10^{-5} m^{2}s^{-1} [13].

In Equation (10), the relative soil moisture, m, is obtained by the following Equation (12) [14].

$$m = \frac{\theta - \theta_r}{\theta_s - \theta_r} \quad (11)$$

$$\varepsilon = \left(1 - \frac{\rho_b}{\rho_p}\right) \quad (12)$$

We can calculate Porosity (using paraffin method) from Equation (12) [15]. Mentioned symbols in Equations (11) and (12) will define Nomenclature section.

III. EXPREMENTAL PROCEDURE

Undisturbed soil samples were properly collected from a fault zone and placed in metal cylinders (diameter of 3.8 cm and height of 7.9 cm). Their weighed moisture, maximum and minimum volumes of moisture were measured and their compositions were determined. Then, the samples were fitted between the small (0.67 liter) and large chamber (12.26 liter) of the apparatus as shown in Figure 2 for Radon diffusion coefficient measurement.

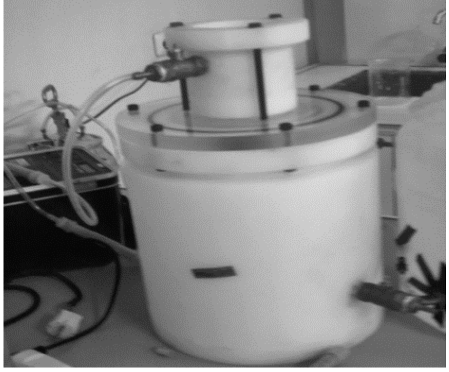

Figure 2. Photograph of Radon diffusion measurement system [16]

In order to determine the leakage of the large chamber, a Teflon cylinder of the same size of the sample holder was fitted between the two chambers and the small chamber was connected to the Radon source. When the Radon concentration reached to 53.5 kBq/m^3, the source was disconnected and the Radon concentration of the large chamber was measured during five days.

For obtaining the Radon diffusion coefficient by steady state method, the small chamber is connected to the Radon source in which the gas diffuses through soil sample and enters to the large chamber. When the Radon concentration of the large chambers reaches a constant value and the Radon concentrations are measured by RAD7 detector.

Since during the Radon concentrations measurement in the large chamber, the air in the detector system is mixed with Radon in the chambers, the Radon concentration measurements should be corrected by the Equation (13).

$$I_{2real} = \frac{V_D + V_2}{V_2} \times I_2 \tag{13}$$

where $V_D = 0.7$ m^3 the detector volume, V_2 is the large chamber volume and I_{2REAL} is corrected Radon concentration of the chamber.

IV. RESULTS

The Radon leakage of the large chamber is shown in Figure 3. By fitting exponential curve to the measured data of Figure 3, the leakage constant of large chamber is obtained to be λ_1=0.4×10^{-6} s^{-1}. Thus, will be λ_t=2.5×10^{-6} s^{-1}. Table 1 shows the components of the soil samples used for Radon diffusion measurement.

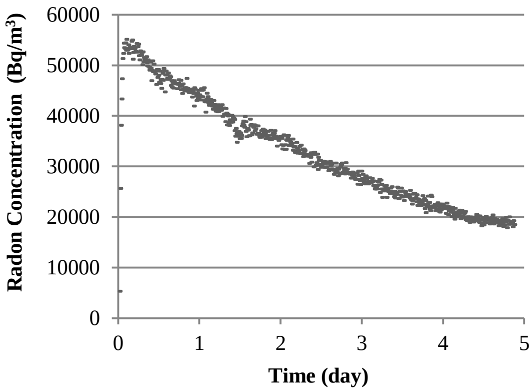

Figure 3. Radon concentration of large chamber versus Time [16]

Table 1. Percentages of the components of soil samples

Sampling depth [m]	Clay	Sediment	Sand	Tissue
0-0.3	15	30	55	Sandy loam

Table 2 shows the moisture content of the soil samples. Maximum and minimum of volume moisture in the soil of the fault zone are 50 and 3, respectively.

Table 2. Weighed moisture, volume moisture and moisture saturation of the soil samples

Sample No.	Volume moisture (%)	Saturation fraction	Weighed moisture (%)	Steady state period (days)
1	29	0.553	20	4
2	4.5	0.035	3.1	3
3	3.8	0.018	2.6	2
4	33.4	0.646	23	-
5	23.2	0.430	16	-
6	19.4	0.350	13.4	-
7	14.5	0.268	10	-

Samples No. 1, 2 and 3 given in Table 2 were examined using steady state method and other samples were examined using transient method. Table 3 shows Radon concentrations in the two chambers and the corrected Radon concentrations in the large chamber for soil sample numbers 1-3.

Table 3. Radon concentration values in the chambers for the quoted soil samples using the steady state method

Radon concentration (kBqm^{-3})				
Sample No	Weighed moisture (%)	I_1	I_2	Corrected I_2
1	20	69.9	41.2	43.5
2	3.1	58.8	53.2	56.2
3	2.6	55.2	48.9	51.7

Table 4. Radon concentration values in the chambers for soil samples (4-7 of Table 2) using the steady state method

Radon concentration (kBqm^{-3})					
Time interval (h)	Weighed moisture (%)	Corrected $I(t_2)$	$I(t_2)$	Corrected $I(t_1)$	$I(t_1)$
24.7	23	27.7	22.9	5.9	5.6
22.8	16	32.3	32.4	5.9	5.6
24	13.4	72.8	68.9	56.9	53.8
23.7	10	64.7	61.2	58	54.9

Table 4 shows the Radon concentrations in the large chamber in two different times for other samples of Table 2 using transient method. Inserting the Radon concentration values in Equations (6) and (8), the diffusion coefficients of the soil samples with various weighed moisture and relative moisture saturation were determined. The results are given in Table 5.

Table 5. Diffusion coefficients with different moisture content

Sample No	Relative moisture saturation ×10^{-2}	Diffusion coefficient ×10^{-6} (m^{2}s^{-1})	Weighed moisture (%)
1	55.3	0.4	20
2	3.5	5.1	3.1
3	1.8	3.5	2.6
4	64.6	0.54	23
5	43	0.47	16
6	35	1.9	13.4
7	26.8	2.8	10

Figure 4 shows the Radon diffusion coefficient of the soil samples versus the relative moisture saturation of the samples, by steady state and transient methods.

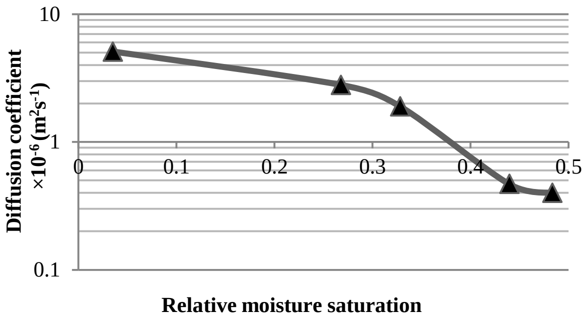

Figure 4. Radon diffusion coefficient of the soil samples versus their relative moisture saturation [16]

The diffusion coefficients have also been worked out using equation 10. Measuring the parameters in Equation (12), the average porosity of the soil sample was obtained 0.37. Hence, the area of soil grains, S=1-ε, will be 0.63×10^{-4} m^2 [17]. As the tissue soil samples are sandy loam, the soil grains are considered sphere. Thus, the average diameter of the soil particles can be estimated from standard soil classifications such as the 12 categories used by the U.S. Soil Conservation Service (SCS) [18].

$$S = 4\pi r^2 \;\rightarrow\; r = 2.25\times10^{-3} \;\rightarrow\; d = 4.5\times10^{-3} m$$

The soil samples permeability and their diffusion coefficients were obtained inserting D_a, ε and, m into Equations (9) and (10). The results are given in Table 6.

Table 6. Diffusion coefficients and Permeability of samples of various moisture content

$D \times 10^{-6}$ (m^{2}s^{-1})	2.23	1.83	1.45	0.52	3.9	3.8	0.9
$k \times 10^{-10}$(m)	3.62	3.22	2.56	0.48	3.85	3.85	1.25
m	0.27	0.35	0.43	0.65	0.02	0.04	0.55

The results of the theoretical diffusion coefficient versus relative moisture saturation of the soil samples are plotted in Figure 5. Then permeability of the soil samples as a function of the relative moisture saturations are plotted in Figure 6.

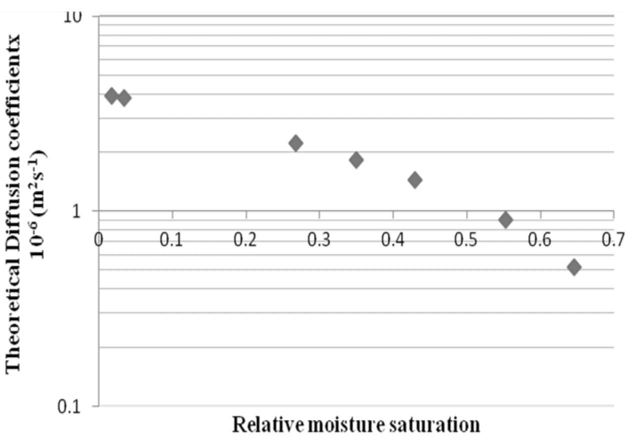

Figure 5. Theoretical diffusion coefficient versus relative moisture saturation of the soil samples

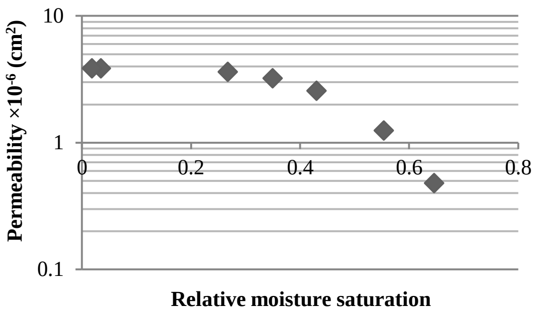

Figure 6. Permeability of soil samples versus relative moisture

V. DISCUSSION

Most of the researchers have overlooked the leakage of their apparatus in determining the Radon diffusion coefficient. But in this study, leakage of the device has also have been considered for precise Radon diffusion coefficient measurements and leakage was very low.

Figure 5 indicates that the diffusion coefficient of soil samples with average porosity of 0.37 and sandy loam texture, decreases as a function of their relative moisture saturation. This decrease is reasonable as increasing the moisture of the samples, the pores of the soil samples are filled with water and the Radon transport through water is much lower than when the pores were filled with air.

The behavior of the diffusion coefficient and permeability of the samples with porosity 0.35 is in agreement with the results obtained by Rogers and Nielson [7]. They obtained the permeability of sandy loam soil with porosity 0.35 and relative moisture saturation of 0.375 to be 1.2×10^{-2} m^2 and the diffusion coefficient to be 1.8×10^{-6} m^{2}s^{-1} [7].

Sandy loam soil with average porosity 0.37 and relative moisture saturation of 0.35, the permeability and diffusion coefficient were found to be 3.22×10^{-10} m^2 and 1.8×10^{-6} m^{2}s^{-1}, respectively. Permeability decreases when the moisture content of the samples increases in the both researches. But the small difference between the two researches is attributed to the porosity coefficients of the regional soil. Porosity coefficient is a function of moisture content of the samples and is constant for fully dry soil. When porosity in wet soil is low, the Radon gas can diffuse through pores of soil [2].

Shwekani et al. studied effect of soil moisture and porosity on the Radon diffusion. They found diffusion process decreases when moisture increases and by increasing porosity, Radon diffuses simpler through soil pores [19]

The other factor that influence on diffusion coefficient is gas effective pathway. Gas effective pathway is the spiral route that one gas molecule passes through textural soil to arrive surface. Each soil sample has a particular effective pathway [20].

VI. CONCLUSIONS

The total gas removal constant (λ_t) was found to be 2.5×10^{-6} s^{-1}. The small difference between the total removal constant and Radon decay constant (2.1×10^{-6} s^{-1}) shows that the chamber leakage is very low but for precise calculation of diffusion coefficient the leakage constant should be considered. However, the obtained diffusion coefficients of the soil samples of the fault zone indicate that the Radon exhalation rate employed as an earthquake precursor is strongly dependent on the soil texture and the moisture content of the soil zone.

NOMENCLATURES

D: Diffusion coefficient [m^2s^{-1}]
$I(z,t)$: Radon concentration at any depth z in the soil sample at any time [Bqm^{-3}]
S: Surface area of soil samples [m^2]
$I_2(t)$: Radon concentration in the large chamber at any time [Bqm^{-3}]
d: Height of the soil samples [m]
V_2: Volume of large chamber [m^3]
I_1(t): Radon concentration in small chamber [Bqm^{-3}]
Δt: Time interval between two measurements
$I_2(t_1)$: Radon concentration in large chamber at t_1 [Bqm^{-3}]
$I_2(t_2)$: Radon concentration in large chamber at t_2 [Bqm^{-3}]
d_a: Average diameter of soil grain in cm
k: Permeability
m: Relative moisture saturation of soil samples
ε: Porosity of soil samples
θ: Volume moisture of soil sample under consideration (%)
θ_r: Minimum volume moisture of soil samples (%)
θ_s: Maximum volume moisture of soil samples (%)
ρ_p: Soil bulk density [kg/m^3]
ρ_b: Density of soil grains (soil without porosity) [kg/m^3]

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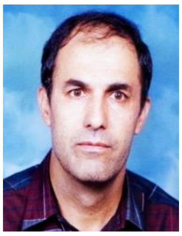

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