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Research Article

Experimental Study on Unsteady Radon Exhalation from the Overburden Layer of the Uranium Mill Tailings Pond under Rainfall

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In order to find out radon reduction performance of the overburden layer on uranium mill tailings (UMTs) pond beach surface after rainfall, the rainfall simulation experiment of the overburden layer was carried out with the self-developed equipment. Based on the radon migration model of the overburden layer on the UMTs pond beach surface, the change rule of radon exhalation in four types of compactness of the overburden layer within 120 hours after rainfall was studied, and the corresponding moisture content was also analyzed. The results show that the radon concentration in the overburden layer of UMTs increases nonlinearly; the dynamic change in moisture content of the overburden layer on the beach surface leads to the unsteady radon exhalation. The variation of radon exhalation shows three stages: increase, linear decrease, and stability tendency. After rainfall, radon exhalation rate increases due to water vapor and there is free radon seepage in pores. With the decrease of free radon production rate, radon exhalation rate gradually decreases until it reaches stability again. When the thickness of the overburden layer reduces, the porosity decreases with the increase in compactness of the overburden layer. While the decrease in radon reduction is more obvious, the less time it takes for radon exhalation to vary from unstable to stable overburden after rainfall.

1. Introduction

Radon, a radioactive inert gas which is the most easily exposed in people's daily lives [1, 2], is published by WHO as one of the 19 main environmental carcinogens [3]. The decay of radon and its daughters can be fatal to the human body [4]. Even decommissioned uranium mill tailings (UMTs) ponds still contain radium (^{226}Ra). The decay of radium causes a continuous buildup of radon atoms. Therefore, if the UMT pond is not properly disposed of, it will pose a fatal danger to the surrounding environment and human safety. The disposal with soil-covering system is the major means of decommissioning the beach surface of UMTs ponds at present, which can effectively inhibit radon exhalation. It is convenient to operate and has remarkable effects on radon reduction. The most common covering material with a wide range of sources is clay or sandy soil. There is lots of research on mixed material overburden layers which may further reduce the radon exhalation of UMTs pond. Many studies on the overburden layer of UMTs ponds have been conducted up till now [5–8]. Moreover, there are many researchers who have considered the influence of external environmental factors on radon exhalation from UMTs pond [9–11]. All of them aim to find the optimum UMT treatment project by considering various factors as much as possible.

It is recognized that the radon migration process in porous media is mainly driven by diffusion and convection. Radon and its daughters, which are produced from radioactive sources, migrate through the pores of porous media and diffuse to the overburden layer surface. Then under the action of concentration difference or external environment, convection and diffusion occur to complete the migration from overburden surface to air. A model for predicting radon migration in porous media was established which considered the effect of the coupling of heat and moisture [12]. A study of the exhalation of radon from a circular tubular cover layer was developed, which led to a formula for the radon exhalation rate under steady-state conditions [13].

The permeability of tailings and the overburden layer are closely related to the unsaturated state [14]. Radon migration models and radon exhalation under steady state have been performed [15, 16]. However, there are few studies on radon migration in an unsteady state induced by water saturation [17].

There is more rain in the south of China, and the moisture content of the soil is relatively sufficient [18]. The beach surface of the tailings pond is flat, and the mechanical macroscopic failure induced by rainfall is not significant [19]. The change in radon exhalation rate on the beach surface is mainly caused by soil evaporation changing moisture content. The process of water evaporation in soil is dynamic, which is often studied by means of soil moisture monitoring [20, 21]. Soil evaporation is affected by soil characteristics and external factors. The evaporation rate of soil is often in direct proportion to its initial moisture content and dry density [22].

In this study, the radon exhalation experiment was carried out on the UMTs pond beach surface after rainfall by a self-made simulation experiment device. By analyzing the increasing trend of radon concentration in the overburden layer of UMTs, the steady-state pattern of radon exhalation from the beach surface after rain was analyzed. Combining with the changes of moisture content and radon exhalation rate after different standing times, the radon exhalation law of UMTs pond beach surface after rainfall was explored. The results may provide a reference for the study of the long-term safety and stability of UMTs pond.

2. Theoretical Basis and Methodology

2.1. Radon Migration from the Overburden Layer of UMTs Pond. Radon control mechanism of the overburden layer on decommissioned UMTs pond beach surface [23] is to reduce the radon diffusion coefficient and increase the migration residence time of free radon in the overburden layer by using the covering medium and make radon decay in the overburden layer through the retention effect, so as to reduce the radon exudation rate. The radon migration model of the overburden layer on the UMTs pond beach surface is shown in Figure 1.

Since covering material has very low nuclide content, the radon produced by it is negligible. According to the general migration equation of radon in porous media, the migration equation of radon in the overburden layer on UMTs pond beach surface in an unstable state can be obtained [24].

$$\frac{\partial C_O}{\partial t} = D_O \Delta C_O - C_O \nabla \cdot v_O - v_O \cdot \nabla C_O - \lambda C_O. \tag{1}$$

Assuming that the overburden layer is continuous at the interface between UMTs and air, the volumetric concentration of radon in UMTs, the pores of the overburden layer, and air are continuous, and the seepage volume velocity is the same.

The continuity equation of radon concentration at the interface between UMTs and the overburden layer (at $x = x_0$) can be expressed as follows:

$$\begin{aligned} C_O|_{x=x_0} &= \frac{\eta_O}{\eta_T} C_T|_{x=x_0}, \\ D_O \mathrm{grad} C_O &= D_T \mathrm{grad} C_T + \left(1 - \frac{\eta_O}{\eta_T}\right) v_T C_T. \end{aligned} \tag{2}$$

The continuity equation of radon concentration at the interface between the overburden layer and air (at $x = x_1$) can be expressed as follows:

$$\begin{aligned} C_O|_{x=x_1} &= \eta_O C_A|_{x=x_1}, \\ D_A \mathrm{grad} C_A &= D_O \mathrm{grad} C_O + \left(1 - \frac{1}{\eta_O}\right) v_O C_O, \end{aligned} \tag{3}$$

where D_O, $D_{T,}$ and D_A are respectively the diffusion coefficients of radon in the overburden layer, UMTs and air ($m^2 \cdot s^{-1}$); C_O, $C_{T,}$ and C_A are respectively the concentration of radon in the overburden layer, UMTs and air ($Bq \cdot m^{-3}$); v_O, v_T are respectively gas seepage velocity in the overburden layer and UMTs ($Bq \cdot m^{-1}$); η_O, η_T are respectively the porosity of the overburden layer and UMTs; λ is the decay constant of radon (s^{-1}).

2.2. Effect of Soil Moisture on Radon Migration. Radon is easily soluble in water and is related to temperature τ (°C). The solubility coefficient K of radon in water is the ratio of radon concentration in the liquid phase to radon concentration in the gas phase [25].

$$K = 0.105 + 0.405 \times e^{-0.05027\tau}. \tag{4}$$

Radon migrates rapidly through fluid (air or water) in pores [26]. After rainfall, a large amount of rainwater infiltrates into the overburden layer on UMTs pond beach surface, and a large amount of free radon dissolves in the water of overburden layer. Radon migrates to the surface with water vapor under soil evaporation. The evaporation flux of an unsaturated soil surface is usually calculated by the Penman–Wilson formula [27].

$$E = \frac{\Gamma Q n + \eta E b}{\Gamma + \eta A}. \tag{5}$$

Where E is evaporation flux ($mm \cdot d^{-1}$); Γ is the slope of the relation curve between saturated vapor pressure and temperature; R_n is the net radiation amount of soil surface; η is the humidity constant; and $E_b = f(u) e_a (B - A)$, where $f(u)$ is the wind function, e_a is the vapor pressure of the air on the soil surface, B is the reciprocal of the relative humidity

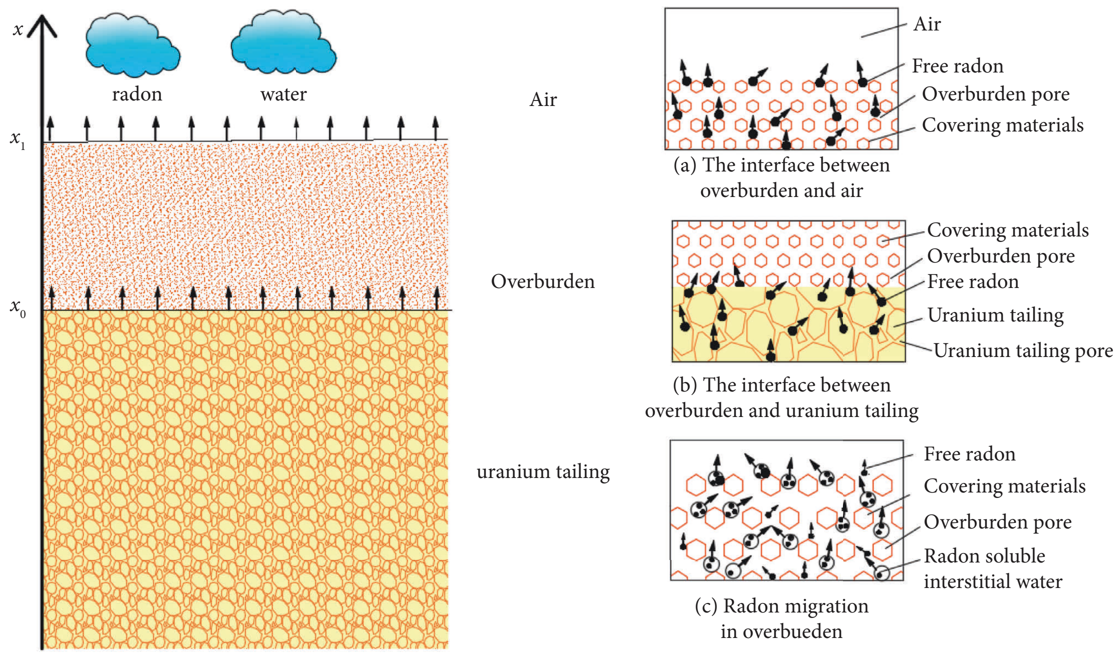


Figure 1: Radon migration model of the UMTs' pond beach overburden.

of the air, and A is the reciprocal of the relative humidity of the soil surface.

3. Experimental Device and Scheme

3.1. Raw Materials. In this experiment, materials to simulate the uranium tailing pond beach surface were taken from a UMT pond in south China. Red soil was selected as the overburden layer, which has a good radon reduction effect and a low cost of transportation and treatment [28]. The compositions of UMTs and red soil were analyzed by XRF, and the results are shown in Table 1. In addition, the uranium grade of uranium tailings was determined, and the uranium content of 0.07% was much higher than that of the natural background. Table 2 summarizes the physical properties of red soil.

3.2. Experimental Device. The device used in the experiment was a self-developed device, which mainly includes a cylindrical chamber, a constant temperature water tank, a rainfall simulator, and a metering/measurement system. The schematic diagram of the experimental device is shown in Figure 2. A cylindrical chamber was used to stack test samples and collect radon gas, with a diameter of 200 mm and a height of 600 mm, respectively. The constant temperature water tank was used to control the temperature and ensure the consistency of the experimental temperature. Its temperature regulation range is 5–50°C, and its accuracy is ±0.5°C. The rainfall simulation range of rainfall simulation is 1.2–40 $L \cdot h^{-1}$. Radon concentration was measured by a RAD7 radon meter, and moisture content was measured by a mass measuring device with an accuracy of 10 g.

3.3. Experimental Scheme. Rainfall was used to simulate the radon exhalation from the overburden layer on the UMTs pond beach surface under the effect of water erosion. In order to effectively and realistically simulate the radon exhalation from the UMTs pond beach surface, the thickness of the UMTs and the overburden layer of red soil were reset to 50 cm and 15 cm, respectively. According to the 2017 rainfall meteorological data from the Hunan Meteorological Bureau of China, a continuous heavy rainfall of 6 h (20 mm/h) was selected. The experimental ambient temperature was 24 ± 1°C. The specific experimental procedures are as follows:

(i) Pre-experimental treatment: firstly, 9.55 kg of dried UMTs was layered and compacted into the cylindrical chamber until the thickness is 15 cm; then, connect the radon collecting cover with the test cylinder by flange and check the air tightness of the device; finally, let the tailing stand for 72 h.

(ii) Experimental settings: The filling mass of dried overburden red soil was 7.49 kg, and the experimental conditions of overburden layer compaction were set as 0.56, 0.62, 0.70, and 0.80 corresponding overburden thickness was 20 cm, 18 cm, 16 cm, and 14 cm. The rainfall in each experimental condition was 1.63 L.

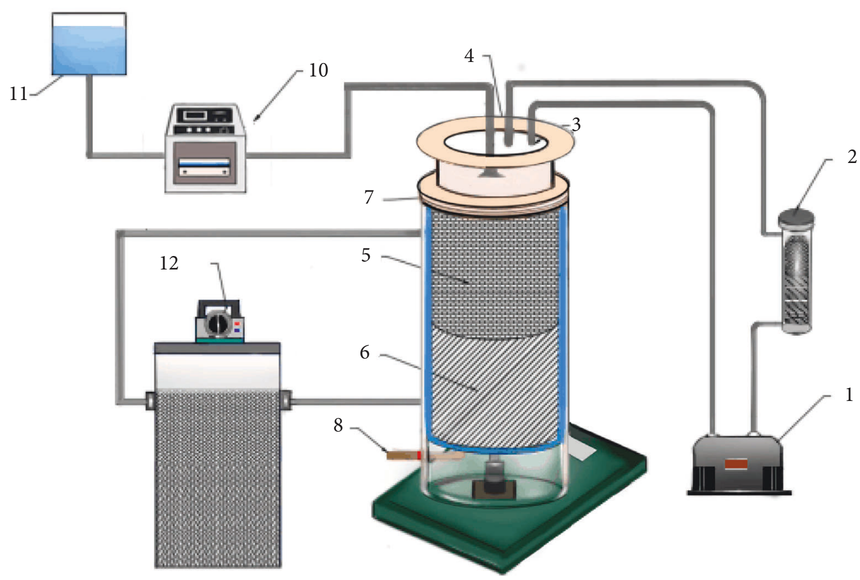
(iii) Experimental test: The RAD7 radon detector was used to measure the changes in radon concentration and moisture content before and after rainfall. The radon concentration measurement needs to decontaminate the experimental device and be recorded every 24 h.

Table 1: Main mineral compositions of test materials.

Test material	Mineral composition content/%							
	SiO_2	Al_2O_3	K_2O	Fe_2O_3	Na_2O	TiO_2	CaO	Others
Uranium tailing	85.250	6.180	3.180	1.630	0.418	0.371	0.276	2.695
Soil	51.672	16.497	4.021	8.516	0.563	0.563	14.078	4.090

Table 2: Physical properties of the tested soil.

Specific gravity, G_s	Plastic limit, w_p (%)	Liquid limit, w_L (%)	Maximum dry density, r_d (g/cm^3)	Optimum moisture content, w_{opt} (%)
2.7	28.0	39.8	2.1	23.3



1-RAD7 radon detector; 2-drying tube; 3-air inlet; 4-air outlet; 5-red soil overburden layer;

6-UMTs; 7-flange; 8-outfall; 9-electronic scale; 10-constant flow pump;

11-water tank; 12-constant temperature water tank

Figure 2: Schematic of a test device.

4. Results and Discussion

4.1. Trend of Radon Concentration after Rainfall. The moisture content of the overburden layer directly affects the radon reduction effect [29] of the overburden layer on UMTs pond beach surface. Soil evaporation changes the characteristics of seepage and pores in the soil, thus affecting radon exhalation in the overburden soil. In order to study the dynamic effect of soil evaporation effects on radon exhalation from the overburden layer after rainfall, the variation of radon concentration in the overburden layer on the UMTs pond beach surface was observed within 5 days of rainfall. The radon concentration accumulation method was used to measure the concentration of radon every 24 h, and the sampling interval was 10 min. The accumulated radon concentration and the increasing trend of the overburden layer with different compaction degrees are shown in Figure 3 after the natural resting for 24 h, 48 h, 72 h, 96 h, and 120 h after rainfall.

By comparing the final cumulative radon concentration in Figures 3(a)–3(d), the tailings overburden layer with lower compaction degree has higher cumulative values under the same conditions and the values exhibit a large difference, which indicated that the compaction degree has an important influence on radon reduction of UMTs pond beach surface. The nonlinear function was used to fit the cumulative radon concentration within 300 min, and the fitting degree (R^2) was all higher than 95%. The growth of radon concentration shows obvious nonlinear changes and the trends are shown in Figure 3, which is inconsistent with the linear growth of radon concentration in porous radioactive media under a steady state [30]. Therefore, it can be inferred that the radon concentration on the overburden layer on the tailings beach surface is an unsteady migration

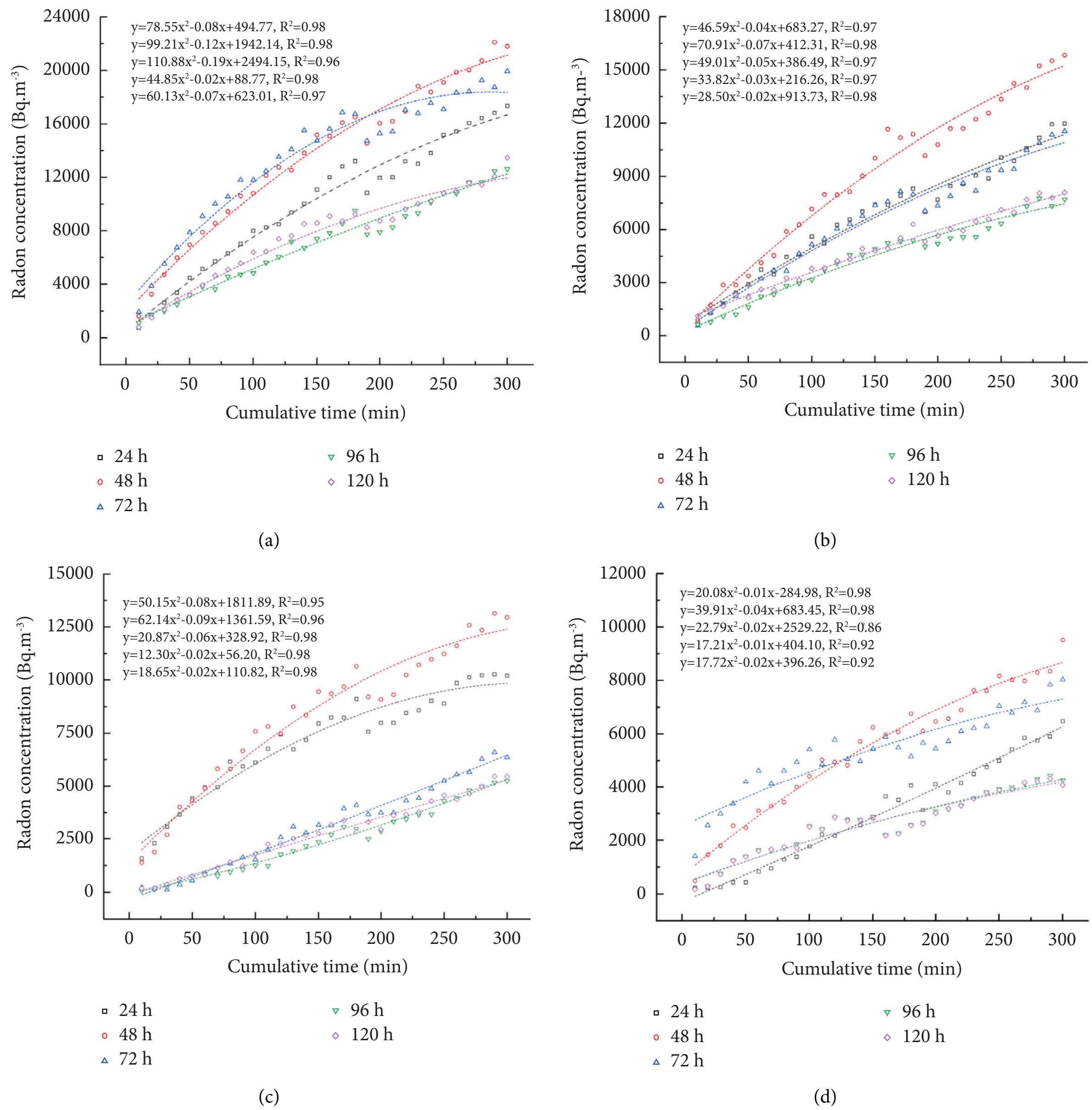


Figure 3: Cumulative radon concentration of the overburden layer with different compaction degrees. (see Figure 3(a)), compaction degree is 0.56,(see Figure 3(b)), compaction degree is 0.62, (see Figure 3(c)), compaction degree is 0.70, (see Figure 3(d)), and compaction degree is 0.80.

process during the resting period, which may be caused by the rainfall changes in the overburden layer.

4.2. Relationship between Radon Exhalation Rate and Soil Moisture. Based on the data of radon cumulative concentration in each group and 15 groups of data before and after, the radon exhalation rates J, J1-15, and J16-30 of the overburden layer were calculated. And the difference between *J,* J1-15, and J16-30 was taken as the radon exhalation error.

The moisture content of samples after 24 h, 48 h, 72 h, 96 h, and 120 h of resting was calculated. The curves of radon exhalation rate change after resting in different compaction degree of tailings overburden layer and moisture content change of experimental samples are obtained, as shown in Figure 4.

It can be seen from Figure 4 that radon exhalation from the tailings overburden layer tends to be stable within 5 days and no longer change greatly. The variation process of the moisture content of the specimens and the radon exhalation rate of the overburden layer with time could be roughly divided into 3 stages. In stage I, moisture content decreased slowly and radon concentration increased. In stage II, the moisture content and radon exhalation decreased linearly, and the decreasing rate of moisture content increased obviously. In stage III, moisture content and radon exhalation went into stable states. Experimental results show that radon exhalation increases first and then decreases with the decrease in moisture content. When radon migration was in

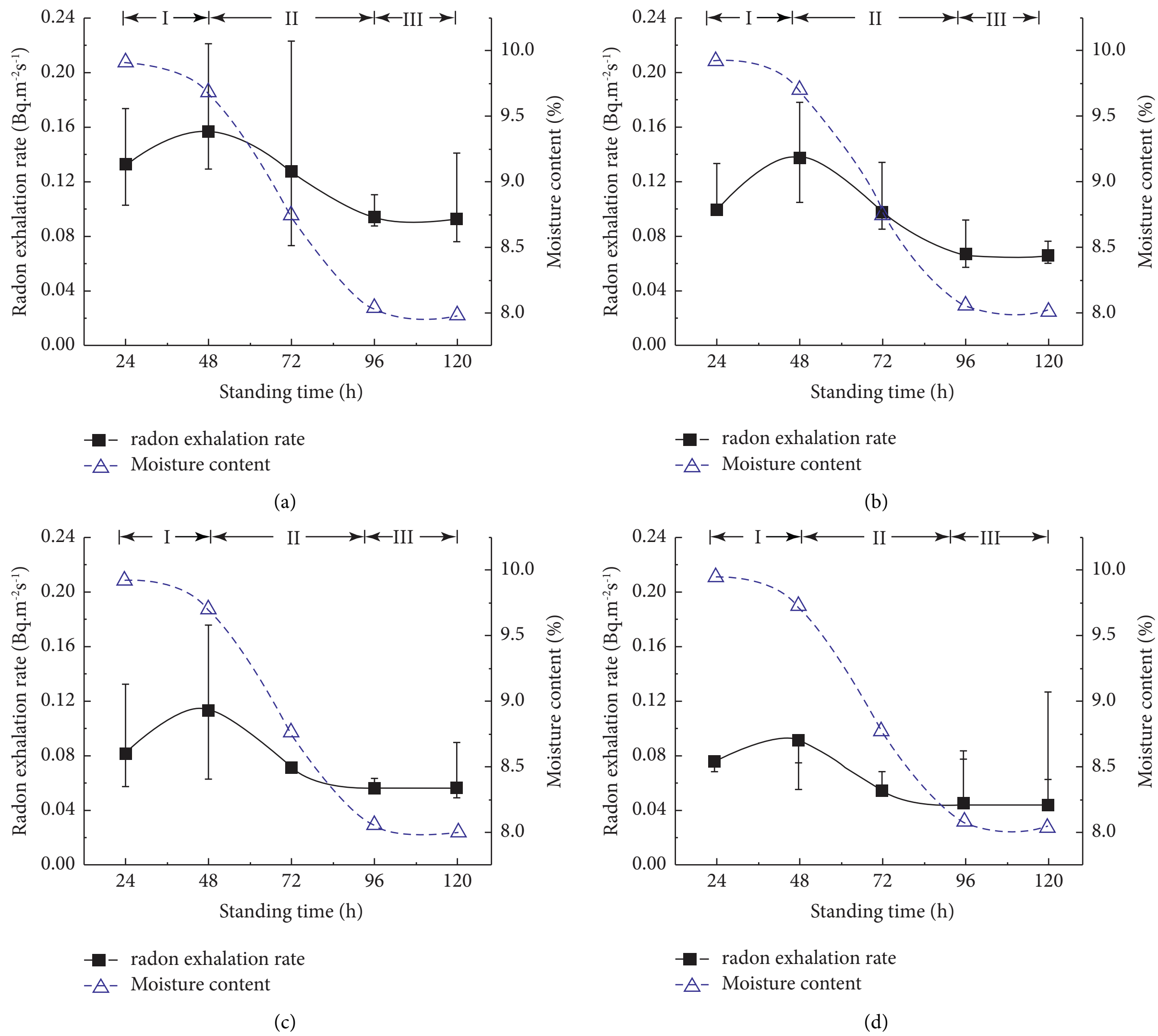


FIGURE 4: Variation curves of radon exhalation rate and moisture content, compaction degree is 0.56 (see Figure 4(a)), compaction degree is 0.62 (see Figure 4(b)), compaction degree is 0.70 (see Figure 4(c)), and compaction degree is 0.80 (see Figure 4(d)).

the way of diffusion mainly, radon exhalation increased first and then decreased with the increase in moisture content [24]. It indicates that the radon migration in the experiment was dominated by water vapor coordinated with seepage migration, which is driven by soil evaporation.

The rainfall in stage I caused a large amount of water on the surface of the overburden layer, which blocked the pores of the overburden layer and hindered the evaporation of internal water. While a large amount of free radon dissolved in water in pores and migrated with the water through seepage, moisture content changed slowly and radon exudation rate increased. In stage II, with the evaporation of water on the surface of the overburden layer, the pore channels of the overburden layer gradually opened, increasing the seepage in pores. The free radon in pores decreased with the migration of seepage. In stage III, the moisture content tended to be stable and the radon migration gradually stabilized as the free water in pores decreased.

The greater the compaction degree of the overburden layer is, the smaller the porosity and thickness. After 120 h, the radon exhalation rate of the overburden layer increased with the compaction degree in the following order: 0.0926, 0.0666, 0.0569, and 0.0441 Bq·m^{-2}·s^{-1}, covering thicknesses of 20, 18, 16, and14 cm. This indicates that reducing the porosity of overburden layer material is more effective than increasing overburden thickness when the thickness is small. The variation trend of moisture content with different compaction degrees was basically the same, while the radon exhalation exhibited large differences. When the compaction degree was 0.65 and 0.80, radon exhalation entered stage III at 96 h and 72 h, respectively. With the increasing compaction degree of the overburden layer, the time needed for radon exhalation stabilization has been reducted.

5. Conclusions

After rainfall, the accumulated radon concentration of the overburden layer on UMTs pond beach surface with different compaction degree showed nonlinear increasing trend. Radon migration in the overburden layer was unsteady with the influence of dynamic changes in moisture content.

The radon exhalation from the overburden layer on the UMTs pond beach surface and the moisture content of the samples all showed three stages: radon exhalation first increases, then decreases, and finally tends to be stable. The moisture content of the samples first slowly decreases, then linearly accelerates to decrease, and eventually becomes stable.

Radon migration in the overburden layer on the UMTs pond beach surface after rainfall is mainly affected by water vapor evaporation of soil, while water vapor was coordinated with radon seepage. It led to unsteady radon exhalation from the overburden layer.

When the thickness of the overburden layer was small, the radon reduction effect of the overburden layer was greatly affected by the porosity. The smaller the porosity was, the more obvious the radon reduction effect was, and the radon reduction performance of the overburden layer stabilized faster after rainfall.

Data Availability

The data from this study are available upon request to the corresponding author.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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