Mechanism of radon exhalation suppression in radioactive tailings using $Na_2O \cdot nSiO_2 - CaCl_2$ modified covering soil

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Abstract: The radon control mechanism of $Na_2O \cdot nSiO_2 - CaCl_2$ modified soil was studied through the laboratory simulation experiment of tailing covering radon control. The radon exhalation rate (J) is negatively correlated with the coverage thickness (H), and it has a non-linear relationship with the temperature. The moisture content variation rate of the covering soil significantly decreases, which helps to reduce soil damage and enhance the resistance of the covering soil to ambient temperature interference. The formation of silicic gel and C−S−H gel effectively optimizes the pore structure and permeability, reduces the diffusion and migration of radon gas in the covering soil, and the average radon exhalation rate is decreased by $1.01 \times 10^{-2} \, \text{Bq/(m}^3 \cdot \text{s})$. The research results show that the $Na_2O \cdot nSiO_2 - CaCl_2$ modified covering soil can effectively improve the radon control performance of the covering soil and reduce the cost of cover treatment.

Key words: radon exhalation; C−S−H gel; radon reduction mechanism; modified covering soil

1 Introduction

Radioactive tailings (RTs) are tailings containing uranium, thorium and other radioactive nuclides, and are mainly disposed by being stored in tailings ponds [1]. They are continuous sources of radon (^{222}Rn). Radon gas is a Group 1 carcinogen that can enter the human body through respiration or via contact with water, significantly increasing the risk of lung cancer [2,3]. Radon control via soil covering is the key aspect in the treatment of the RTs pond, and has been the focus of research worldwidely. The covering layer is susceptible to failure due to permeability degradation during long-term service, and it is difficult to meet the demand of long-term radon control under complex geological conditions [4−6]. Therefore, developing modified covering soils to optimize pore structure, enhance crack resistance, and reduce permeability has become the research direction of RTs pond treatment [7].

The radon control performance of the covering layer is closely related to the thickness, temperature, humidity, and pore characteristics [8,9]. The radon exhalation rate is largely related to the thickness of the covering medium [10,11]. The increase of covering soil thickness will prolong the radon migration path, and the radon exhalation rate will decrease [1,12,13]. However, if the thickness of the covering soil is too large, the increase of self-weight will lead to a decrease of soil porosity [14]. This will lead to the decrease of water regulation ability and crack resistance of the covering soil. High temperature will accelerate soil water evaporation, increase pore connectivity and radon

diffusion coefficient [15−17]. The pore size of soil also has a significant influence on radon migration. Micropores (<1 μm) can absorb radon by capillary action, while macropores (>10 μm) are the main channels for radon migration [18]. In the humidity saturated state, water molecules will occupy the pore space and inhibit radon migration [19]. However, long-term over-wetting of soil will induce soil softening and crack expansion [3,20]. Therefore, reasonable soil thickness and water retention characteristics can significantly improve the radon resistance of covering soil.

Soil modification is primarily categorized into physical and chemical modifications, such as mechanical compaction, soil reinforcement, and cement grouting [21−24]. The $Na_2O·nSiO_2−CaCl_2$ modification is a composite soil modification technique characterized by high reactivity and gel-forming capability, which can enhance the microstructure and chemical properties of the soil [25,26]. $Na_2O·nSiO_2$ (sodium silicate) is an alkaline activator that reacts with $CaCl_2$ to form C−S−H gels (calcium silicate hydrates) that fill soil pores and reduce permeability [27−29]. Through ion exchange and gelling, macropores in the soil can be transformed into nanoscale pores [29], and radon diffusion channels can be reduced. The reaction product silica gel [$nSiO_2·(m−1)H_2O$] has a high specific surface area and gelling activity, and can be adsorbed on the surface of soil particles. The gel covers the surface of soil particles, blocking the water migration channel and reducing the permeability coefficient [30].

To improve the radon control performance of traditional covering soil, this study proposed a synergistic modification scheme based on $Na_2O·nSiO_2−CaCl_2$. Indoor physical simulation experiments were conducted to model radon control in the modified covering soil for the RTs pond. The effects of soil thickness and ambient temperature on water retention and radon control performance of $Na_2O·nSiO_2−CaCl_2$ modified soil were studied by comparative experiments. This study can provide a novel solution for the long-term management of the RTs pond.

2 Experimental

2.1 Test materials

The uranium tailing used in the test was taken from a uranium tailing in South China, which was a high silicon sand type radioactive tailing. The tailing had a density of 2.60 g/cm^3 and a radium content of 8.61 Bq/g. The red soil used in the mulch was taken from the local red soil in Hengyang City, Hunan Province, China. The content of Al_2O_3 and CaO in the soil was high, and the natural soil sample was plastic and reddish-brown. According to the Test Methods of Soils for Highway Engineering (JTG E40—2020), the physical property parameters of soil were measured (Table 1). The soil density was 2.41 g/cm^3, the optimal water content and standard maximum dry density were 18.42% and 1.92 g/cm^3, and the plastic limit and liquid limit were 17.62 wt.% and 33.64 wt.%, respectively.

Table 1 Physical properties of soil

Parameter	Value
Density/(g·cm^{-3})	2.41
Optimal moisture content/%	18.42
Maximum dry density/(g·cm^{-3})	1.92
Plastic limit/wt.%	17.62
Liquid limit/wt.%	33.64

Sodium silicate ($Na_2O·nSiO_2$) and calcium chloride ($CaCl_2$) were selected for soil modification. Sodium silicate was a white crystalline powder, purchased from Qingdao Bay Chemical Co., Ltd. (China). Its modulus was 2.87, and the bulk density was 0.55 g/cm^3. The contents of Na_2O and SiO_2 were 21.73 wt.% and 60.44 wt.%, respectively. Calcium chloride was purchased from Sinopsin Group Chemical Reagent Co., Ltd. (China). It was a white powder with a purity of 98.0wt.%.

2.2 Radon exhalation test

The experiment was carried out in a laboratory with a diameter of 240 mm and a height of 600 mm, and the sealing cover with a height of 100 mm was used at the top. A constant temperature water tank with an accuracy of ±0.5 °C was used for constant temperature control, RAD−7 radon detector was used for radon concentration measurement, and the humidity was controlled below 10%. The radon detector had been calibrated in the National Standard Radon Chamber at the University of South China. The experimental device is shown in Fig. 1.

The initial water content of the experimental covering soil was 15% and density was 1.40 g/cm^3.
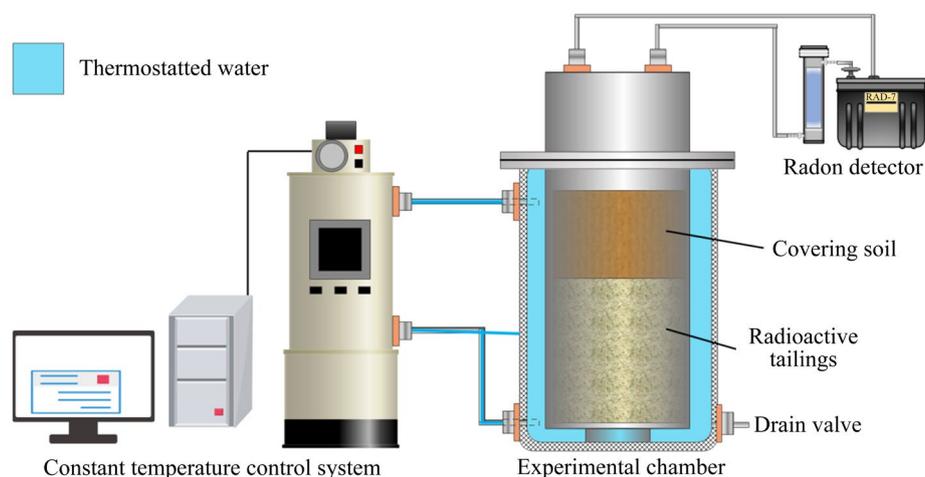
Fig. 1 Schematic diagram of test device

The pure soil experimental group was added with pure water. In the modified soil cover group, the mass ratio of $Na_2O \cdot nSiO_2$ to $CaCl_2$ was 2:1, and the addition amount was 2% of the total mass of the soil cover. Before the experiment, sodium silicate and calcium chloride were configured as solutions, respectively. The filling thickness of the tailings was 20 cm, and the covering thicknesses were 4, 8, 12, and 16 cm, respectively. After covering 2 cm of red soil, $CaCl_2$ solution and $Na_2O \cdot nSiO_2$ solution were alternately sprayed on the covering soil.

After loading the sample, the ambient test temperature (15, 20, 25, and 30 °C) was set according to the experimental plan. After the sample was sealed for 5 d and then opened for 2 d, the test sample preparation was finally completed. The RAD−7 radon detector was used to measure the cumulative radon concentration of the samples [31,32]. The measuring mode of the radon detector was set as "sniff mode", the cycle time was 10 min and the total time was 150 min.

2.3 Soil evaporation test

The test samples were divided into the pure soil covering group (PS) and the $Na_2O \cdot nSiO_2-CaCl_2$ modified covering group (MS). The cylindrical specimens with a diameter of 50 mm and a height of 4, 8, 12, and 16 cm were used for evaporation experiments. In order to ensure the uniform distribution of water and solution, the soil was alternately filled and the solution was sprayed with 2 cm as the layered unit. The evaporation test was carried out in a constant temperature and humidity test chamber with a temperature of (35 ± 0.5) °C and a humidity of (60 ± 2)%.

2.4 Experimental data processing

In order to study the radon exhalation rates of pure soil and modified soil samples at different ambient temperatures and soil thicknesses, the closed chamber method was used to measure the radon exhalation. The formula for calculating the radon exhalation rate of the covering soil layer is as follows [7,33]:

$$J = \frac{\lambda V \left[C - C_0 \exp(-\lambda t) \right]}{S \left[1 - \exp(-\lambda t) \right]} \tag{1}$$

where J is the radon exhalation rate on the surface of the sample, $Bq \cdot m^{-3} \cdot s^{-1}$; S is the area of radon exhalation surface of the sample, m^2; V is the volume of radon collector, m^3; λ is the decay constant of radon, and $\lambda=2.1\times10^{-6}\,s^{-1}$; C is the cumulative radon concentration in the radon collector at time t, $Bq \cdot m^{-3}$; C_0 is the initial radon concentration in the radon collector, $Bq \cdot m^{-3}$; t is the radon collection time, s.

Because the sampling time t is short, Eq. (1) can be simplified as [32]

$$J = \frac{(C - C_0) \cdot V}{S \cdot t} \tag{2}$$

3 Result and discussion

3.1 Radon exhalation of pure and modified covering soils

The cumulative radon concentrations of the pure soil and $Na_2O \cdot nSiO_2-CaCl_2$ modified covering

soil samples are shown in Figs. 2 and 3, respectively. The accumulation method is one of the main approaches for measuring radon exhalation rate. The radon exhalation rate can be calculated by fitting the concentration−time curve using the least squares method [34]. The linear correlation coefficient (R^2) between the cumulative radon concentration and the cumulative time is above 0.916. The results show that the radon concentration increases linearly, and the effects of radon leakage and back-diffusion on the measurement of cumulative radon concentration in the overburden are negligible [31]. Under the same conditions, the cumulative radon concentration within 100 min after the modified covering soil is generally lower than that of the pure soil (Figs. 2 and 3). This indicates that the radon exhalation rate of the modified soil decreases. The radon exhalation rate of each sample can be calculated by Eq. (3) [35,36]:

$$J = k\frac{V}{S} \tag{3}$$

where k is the linear fitting slope of the cumulative radon concentration−time curve.

The radon exhalation rates of samples at different ambient temperatures and covering thicknesses are calculated, as listed in Table 2. The radon exhalation rate of the sample decreases with the increase of covering thickness. Radon migration is mainly dependent on molecular diffusion and convection [37,38]. The increase in the thickness of the covering layer can prolong the diffusion path required for radon migration from the radon source area to the surface. According to Fick's first law, the diffusion flux is proportional to the concentration gradient and inversely proportional to the diffusion distance [39]. As the thickness (L) of the covering layer increases, the concentration gradient ($\Delta C/L$) of radon in the covering layer gradually decreases, which leads to a reduction in the diffusion flux and a decrease in radon exhalation rate.

Temperature can cause heat convection in the covering soil layer, which can accelerate the radon migration and lead to the increase of radon exhalation [40,41]. In Table 2, the radon exhalation rate of the covering soil samples does not show a single increasing trend with the increase of temperature, but presents a nonlinear correlation.
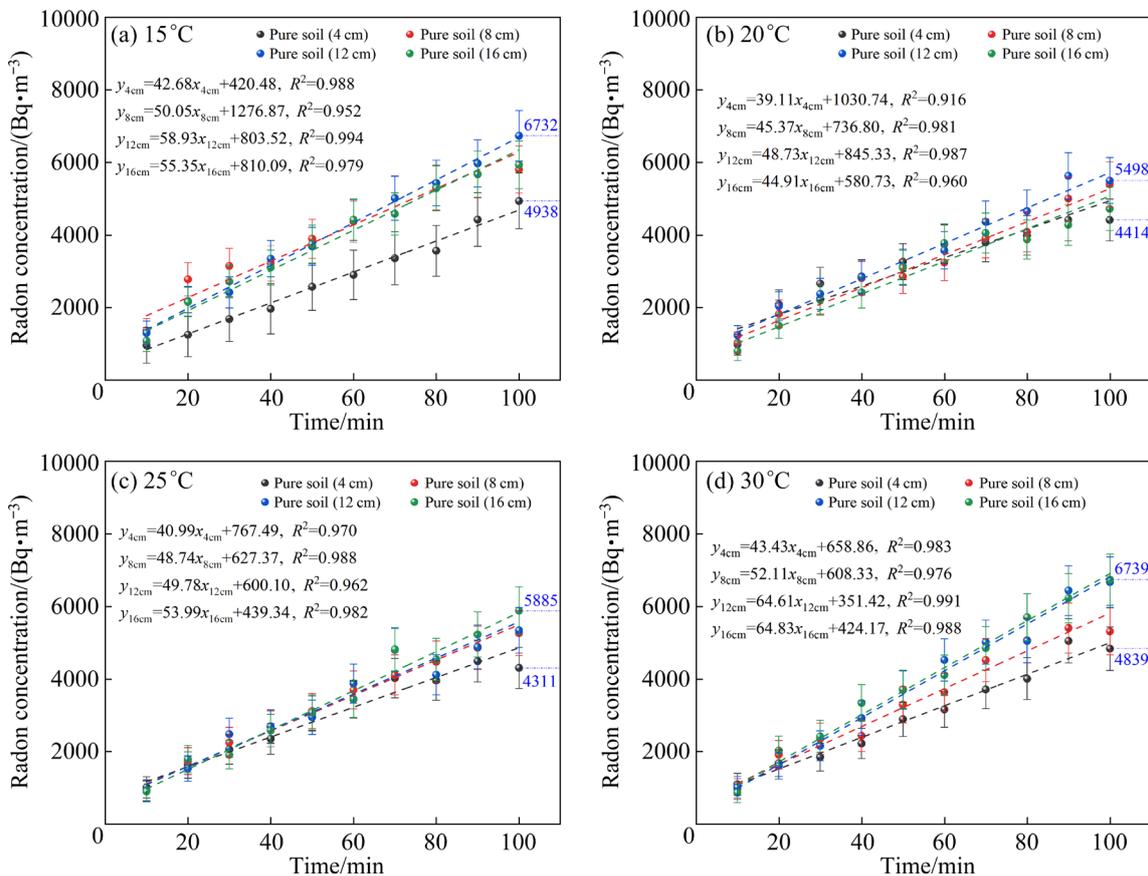

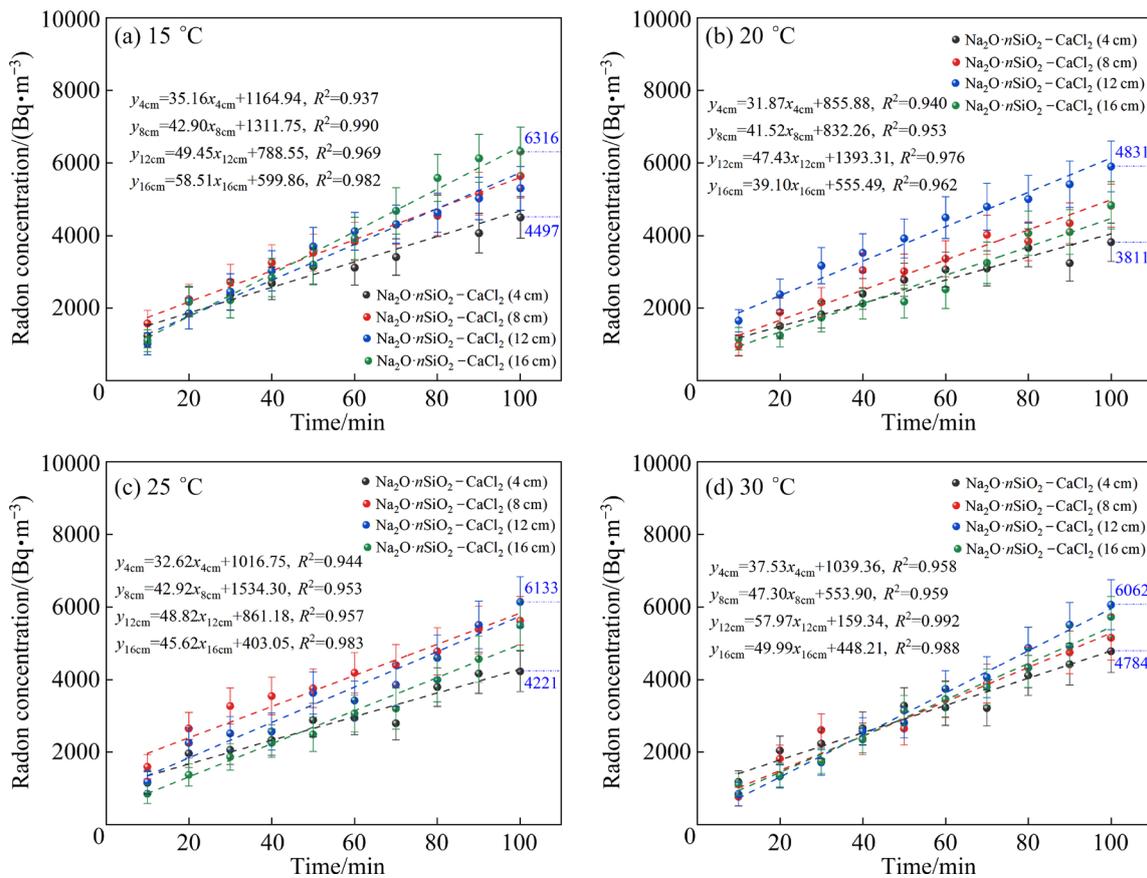**Fig. 2** Cumulative radon concentration of pure soil samples

Fig. 3 Cumulative radon concentration of $Na_2O \cdot nSiO_2 - CaCl_2$ modified samples

Table 2 Radon exhalation rate of covering soil samples before and after modification (10^{-2} Bq/(m³·s))

Thickness/ cm	15 °C		20 °C		25 °C		30 °C	
	Pure soil	Modified soil	Pure soil	Modified soil	Pure soil	Modified soil	Pure soil	Modified soil
4	11.38	9.38	10.43	8.50	10.93	8.70	11.58	10.01
8	10.01	8.58	9.07	8.30	9.75	8.58	10.42	9.46
12	7.86	6.59	6.50	6.42	6.64	6.51	8.61	7.73
16	3.68	3.90	2.99	2.61	3.60	3.04	4.32	3.33

Taking 4 cm pure soil sample as an example, the highest radon precipitation rate at 30 °C is 11.58×10^{-2} Bq/(m³·s). Temperature can affect the moisture content of soil cover, and the moisture content of soil is higher when the temperature is lower. The water dissolves the radium element, thereby increasing the radon concentration in the pore water [42]. At the same time, the adsorption of radon atoms on the surface of soil particles is reduced, and the release efficiency of radon is improved [43]. The radon exhalation rate of 4 cm pure soil sample at 15 °C (11.38×10^{-2} Bq/(m³·s)) is higher than that at 20 °C (10.43×10^{-2} Bq/(m³·s)). By comparing the radon exhalation rate before and

after modification, the radon exhalation rate of the modified soil is obviously lower than that of pure soil under the same experimental conditions. The results show that $Na_2O \cdot nSiO_2 - CaCl_2$ can effectively improve the radon control performance of the pure soil.

3.2 Change of water content of pure soil and modified covering soil

Radon is soluble in water, and water evaporation can accelerate radon percolation, so the water content of soil cover is an important factor affecting radon exhalation [44]. Figure 4 shows the change curves of the water content of pure soil and
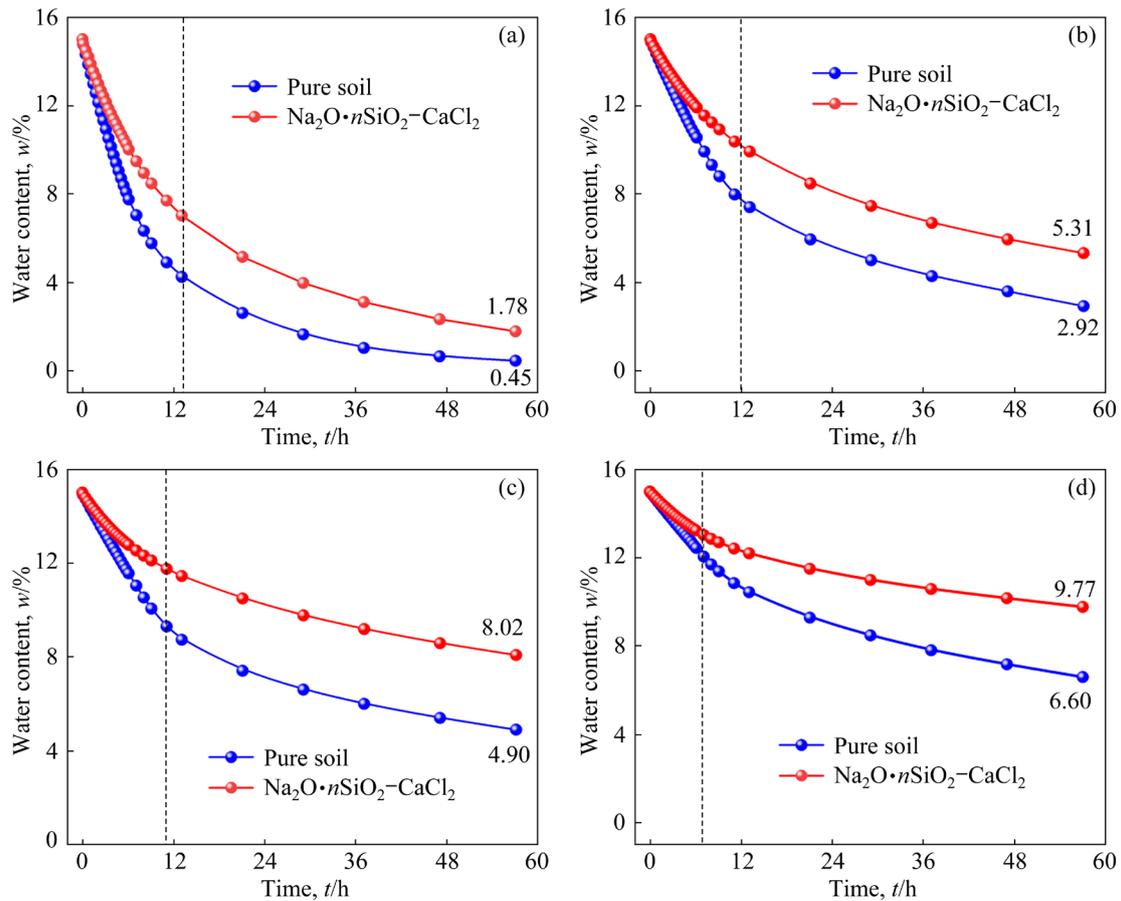
Fig. 4 Water content of samples with different thicknesses: (a) 4 cm; (b) 8 cm; (c) 12 cm; (d) 16 cm

modified covering soil at 35 °C. The water content of the modified soil is higher than that of the pure soil, indicating that the modified soil has better water retention characteristics. By comparing Figs. 4(a−d), it can be seen that the difference of water content in the same evaporation time increases with the thickness of the covering soil. After drying, the moisture contents of 4 cm-thick pure soil and modified covering soil are only 0.45% and 1.78%, respectively, which are close to that of dry soil. The water contents of 16 cm-thick pure soil and modified covering soil are 6.6% and 9.77%, respectively, and the difference of water content between them is 3.17%.

In order to further analyze the evaporation rate of covering soil, the water content change rate (v_w) is used to represent the change in soil water content within a unit of time.

$$v_w = \frac{w_{t_1} - w_{t_2}}{t_2 - t_1} \tag{4}$$

Figure 5 shows the change rates of water content of the pure soil and the modified soil. The water content change rate of Na$_2$O·nSiO$_2$−CaCl$_2$ modified soil becomes significantly smaller, and the maximum change rate of pure soil is 1.125, while that of modified soil is 0.816 (Fig. 5). The difference of the water content change rates before and after modification increases with the increase of thickness. When the thickness of covering soil is 4 cm, the water content change rate of them is similar. The water content change rate directly affects the dynamic balance of soil pore water. When the evaporation rate is high, the soil surface water is lost rapidly and a significant moisture gradient can be formed [45,46]. This leads to the upward migration of water by capillary action, which aggravates the radon seepage. At the same time, the rapid change of water content can lead to the rapid change of soil microstructure, the formation of soil damage, and the increase of the radon migration channel. Therefore, the Na$_2$O·nSiO$_2$−CaCl$_2$ modified covering soil can effectively reduce the change rate of water content of the soil cover, and help to improve the radon control performance of the covering soil.

3.3 Radon control mechanism of Na₂O·nSiO₂−CaCl₂ modified covering soil

As shown in Fig. 6, $Na_2O \cdot nSiO_2$ (sodium silicate) and $CaCl_2$ can chemically react in the soil pores to produce silica gel $nSiO_2 \cdot (m-1)H_2O$ and calcium silicate hydrate (C−S−H gel) [27]. Silicic acid gel has strong cementation and can fill the pores among soil particles [28]. At the same time, the soil particles can be cemented together to improve the integrity of the soil [47], to improve the radon control performance of the covering soil (Fig. 6(b)). The $Ca(OH)_2$ produced in the reaction can dissociate Ca^{2+} and OH^- from the covering soil. The surface of soil particles is usually negatively charged and absorbs cations. Due to the high valence of Ca^{2+}, there is an ion exchange with low-valent cations (such as Na^+ and K^+) originally adsorbed on the soil particles surface [48].

Ca^{2+} ions become more firmly adsorbed onto soil particle surfaces, resulting in a thinner double electric layer at the particle interfaces (Fig. 6(c)) [49]. The electrostatic repulsion among the particles is reduced, which makes the soil particles close to each other and condense, and enhances the stability of the covering soil. At the same time, the silicic acid gel fills soil pores in colloidal form. These
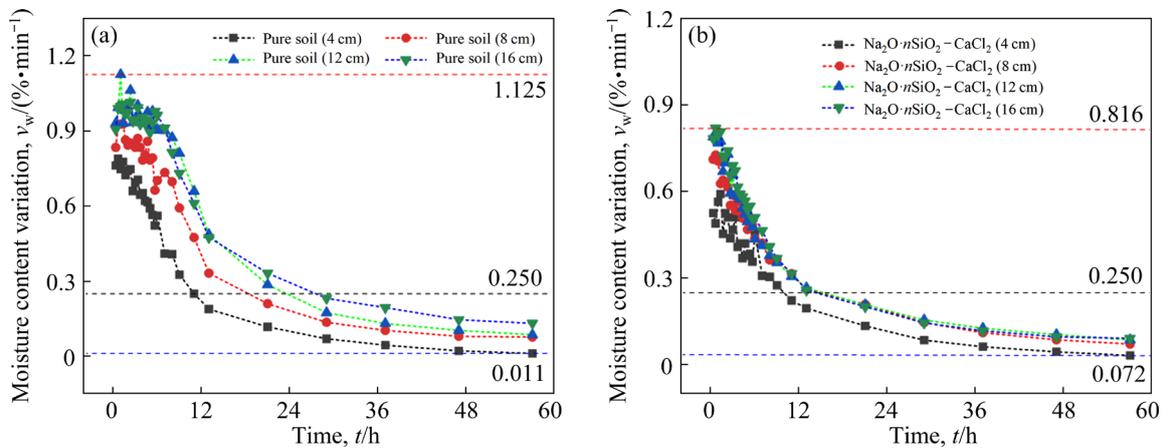

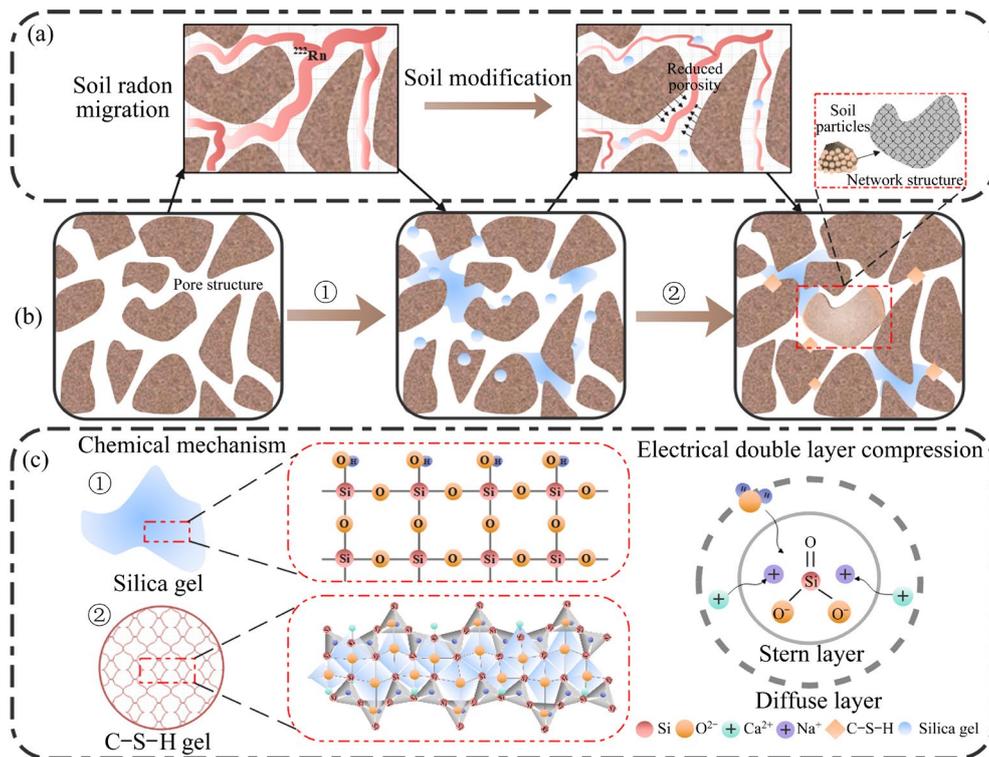**Fig. 5** Moisture content variations of overburden soils

Fig. 6 Microscopic radon control mechanism of $Na_2O \cdot nSiO_2$−$CaCl_2$ modified covering soil

colloidal particles occupy intergranular micropores, reducing pore size and permeability of the overlying soil layer [50], thereby suppressing the diffusion and migration of radon gas within the soil matrix (Fig. 6(a))

Figure 7(a) shows the radon exhalation rates of pure soil and $Na_2O \cdot nSiO_2−CaCl_2$ modified covering soil at different temperatures. ΔJ is the fluctuation range of radon exhalation rate with different thicknesses at the same temperature:

$$\Delta J = J_{max} − J_{min} \qquad (5)$$

The use of $Na_2O \cdot nSiO_2−CaCl_2$ to modify the covering soil reduces the permeability of the covering soil [45,51], and can effectively reduce the influence of ambient temperature on the radon control performance of the covering soil. As shown in Fig. 7(a), the radon exhalation rate of the $Na_2O \cdot nSiO_2−CaCl_2$ modified covering soil decreases uniformly at different temperatures, and ΔJ decreases somewhat. The mean value and fluctuation range (ΔJ_{PS}) of radon exhalation in pure soil at 30 °C are 8.73×10^{-2} and 7.26×10^{-2} Bq/(m³·s), respectively. After modification, the mean radon exhalation and the fluctuation range (ΔJ_{MS}) are 7.63×10^{-2} and 6.68×10^{-2} Bq/(m³·s), respectively.

Figure 7(b) shows the radon exhalation rate of pure soil and $Na_2O \cdot nSiO_2−CaCl_2$ modified covering soil under different cover thicknesses. The radon exhalation rate (J) and the cover thickness (H) conform to $J = a − b\exp(cH)$. Before and after modification, the cover thickness−radon exhalation rate models are expressed as follows:

$$J_{PS} = 0.132 − 0.012\exp(0.128H_{PS}) \qquad (6)$$

$$J_{MS} = 0.098 − 0.002\exp(0.208H_{MS}) \qquad (7)$$

It can be seen from Fig. 7(b) that the radon exhalation rate of the modified $Na_2O \cdot nSiO_2−CaCl_2$ covering soil decreases with different cover thicknesses, and the decrease of radon exhalation rate is greater at smaller cover thicknesses. The analysis of the error band of radon exhalation rate shows that the influence of temperature on radon exhalation rate fluctuates little. The $Na_2O \cdot nSiO_2−CaCl_2$ modified covering layer soil effectively reduces the soil porosity and improves the soil integrity. The modified covering layer soil reduces the radon gas migration path. The average exhalation rate of radon decreases from 7.99×10^{-2} to 6.98×10^{-2} Bq/(m³·s), reduced by

1.01×10^{-2} Bq/(m³·s). $Na_2O \cdot nSiO_2−CaCl_2$ modified covering soil can effectively reduce the thickness of covering soil and the cost of covering treatment. The effect of ambient temperature on radon control performance can be effectively reduced.
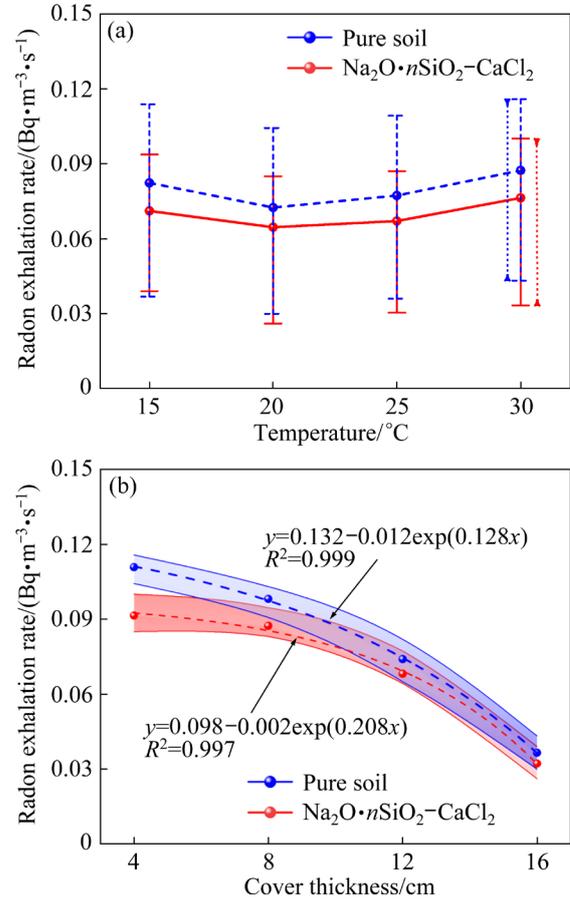

Fig. 7 Radon exhalation rate of pure soil and $Na_2O \cdot nSiO_2−CaCl_2$ modified covering soil: (a) At different experimental temperatures; (b) With different cover thicknesses

4 Conclusions

(1) The radon exhalation rate decreases with the increase of covering thickness, and the radon exhalation rate (J) and the covering thickness (H) conform to $J = a − b\exp(cH)$. Due to the coupling effect of temperature and water content, the radon exhalation rate of the overlying soil layer has a nonlinear relationship with temperature.

(2) The increase of the water content change rate aggravates the radon seepage, and the rapid change of water content can lead to the rapid change of soil microstructure and increase the channels for radon exhalation. The water content change rate of $Na_2O \cdot nSiO_2−CaCl_2$ modified soil is

significantly reduced, which is helpful to improve the radon control performance of the soil.

(3) The microscopic mechanism of radon control in soil modified by $Na_2O \cdot nSiO_2 - CaCl_2$ is revealed. The formation of silicic gel and C−S−H gel improves the pore structure and permeability of the covering soil, and reduces the diffusion and migration of radon in the covering soil.

(4) The average radon exhalation rate of the $Na_2O \cdot nSiO_2 - CaCl_2$ modified covering soil is decreased by $1.01 \times 10^{-2}\,Bq/(m^3 \cdot s)$. The $Na_2O \cdot nSiO_2 - CaCl_2$ modified covering soil can effectively reduce the thickness of covering soil, improve the ability of soil cover to resist ambient temperature interference, and reduce the cost of covering treatment.

CRediT authorship contribution statement

Guo-kai XIONG: Data curation, Visualization, Investigation, Writing − Original draft; **Hong WANG:** Conceptualization, Investigation, Formal analysis, Data curation, Writing − Original draft; **Xiang-jiang WANG:** Conceptualization, Project administration, Theoretical analysis.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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放射性尾矿 Na₂O·*n*SiO₂−CaCl₂ 改性覆土降氡机理

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摘　要：通过尾矿覆盖控氡室内模拟实验，研究了 Na₂O·*n*SiO₂−CaCl₂ 改性土的控氡机理。氡析出率(J)与覆盖厚度(H)呈负相关，与温度呈非线性相关。覆土含水率变化速率明显变小，有助于减少土体损伤，提升覆土的抗环境温度干扰能力。硅酸凝胶和 C−S−H 凝胶的形成有效优化孔隙结构和渗透性，减少氡气在覆土中的扩散与迁移，其平均氡析出率下降了 $1.01 \times 10^{-2}\,\mathrm{Bq/(m^3 \cdot s)}$。研究结果表明，Na₂O·*n*SiO₂−CaCl₂ 改性覆土可有效提升覆土控氡性能，降低覆盖治理成本。

关键词：氡析出；C−S−H 凝胶；降氡机理；改性覆土

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