

Research Article

Effect of Colloidal Silicate on the Migration Behaviour of Strontium in Groundwater Environment of Geological Disposal Candidate Site

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Various colloids are present in the natural groundwater environment, and colloids act on the processes involved when radionuclides leak from a repository in a high-level waste disposal site. This paper investigates the effect of colloidal silicate in natural groundwater environments on the migration behaviour of Sr(II). Three different experimental cases have been designed: (1) effect in the presence of colloidal silicate, (2) effect in the presence of a porous medium, and (3) effect in the presence of both colloidal silicate and porous medium (referred to as CS, PM, and PC, respectively). Batch experiments were used to study the effect of influencing factors on Sr(II) migration behaviour, such as the amount of CS, solid-to-liquid ratio, pH, contact time, and initial concentration of Sr(II). The experiments showed that the effect of PC on the migration behaviour of Sr(II) was greatest, and the presence of CS enhanced the sorption. The colloid amount, pH, and solid-to-liquid ratio significantly affected the migration behaviour. The more the colloids were added, the better the adsorption effect. The optimal pH and solid-to-liquid ratio were 6 and 20 : 1, respectively. The alkaline environment is more conducive to colloid sorption. When the solid-to-liquid ratio was 20 : 1, the sorption percentage of PC is 0.5 times larger than PM. Although the PC has a longer adsorption equilibrium time, the percentage of adsorption can be larger than that in the other two cases. The kinetics and isotherms of Sr(II) were best described by the pseudo-second-order and Langmuir models. It was inferred that strong chemical interactions and/or surface complexation contributed primarily to Sr(II) sorption, and the process was on the monolayer adsorption of the outer surface. These findings provide valuable information for the migration behaviour of strontium in groundwater environments of geological disposal site. At the same time, it provides information for the implementation of permeable reactive barrier technology to control the transport of radioactive Sr(II) and its species in natural surface and groundwater.

1. Introduction

Since the development and utilization of nuclear energy, the nuclear industry has developed rapidly, and a large amount of nuclear waste has been created [1–3]. Presently, high-level waste is being managed by immobilizing it in solid form (glass and synroc), which is to be buried in deep geological repositories in the near future. However, over thousands of years, there is a finite probability of these minor radionuclides

entering the geosphere, through earthquakes, volcanic eruptions, leaching, and corrosion of the waste form, and underground water movement [4]. It is therefore imperative to study the migration behaviour of these minor radionuclides in the aquatic environment.

Strontium (Sr) is an important product of nuclear fission and is among the typical components of radioactive waste [5, 6]. Sr(II) has good mobility and a half-life of 28.8 years. Its migration speed in geological media is faster than many

actinide, fission, and transuranic nuclides [7, 8]. It is a regulatory object that needs special attention in geological disposal and has been ranked by the US Department of Energy as the third most important nuclear element that can be easily transferred to the environment [9–11]. The chemical properties of Sr(II) are similar to those of Ca or Ba. However, the ability of the human body to distinguish between Sr(II) and Ca(II) is not strong. Sr(II) can easily replace Ca(II) and accumulate in the human body, resulting in osteosarcoma, leukaemia, and other diseases [12, 13]. Therefore, there is high value in taking Sr(II) as the research representative to study the effect of colloids on the migration of nuclides in the groundwater environment for the geological disposal of radioactive nuclides.

At present, many studies have been conducted on the migration of heavy metal elements in aqueous solution or soil medium. Some materials, such as illite [14–16], attapulgite [17], natural clayey sandstone [18], kaolinite [19], montmorillonite [20], and Na-rectorite [21], may play an important role in the migration behaviour of heavy metal elements. Furthermore, colloids have been found to have a significant influence on the migration of radionuclides in the water environment. Yu et al. [20] and Zhao et al. [21] showed that the presence of HA enhances $^{90}\text{Sr}(\text{II})$ sorption on Na-montmorillonite at $\text{pH} < 7.0$ but reduces $^{90}\text{Sr}(\text{II})$ sorption at $\text{pH} > 7.0$. Feng et al. [22] found that the adsorption behaviour of arsenic on soils was strongly dependent on the concentrations of $\text{Fe}(\text{OH})_3$ and H_2SiO_3 colloid. Arsenic adsorption on soils increased significantly with decreasing H_2SiO_3 colloid concentration, but the effect of $\text{Fe}(\text{OH})_3$ colloid on the adsorption behaviour of arsenic in soils is complicated.

The common colloids present in natural water are based on oxides of Si, Al, and Fe [23, 24]. Ultrafiltration analysis confirmed that the elemental composition of colloids in the groundwater of the study area was Si, Fe, Na, etc. In the treatment of wastewater containing radionuclides, silica gel is often used as a sorbent [25]. Many studies have shown that silica gel has sorption properties for nuclides [26, 27]. Lu and Mason [28] found that the isotope exchange behaviour of radioactive Sr(II) onto montmorillonite and silica colloids is an important pathway for the transport of Sr(II) and its species in aqueous systems. At present, one of the important aspects, which is not clearly understood, is the effect of CS in groundwater environment on the migration behaviour of Sr(II). Various authors have reported the influence of colloids or a single component of PM on the migration behaviour of nuclides. Luo et al. [29] studied the sorption behaviour of V(V) onto natural soil colloids and found that the adsorption capacity has different changes with changing environmental conditions such as temperature and pH. Kaygun et al. [17] studied the adsorption behaviour of Sr and Cs onto attapulgite, and the adsorption efficiency was quite high at pH levels from 4 to 8. Therefore, research on the effect of colloids in groundwater environment on the migration behaviour of Sr can make up for this deficiency.

The effect of CS in natural groundwater environments on the migration behaviour of Sr(II) was studied by batch experiments, which explored the impact of various factors

(colloid amounts, solid-liquid ratio, pH, initial concentration of Sr(II), and contact time). The sorption mechanism, sorption kinetics, and isotherms of CS and PM on Sr(II) were discussed. The results provide information for the implementation of permeable reactive barrier technology to control the transport of Sr(II) and its species in natural surface and groundwater.

2. Materials and Methods

2.1. Materials. Study area and reagent source: the study area is in a preselected high-level waste disposal area in Beishan, Gansu Province, China. The sampling location is $41^\circ 49' 17''$ N and $97^\circ 00' 50''$ E. Soil and groundwater samples were collected from the area. Inductively coupled plasma mass spectrometry (ICP-MS) (NexION 300X, PerkinElmer Inc., USA) was used to examine the elements in groundwater. Inductively coupled plasma emission spectrometry (ICP-ES) (Spectro Arcos Eop, Spectro Analytical Instruments GmbH, USA) was used to detect the metal components in the soil samples. Nanosilica was purchased from Sinopharm Group Chemical Reagent Co., Ltd., China. Strontium nitrate was obtained from Tianjin Guangfu Fine Chemical Research Institute, China. Hydrochloric acid and sodium hydroxide were purchased from Beijing Beihua Fine Chemicals Co., Ltd., China. All chemicals used in this study were analytically pure.

Preparation of CS: 50 mg of nanosilica is taken into a 500 ml volumetric flask and dilute with groundwater to a final volume of 500 ml. It is placed into a sonicator for a 30 min ultrasonic bath to evenly disperse the sample and filtered through a $0.45\ \mu\text{m}$ membrane. The concentration of CS was $100\ \text{mg L}^{-1}$.

Preparation of Sr(II) stock solution: 1.2 g of $\text{Sr}(\text{NO}_3)_2$ is placed into a volumetric flask and diluted with groundwater to a final volume of 500 ml. The concentration of Sr(II) in the solution was determined to be $500\ \text{mg L}^{-1}$.

2.2. Batch Tests. The influencing factors including the amount of CS, solid-to-liquid ratio, pH, contact time, and initial concentration were investigated using the batch technique. The volume of the solution was 100 ml for all experiments (when the amount of colloid was 100 ml, the solution volume was 100 ml by default). In the process, the experimental time was 1440 min, the pH values of the solutions were carefully adjusted by either 0.1 M HCl or 0.1 M NaOH, and the solid-to-liquid ratio was controlled by adding different amounts of PM. The initial concentrations of Sr(II) varied from 0.5 to $1000\ \text{mg L}^{-1}$, and the amount of colloid varied from 0 to 100 ml. The variable quantity of Sr(II) was determined from the difference between the initial and final concentrations of Sr(II). Experimental conditions were changed by changing specific experimental parameters; other parameters remained unchanged. The different controlled conditions are shown in Table 1. Three groups of controls were designed for each part of the experiment. The experimental results were taken as the average of the three groups.

TABLE 1: Controlled and various factors of experiments.

Various factors	Colloid amounts (ml)	Contact time (min)	pH	Sr(II) initial concentration (mg L ⁻¹)	Solid-to-liquid ratio (g L ⁻¹)
Colloid amounts (ml)	10, 50, 100	1440	6	10	20
Contact time (min)	10	10, 20, 60, 120, 300, 600, 1440, 2880, 4320	6	10	20
pH	10	1440	3, 4, 5, 6, 7, 8, 9, 10	10	20
Sr(II) initial concentration (mg L ⁻¹)	10	1440	6	0.5, 1, 5, 10, 50, 100, 200, 500, 800, 1000	20
Solid-to-liquid (g L ⁻¹)	10	1440	6	10	1:1, 2:1, 5:1, 10:1, 20:1, 100:1

3. Results and Discussion

3.1. Composition of the Water and Soil Samples. ICP-MS was used to determine the trace elements in groundwater. The pH of groundwater was 7.3, and the chemical composition is summarized in Table 2. The purpose of this step is to further understand whether the composition of groundwater will have an unnecessary effect on the experimental results. The metal components in the soil samples were determined by ICP-ES. The chemical composition of the soil samples is summarized in Table 3. Similarly, the detection of chemical composition in soil samples is to determine whether there are silicic colloids in the samples to affect the experimental results.

3.2. Characterization of Soil Samples. In Figure 1, it can be seen that the loose porous mineral skeleton of the soil samples is basically bare, and the soil is mainly composed of small particles of approximately 150 nm in size. The mineral skeleton is covered and filled with the clay material, and it is difficult to separate with no certain shape. The surface micropores and pores are relatively numerous and have a strong adsorption effect on metals and radionuclides.

3.3. Effect of Colloid Amount. When PM worked alone, the amount of PM was 1 g, 5 g, and 10 g. When CS worked alone and PC worked, the volume of CS was 10 ml, 50 ml, and 100 ml, and the amount of PM was 1 g. The results are shown in Figures 2–4.

When CS worked alone, the equilibrium time of each addition amount to Sr(II) was approximately 10 h, which was not affected by the amount of CS. The sorption percentage was approximately 28%, 31%, and 36%, respectively. The reason may be that Sr(II) contacts with sorption sites on the surface of colloid [30]. First, there is the outer layer of complexation and ion exchange. Then, there is the inner complexation [30]. Ultimately, the sorption reaches equilibrium within 10 h.

When PM worked alone, the equilibrium time of each amount to Sr(II) was approximately 10 h, which was not affected by the amount of PM. The sorption percentage was approximately 33%, 41%, and 46%, respectively. When CS and PM worked together, the experimental equilibrium time did not vary with the amount of colloid and was always

TABLE 2: Chemical composition of groundwater.

Test items	Mass concentration (mg L ⁻¹)
K	6.42
Na	159
Ga	82.7
Mg	22.4
Fe	<0.005
Sr	1.39
Mn	0.0067
B	0.265
Ba	0.0685
Fluoride	0.0904
Chloride	223
Nitrate	1.82
Sulphate	524
CO ₃ ²⁻	0
HCO ₃ ⁻	182

TABLE 3: Chemical composition of soil samples.

Test items	Mass concentration (mg/kg)
K	20355.50
Na	13726.50
Ga	18111.80
Mg	3652.82
Al	49402.50
Fe	15279.00
Sr	114.58
Si	467.56

approximately 24 h. The sorption percentage was approximately 80%, 87%, and 91%, respectively.

By comparison, when CS interacted with PM, the equilibrium time was longer than CS or PM alone.

3.4. Effect of Solid-to-Liquid Ratio. The experiments were designed with solid-to-liquid ratios of 1:1, 2:1, 5:1, 10:1, 20:1, and 100:1 (g L⁻¹), and the amount of Sr(II) in the sample was 1 mg. With the presence or absence of CS, the results of three media on the sorption of Sr(II) are shown in Figure 5.

When the solid-to-liquid ratio was less than 20:1 and with increasing solid-to-liquid ratio, the amount of Sr(II) removal and the sorption percentage increased gradually.

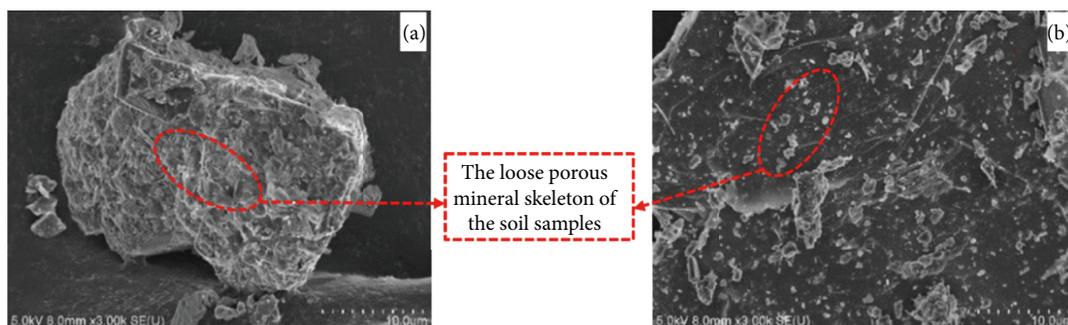


FIGURE 1: SEM of soil samples.

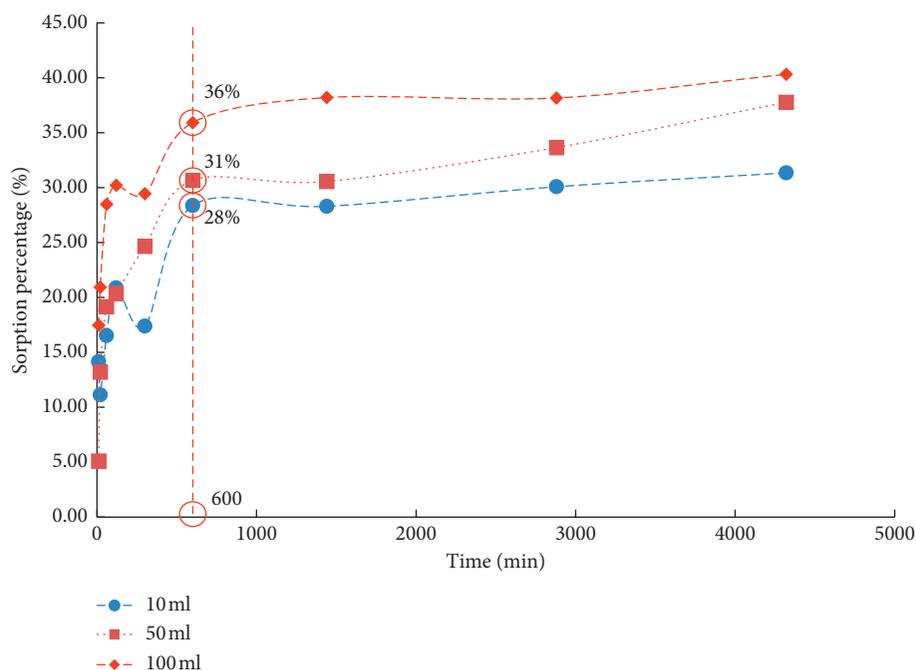


FIGURE 2: Effect of CS addition on equilibrium time: each of the red circles is the point where the experimental phenomenon changes significantly, and the data are the sorption percentage and time of the points.

When the solid-to-liquid ratio was greater than 20:1, the sorption tended to be stable. The sorption percentage affected by CS increased from 29% to 81%. When CS and PM worked together, the trendline was similar to PM alone. However, the effect of sorption increased by approximately 0.5 times. With the increasing solid-to-liquid ratio, the amount of PM in the system increased, as did the total amount of sorption sites [31, 32]. In other words, PM can provide a certain amount of sorption sites. Therefore, the sorption increased correspondingly in the solid phase media [33]. When the solid-to-liquid ratio was greater than 20:1, Sr(II) and the sorption points fully contacted, so the sorption percentage no longer increased.

3.5. Effect of pH. The designed pH levels of the experiment were 3, 4, 5, 6, 7, 8, 9, and 10, and the error was less than 0.1. The amount of Sr(II) in the sample was 1 mg. In each case, the effect of pH on the Sr(II) sorption is shown in Figure 6.

When CS worked alone and $\text{pH} < 6$, with the increasing pH, the sorption percentage increased from 11% to 36%. When $\text{pH} > 6$, the sorption percentage decreased to 14% with the increasing pH. In general, the neutral and alkaline environments are more conducive to colloid sorption [12, 34]. The reason may be that when pH is low, H^+ of higher concentration adsorbs competitively with Sr^{2+} , and the effect of sorption on Sr(II) is poor. With the increasing pH and the decreasing concentration of H^+ , competition decreases. The colloid plays a more important role, and the adsorption amount of Sr(II) increases. The alkaline environment can dissolve the colloid, which is not conducive to the colloid sorption [35]. At the same time, the concentration of OH^- in strong alkaline environment is higher, which reacts with Sr(II) in solution, forming precipitate of $\text{Sr}(\text{OH})_2$. Under the combined action, Sr(II) in solution is removed to a certain extent.

When PC worked and $\text{pH} < 4$, the sorption percentage of Sr(II) increased slowly from 40% to 60%. In general, the

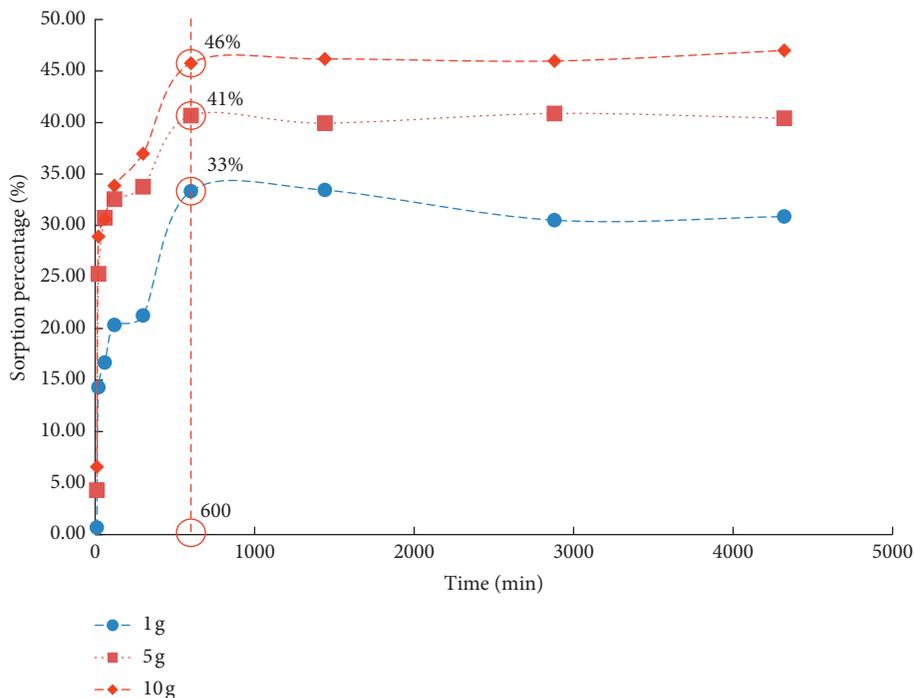


FIGURE 3: Effect of PM addition on equilibrium time: each of the red circles is the point where the experimental phenomenon changes significantly, and the data are the sorption percentage and time of the points.

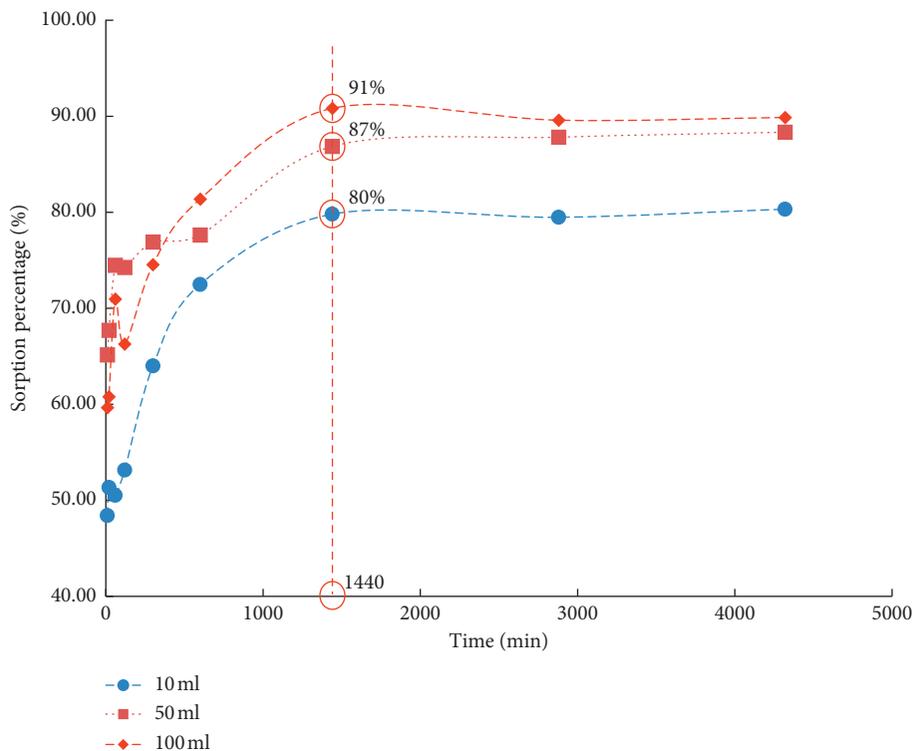


FIGURE 4: Effect of PC addition on equilibrium time: each of the red circles is the point where the experimental phenomenon changes significantly, and the data are the sorption percentage and time of the points.

alkaline environment is more conducive to colloid sorption. The reason is the same as above, but the difference is that PM may be complex with CS forming macromolecules to adsorb Sr(II) [36]. The effect of sorption is better.

In contrast, the sorption of CS and PM is strongly influenced by pH, and the effect of pH on the PC is relatively small. The combined effect increases by approximately 0.5 times relative to PM alone.

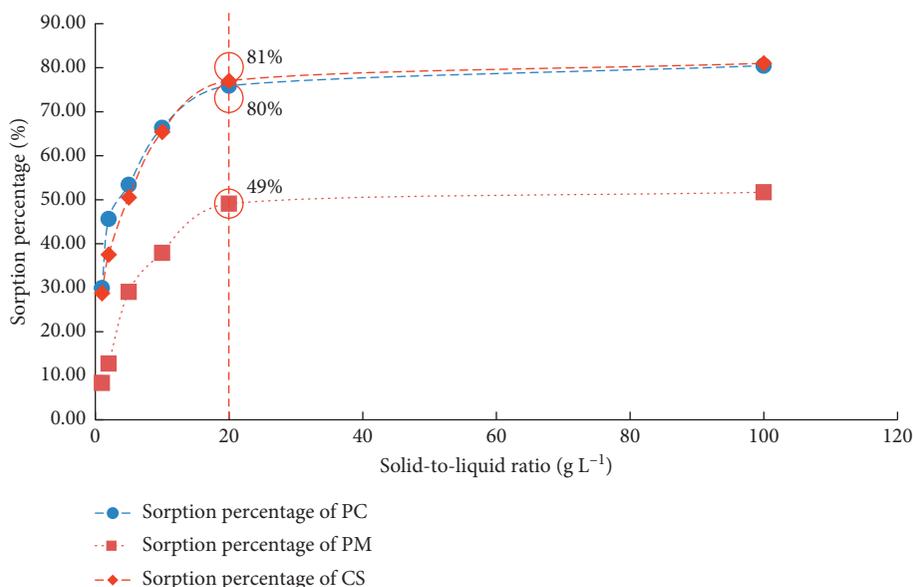


FIGURE 5: Sorption percentage changes with solid-to-liquid ratio.

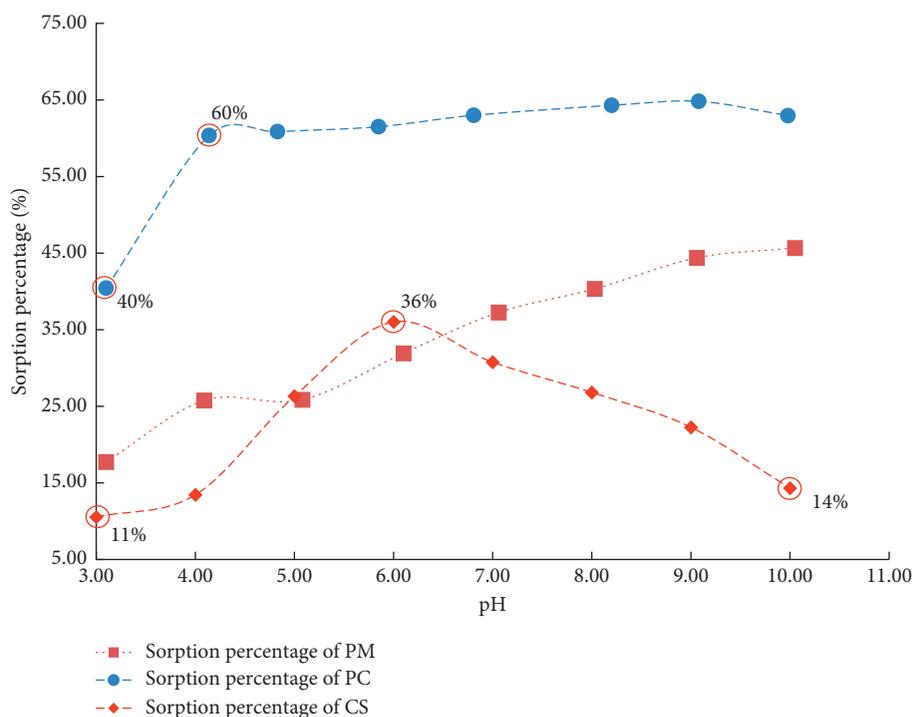


FIGURE 6: Sorption percentage changes with pH.

3.6. *Effect of Sorption Time.* Under the same experimental control conditions, the oscillation times were varied to 10, 20, 60, 120, 300, 600, 1440, 2880, 4320, and 5760 min per sample, and the Sr(II) element content was determined. In each case, the effect of experimental time is shown in Figure 7.

When CS worked alone, the sorption percentage of Sr(II) increased rapidly within 10 h and no longer increased after 10 h. At this time, it was up to sorption equilibrium. The sorption percentage increased from 14% to 31%. The reason

is that Sr(II) contacts with sorption sites on the surface of colloids. First, there is the outer layer of complexation and ion exchange. Then, there is the inner complexation [30]. Ultimately, the sorption reaches equilibrium within 10 h.

Under the combined action of CS and PM, the removal rate of Sr(II) and the sorption percentage increased rapidly within 10 h, increased slowly after 10 h, and no longer increased after 24 h. It tended to equilibrium. The sorption percentage increased from 48% to 70%. The reason is the same as above.

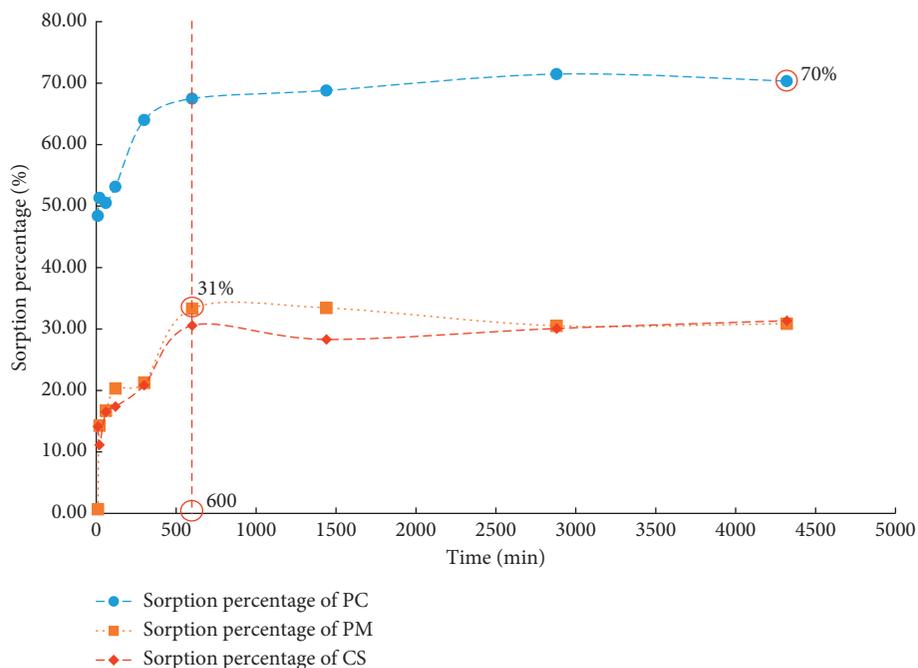


FIGURE 7: Sorption percentage changes with experimental time.

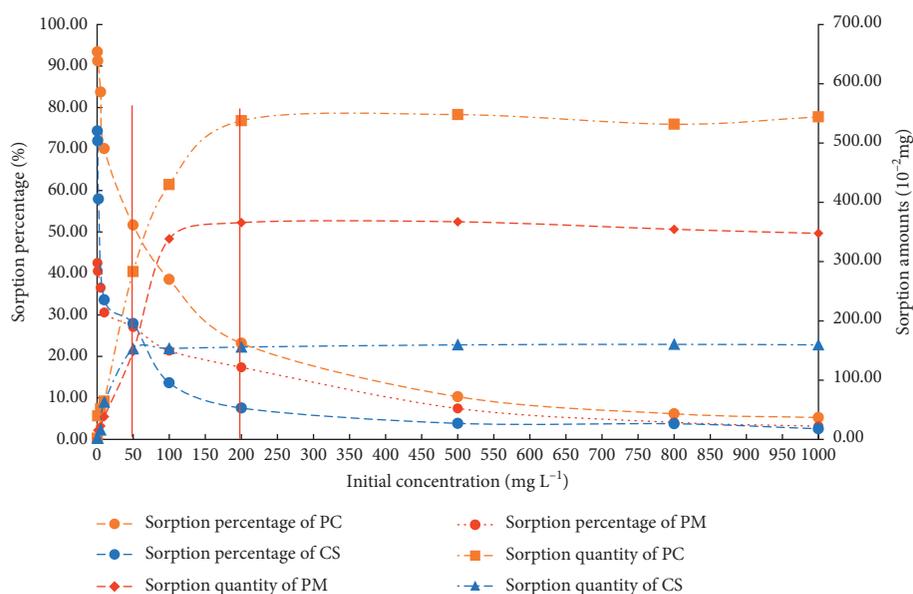


FIGURE 8: Sorption percentage and amounts of Sr(II) change with initial concentration.

By comparison, when PC alone worked, the equilibrium time was the same as CS alone and longer than PM alone. However, the sorption effect is approximately 2.3 times that of PM alone.

3.7. Effect of Initial Concentration. The initial concentration of Sr(II) was 0.5, 1, 5, 10, 50, 100, 200, 500, 800, and 1000 mg L⁻¹. The initial concentration of Sr(NO₃)₂ solution was determined after the sample was prepared. In each case, the effect of Sr(II) initial concentration is shown in Figure 8.

When CS worked alone and the initial concentration of Sr(II) was less than 50 mg L⁻¹, the sorption percentage of Sr(II) increased with increasing concentration. When the concentration was greater than 50 mg L⁻¹, the sorption percentage decreased from the initial 28% to 3%. As the initial concentration increases, the chance of Sr(II) and CS contacting increases and Sr(II) adsorption quantity increases [30]. At the same time, the competition between Sr²⁺ is strengthened, so the sorption percentage decreases [37].

Under the combined action of CS and PM, the adsorption quantity of Sr(II) increased rapidly with increasing

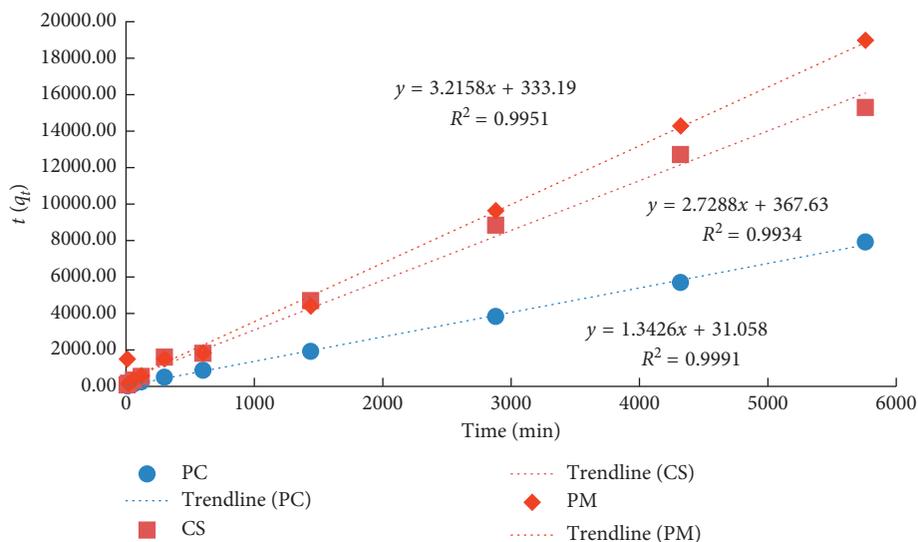


FIGURE 9: Pseudo-second-order kinetic model Sr(II) in the three media.

initial concentration when it was less than 200 mg L^{-1} . The reason is that as the initial concentration increased, the higher binding energy points were fully utilized. At the same time, the sites with lower binding energy also gradually functioned [37, 38]. When the initial concentration was greater than 200 mg L^{-1} , the sorption reached equilibrium. At this time, all points adsorbing Sr(II) in the solution could be in full use. The amount of sorption and desorption was balanced. As the concentration increased, the sorption percentage decreased from 93% to 5% and the competitive sorption between Sr^{2+} intensified.

The sorption effect when CS interacts with PM is obviously better than the cases of the two substances alone. When PC worked, the adsorption of Sr(II) increased by approximately 0.5 times relative to PM alone. With the presence of PM, the adsorptive saturation concentration increased from 50 mg L^{-1} to 100 mg L^{-1} . When PC worked, it was more suitable for a high-concentration Sr(II) environment than PM or CS alone.

3.8. Sorption Kinetics. To study the sorption mechanism of CS on Sr(II), the experimental data were processed by pseudo-first-order and second-order kinetic equations to describe the kinetic behaviour of the reaction.

The pseudo-first-order model is as follows [39]:

$$\ln(q_1 - q_t) = \ln q_1 - K_1 t. \quad (1)$$

The pseudo-second-order model is as follows [40]:

$$\frac{t}{q_t} = \frac{1}{K_2 q_2^2} + \frac{t}{q_2}, \quad (2)$$

$$h = K_2 q_2^2,$$

where K_1 (min^{-1}) and K_2 (min^{-1}) are the adsorption rate constants, which reflect the speed of adsorption and are proportional to the speed of the adsorption reaction; q_1 , q_2 , and q_t represent the amounts (mg/g) of Sr(II) adsorbed at

equilibrium and at time t , respectively; and h is the initial adsorption rate (mg/g-min).

Based on the results of the experimental data, we found that the correlation coefficients (R^2) fitted by the pseudo-second-order kinetic equation were larger. The results are shown in Figure 9. R^2 was greater than 0.99 in all three cases, reaching a significant correlation ($P < 0.0001$). We speculate that the sorption process is mainly chemical sorption [41].

3.9. Sorption Isotherm. Freundlich and Langmuir isotherm sorption equations were used to fit the sorption process of the three media on Sr(II).

The Langmuir isotherm sorption equation is as follows [42]:

$$\frac{q_e}{q_m} = \frac{K_L C_e}{1 + K_L C_e}. \quad (3)$$

The Freundlich isotherm sorption equation is as follows [37]:

$$q_e = K_F C_e^{1/n}, \quad (4)$$

where q_e is the sorption capacity of Sr(II) under different initial mass concentrations, mg/g; C_e is the mass concentration of residual Sr(II) in the solution at equilibrium sorption, mg L^{-1} ; q_m is the maximal saturated sorption capacity, mg/g; K_L is the Langmuir constant, mg^{-1} ; and K_F and n are empirical constants.

As can be seen from Figure 10, the correlation coefficients (R^2) of PC, PM, and CS adsorbed Sr(II) fitted by the Freundlich model are 0.9152, 0.93, and 0.7596, respectively. As shown in Figure 11, the comparison of R^2 between the Langmuir and Freundlich models showed that the Langmuir model exhibited a better fit for the experimental data than the Freundlich model. The Langmuir isotherm sorption equation assumes that the surface of the adsorbent is homogeneous and that there is no interaction between the adsorbates, and it is monolayer sorption. Sorption occurs only

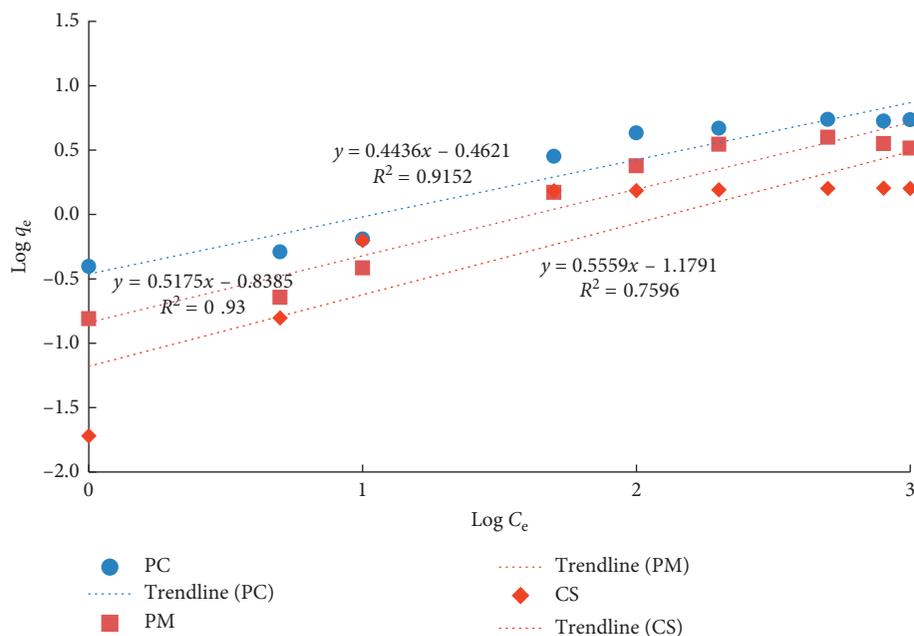


FIGURE 10: Freundlich model of Sr(II) in the three media.

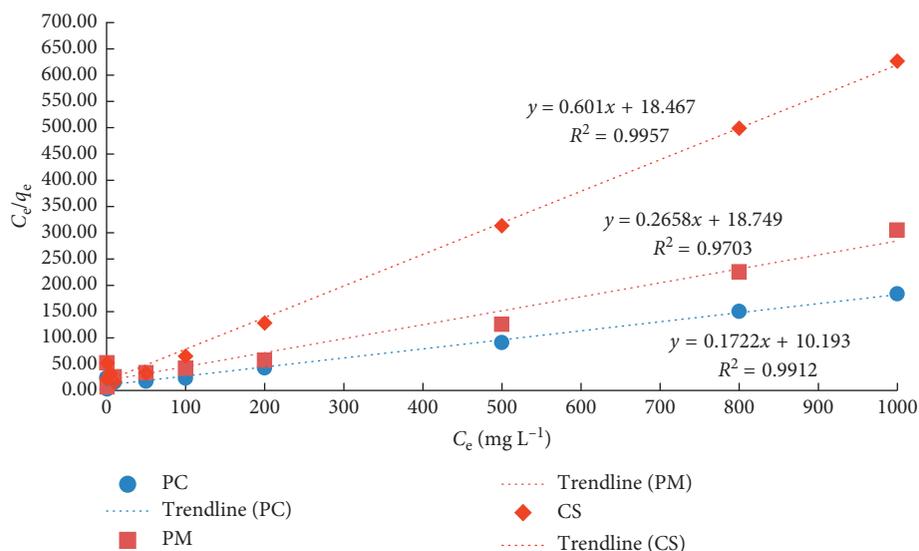


FIGURE 11: Langmuir model of Sr(II) in the three media.

on the outer surface of the adsorbent. The Freundlich isotherm sorption equation can be applied to both single-layer sorption and nonuniform surface sorption [36, 43]. In the present study, the results indicated that the binding sites of the sorbent were distributed uniformly on its surface and the adsorption of Sr(II) was regarded as monolayer adsorption. The characteristics of the adsorption isotherms in this study were consistent with those in previous reports [36, 43].

4. Conclusion

The results suggest that CS promoted the sorption of Sr(II) in groundwater environment. The adsorption amount of Sr(II) was highly dependent on the amount of CS, solid-to-liquid

ratio, and pH. The best solid-to-liquid ratio was 20:1. The sorption percentage reached a maximum at pH=6 and decreased as the acidity or alkalinity of the solution increased. The sorption time studies showed that PC adsorbed approximately 2.67 more Sr(II) than PM and CS. Moreover, the solid-to-liquid ratio and equilibrium times (24 h for PC and 10 h for PM and CS) revealed that PM was the dominant force for adsorbing Sr(II). The initial concentration studies showed that PC worked with a higher sorption saturated concentration (500 mg L⁻¹) compared with PM working alone (200 mg L⁻¹), which indicated that PC was more suitable for a high concentration of Sr(II). The sorption behaviour of Sr(II) followed the pseudo-second-order kinetic model and the Langmuir isotherm, demonstrating that

the adsorption of Sr(II) and the adsorbent was mainly chemical adsorption and formed a monolayer on the outer surface. This study confirmed that the presence of CS in groundwater environment promotes the adsorption of Sr(II), and the results provide information for the implementation of permeable reactive barrier technology to control the transport of radioactive Sr(II) and its species in natural surface and groundwater.

Data Availability

All of the data used in this article support the findings of the study.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Acknowledgments

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