Investigation of co-transport behavior of strontium and bentonite colloids in granite disposal environment

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Abstract

Colloids are prevalent in nuclear waste repositories, with bentonite colloids posing an uncontrollable risk factor for nuclide migration processes. In this study, static adsorption experiments were coupled with dynamic shower experiments to comprehensively investigate the influence of bentonite colloids on Sr2+ migration in granite, considering adsorption capacity. Bentonite colloids have a considerably greater adsorption capacity than both bentonite and granite, with a maximum adsorption of 30.303 mg/g. The adsorption behavior of bentonite colloids on Sr2+ is well described by the Langmuir isotherm and pseudo-second-order kinetic models, indicating that a single-layer chemical adsorption process is controlled by the site activation energy. The adsorbed Sr2+ is unevenly distributed on the colloids, and the adsorption mechanism may involve ion exchange with Ca. Bentonite colloids exhibit superior adsorption in neutral environments. The cations in groundwater inhibit Sr2+ adsorption, and the inhibition efficacy decreases in the order Fe3+>Ca2+>Mg2+>K+. The presence of bentonite colloids in a granite column slightly influences the retention of Sr2+ in the column while markedly reducing the Sr2+ penetration time from 70 h to 18 h. However, the coexistence of Co2+, Ni2+, and Cs+ in a multinuclide system weakens the ability of the colloids to promote Sr2+ migration. In comigration of colloid and multinuclide systems, the adsorption of nuclides by bentonite colloids causes the nuclide migration speed to decrease in the order Sr2+>Cs+>Ni2+>Co2+. This study provides insights into Sr2+ migration in cave repositories for low- and medium-level radioactive waste.

Full Text

Preamble

Investigation of Co-Transport Behavior of Strontium and Bentonite Colloids in Granite Disposal Environments

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Colloids are prevalent in nuclear waste repositories, with bentonite colloids posing an uncontrollable risk factor for nuclide migration processes. In this study, static adsorption experiments were coupled with dynamic column experiments to comprehensively investigate the influence of bentonite colloids on Sr^{2+} migration in granite, with particular consideration of adsorption capacity. Bentonite colloids exhibit a considerably greater adsorption capacity than both bentonite and granite, achieving a maximum adsorption of 30.303 mg/g. The adsorption behavior of bentonite colloids for Sr^{2+} is well described by the Langmuir isotherm and pseudo-second-order kinetic models, indicating that a single-layer chemical adsorption process is controlled by site activation energy. The adsorbed Sr^{2+} is unevenly distributed on the colloids, and the adsorption mechanism may involve ion exchange with Ca.

Bentonite colloids demonstrate superior adsorption in neutral environments. Cations present in groundwater inhibit Sr^{2+} adsorption, with inhibition efficacy decreasing in the order $Fe^{3+} > Ca^{2+} > Mg^{2+} > K^{+}$. The presence of bentonite colloids in a granite column slightly influences the retention of Sr^{2+} in the column while markedly reducing the Sr^{2+} breakthrough time from 70 h to 18 h. However, the coexistence of Co^{2+}, Ni^{2+}, and Cs^{+} in a multinuclide system weakens the ability of colloids to promote Sr^{2+} migration. In comigration systems involving colloids and multiple nuclides, the adsorption of nuclides by bentonite colloids causes nuclide migration speed to decrease in the order $Sr^{2+} > Cs^{+} > Ni^{2+} > Co^{2+}$. This study provides insights into Sr^{2+} migration in cave repositories for low- and medium-level radioactive waste.

Keywords: Granite; Radioactive cave disposal; Bentonite colloid

Introduction

Low- and medium-level radioactive waste is primarily disposed of in near-surface facilities. In recent years, there has been growing interest in rock cave disposal

to enhance safety. The Chinese 14th Five-Year Plan includes provisions for constructing the first rock cave disposal facility for low- and medium-level radioactive waste in Guangdong Province. The cave repository is situated in a granite layer with four barriers to prevent nuclide migration. Gaomiaozi bentonite serves as the backfill material, with granite as the surrounding rock [**?**].

However, barrier systems can fail under unforeseen circumstances such as extreme weather and geological activities during long-term disposal, enabling groundwater to transport nuclides such as ^{137}Cs, ^{60}Co, ^{90}Sr, and ^{131}I into the biosphere, thereby causing irreversible harm to the ecological environment [**?**]. Among these nuclides, ^{90}Sr is a prominent fission product of radioactive waste known for its solubility and mobility in water [**?**, **?**, **?**]. This nuclide is highly biologically toxic and can inflict considerable damage to bone marrow and bone tissue, increasing the risk of cancer and leukemia [**?**]. Therefore, close monitoring of the adsorption and migration of ^{90}Sr in repository barrier materials is essential for long-term safety assessments.

Colloids are typically particles with diameters of 1 nm to 1 µm suspended in a fluid [**?**]. Colloids have large specific surface areas and charged surfaces, resulting in high affinity for the vast majority of nuclides in repositories. The many types of colloids in the water environment of waste repositories include inorganic colloids derived from clay minerals, iron and aluminum oxides/hydroxides from the corrosion of waste containers, organic colloids formed from organic macromolecules and humic acid, and biological colloids such as bacteria and viruses. Bentonite is used as backfill material and is abundant throughout the repository. Consequently, bentonite colloids are the primary colloidal species in nuclear waste repositories [**?**]. Bentonite colloids are produced by underground erosion and soaking of bentonite. However, the subsequent continuous loss of bentonite and degradation of engineering barriers [**?**] threaten the blocking capacity of the repository' s engineering barrier system. Bentonite colloids have large specific surface areas and can chemically adsorb or form complexes with nuclides, thereby transporting nuclides and affecting their migration behavior in the repository. Several studies have confirmed that bentonite colloids significantly affect the migration of radionuclides in repositories [**?**, **?**, **?**, **?**].

Consequently, considering the impact of bentonite colloids is crucial for assessing repository security. Granite serves as the final barrier in a repository, bridging the engineered barrier and the biosphere. Bentonite colloids transporting radionuclides through the engineered barrier come into direct contact with granite. During this interaction, bentonite colloids may be retained in the granite regardless of radionuclides, thereby penetrating the granite and enabling nuclides to be retained in the granite. Bentonite colloids affect nuclide migration in granite through a complex and reversible process that may have different effects on different nuclides. Numerous studies have shown that the presence of bentonite colloids may diminish the ability of surrounding rock barriers to block nuclides. Katezirina et al. [**?**] investigated the migration behavior of ^{137}Cs on fractured granite in the presence of bentonite colloids and found that bentonite

colloids transport a small portion of ^{137}Cs through the granite. Similarly, Zheng et al. [**?**] investigated the impact of gibbsite and sodium bentonite colloids on U(VI) transport in saturated granular granite columns, finding that the strong binding affinity of U(VI) to the colloids enhanced U(VI) transport.

However, the interactions and cotransport modes of colloids and nuclides may also inhibit or eliminate the effect of bentonite on nuclide migration in granite [**?**]. Elo et al. [**?**] conducted column experiments and showed that mobile bentonite colloids have a negligible effect on Np(V) migration in granite. By contrast, Timothy et al. [**?**] found that Am(III) transport by bentonite colloids is governed by the desorption of Am(III) from specific binding sites on the colloids, with bentonite colloids weakly promoting Am(III) transport. Most studies on bentonite colloids have focused on either the adsorption properties of colloids or the direct impact of colloids on nuclide migration. However, few experiments have been conducted on joint adsorption and migration to comprehensively investigate the influence of bentonite colloids on nuclide migration in granite in terms of adsorption capacity.

Hence, in this study, strontium (Sr) was selected as the target nuclide, sampled from the surrounding granite and backfill material (bentonite) in the first cave repository for low- and medium-level radioactive waste in China. The physical and chemical properties of granite and bentonite in the area were characterized, and bentonite colloids were produced using local groundwater. Batch adsorption experiments were performed to compare the adsorption of Sr^{2+} on bentonite colloids, bentonite, and granite. The mechanism of Sr^{2+} adsorption onto bentonite colloids was elucidated by combining empirical models with characterization methods. Accordingly, dynamic migration experiments were conducted to investigate the impact of bentonite colloids on the migration behavior of Sr^{2+} in granite columns, with particular focus on the influence of a multinuclide system on Sr^{2+} migration. This study provides insights for accurate estimation of the potential risk of Sr migration in cave repositories for radioactive waste.

Materials and Methods

A. Geological Materials

Granite samples were obtained from granite rock formations near a low- and medium-level radioactive waste repository in Guangdong Province. The samples were extracted from a depth of 173 m with a specific surface area of 2.54 m^2/g. The bentonite samples were obtained from Gaomiaozi, Inner Mongolia and had a measured specific surface area of 38.278 m^2/g. The elemental and oxide compositions of both granite and bentonite samples were determined using an Axios series X-ray fluorescence spectrometer (XRF). The findings are presented in Table 1 . Groundwater samples were extracted from boreholes proximal to the cave repository at depths ranging from 148 to 173 m. During sampling, the groundwater had a flow rate of 41.5 L/min, conductivity of 298.5 µS/cm, and pH of 6.7. Groundwater samples were analyzed for major cation

and anion compositions using inductively coupled plasma-mass spectrometry (ICP-MS) and ion chromatography, respectively, and the results are shown in Table 2 .

TABLE 1: Oxide mass percentage of granite and bentonite | Element(%) | SiO_2 | Al_2O_3 | K_2O | Fe_2O_3 | CaO | Na_2O | MgO | |———|—|——|——|——|——|——|——| | Granite | 74.02| 15.56 | 1.82| 2.31 | 1.25| 1.50 | 2.86| | Bentonite | | | | | | | |

TABLE 2: Chemical composition of groundwater near the cave repository | Element($mg \cdot L^{-1}$) | Al^{3+} | Ca^{2+} | Mg^{2+} | K^+ | Si^{4+} | Mn^{2+} | Fe^{3+} | $SO_4{}^{2-}$ | $NO_3{}^-$ | Cl^- | |————-|—|—|—|—|—|—|—|—|—|—|

B. Experimental Instruments and Materials

Transmission micrographs of the samples were obtained using a field-emission transmission electron microscope (FETEM) (Libra200FE, Zeiss, Germany). Electron micrographs were obtained using a high-resolution cold-field emission scanning electron microscopy/energy dispersive X-ray spectroscopy (SEM-EDS) (Ultra55, Zeiss, Germany). The Zeta potentials of the samples were measured using a potentiometer (ZetaPALS, Brookhaven, USA). The mineral compositions were determined by an X-ray diffractometer (XRD) (TD-3500, Panaco, Netherlands). Functional group compositions were determined using a Fourier transform infrared (FT-IR) absorption spectrometer (Frontier, Perkin Elmer, USA). Colloid mass concentrations were measured using a gravimetric method combined with ultraviolet (UV) spectrophotometry (V-5800, Yuanyi, China). The residual Sr^{2+} concentration in solution was measured by ICP-MS (iCAP6500, Thermo Fisher, USA).

Hydrochloric acid (HCl) was obtained from China West Long Science Co., Ltd. (China). Sodium hydroxide (NaOH, 99%), ferric chloride ($FeCl_3$, 99%), calcium chloride ($CaCl_2$, 99%), magnesium chloride ($MgCl_2$, 99%), and potassium chloride (KCl, 99%) were obtained from Tianjin Zhiyuan Chemical Reagent Co., Ltd. (China). Cesium chloride (CsCl, >99%) and strontium chloride hexahydrate ($SrCl_2 \cdot 6H_2O$, >99%) were procured from Aladdin Reagent Factory (China), and cobalt chloride hexahydrate ($CoCl_2 \cdot 6H_2O$, >99%) and nickel chloride hexahydrate ($NiCl_2 \cdot 6H_2O$, >99%) were purchased from Macklin Reagent Company (China). All chemical reagents used in this study were of analytical grade and were not further purified before use.

C. Preparation of Bentonite Colloids

Ten grams of bentonite powder (200 mesh) were weighed and dispersed in 1 L of ultrapure water with a resistivity of 18.25 $M\Omega \cdot m$. The powder was dispersed by ultrasonic agitation for 30 min, followed by magnetic stirring for 4 h to ensure uniform mixing. The mixture was allowed to settle at room temperature for two days. The resulting sample was centrifuged at 4000 rpm for 15 min to remove particles larger than 1 μm. The separated solution was transferred to

a dialysis bag for purification. The dialysis water was periodically replenished, and the pH and conductivity were monitored using pH and conductivity meters, respectively. The conductivity of the solution decreased from 160 µS/cm to 10.58 µS/cm during dialysis, indicating that the colloidal dispersion had low ionic strength and therefore an appropriate dispersion coefficient. The prepared colloidal samples were stored in a sealed conical flask at room temperature until further use.

The colloidal concentration was determined using the differential gravity method combined with UV spectrophotometry, following the method used in our previous study [**?**]. A colloidal sample solution with a desired concentration gradient was prepared based on the mass concentration obtained using the differential gravity method. The full spectrum of the colloidal solution was scanned using a UV-visible spectrophotometer, and the characteristic wavelength of the solution was determined to be 240 nm. The colloid mass concentration was determined by generating a standard curve ($R^2 = 0.999$) from the data.

D. Adsorption/Desorption Experiments

Batch adsorption experiments were conducted on two groups: a powder (S1) and colloidal (S2) group, to investigate the adsorption behavior of Sr^{2+} on geological barrier materials and colloids, respectively. Each experiment was repeated three times, and the average measurement was recorded. For Group S1, several portions of granite (Gt) and bentonite (Bt) powder were weighed and placed in polypropylene centrifuge tubes, to which 4.5 mL of deionized water was added. Group S2 was prepared by adding a bentonite colloidal (BC) solution to the centrifuge tubes. Samples from both groups were equilibrated overnight at room temperature. Subsequently, 4.5 mL of Sr^{2+} solution was added to each sample to achieve the desired nuclide concentration. In experiments investigating the influence of specific ions, 4.5 mL of a mixed solution of Sr^{2+} and the respective ions were added to the sample. The pH values of the samples were adjusted using trace quantities of hydrochloric acid and sodium hydroxide. The samples were placed in an oscillating box and agitated until adsorption equilibrium was reached. The equilibrated samples were centrifuged at 10,000 r/min for 30 min to separate the solid and liquid phases. The supernatant of the Group S1 samples was directly diluted, and the residual Sr^{2+} concentration was determined using ICP-MS. To ensure that the bentonite colloids were completely separated from the adsorbed solution, the supernatant of the S2 group was passed through a 0.1 µm filter membrane before the residual Sr^{2+} concentration in the liquid phase was determined.

A desorption experiment was conducted after completion of the adsorption experiment. The suspension that remained at the end of the adsorption experiment was removed to yield a granite sample with adsorbed Sr^{2+}. An equal volume of solution was added to the sample at the same pH, and the mixture was shaken under the same experimental conditions as those used for the adsorption experiment to reach desorption equilibrium. The sample was centrifuged, filtered, and

analyzed using ICP-MS to determine the Sr^{2+} concentration in the supernatant.

E. Migration Experiment

The migration experiment was performed using a dynamic column with a length of 20 cm and diameter of 2.5 cm, which was filled with a soil column with an actual length of 14 cm. The experimental setup is illustrated in Figure 1 [Figure 1: see original paper]. Coarse-grained granite (60-80 mesh) was wet-packed into the columns. Nylon nets were placed at both the upper and lower ends of the column to prevent the granite powder from escaping during the filling process. The packed granite was continuously washed with an upward flow of ultrapure water at a constant rate until water flow equilibrium was reached in the column to ensure stable moisture content throughout the experiment.

The Sr^{2+} migration experiment was performed on the equilibrated column in two stages: transport and elution. Prior to conducting the experiment, a Sr^{2+} solution (Sr^{2+} and ionized water), BC-Sr^{2+} solution (bentonite colloid, Sr^{2+}), and mixed solution (bentonite colloids, Sr^{2+}, Cs^{+}, Co^{2+}, Ni^{2+}) were prepared, each with a nuclide concentration of 100 mg/L along with a bentonite colloid concentration of 250 mg/L, to simulate the expected near-field range of spent fuel storage. During the transport stage, the prepared solution was continuously added to the lower end of the granite column at a fixed flow rate of 4 mL/h via a peristaltic pump. Samples were periodically withdrawn and diluted for testing until the nuclide concentration stabilized. During the elution stage, ultrapure water was continuously pumped into the granite column at a constant flow rate of 4 mL/h. Regular sampling and testing were conducted until the simulated nuclide concentration in the detection solution approached zero.

F. Data Analysis

The adsorption capacity for Sr^{2+} (q) was determined for granite, bentonite, and bentonite colloid. The migration of Sr^{2+} in the granitic media was analyzed by calculating the longitudinal diffusion coefficient (D), which is expressed as:

$$C_0 - C_e V = L/t_{0.5} \left(\frac{L - V t_{0.16}}{t_{0.16}} \frac{L - V t_{0.84}}{t_{0.84}} \right)^2$$

The adsorption data obtained at different contact times were fitted using pseudo-first-order and pseudo-second-order kinetic models and the Weber-Morris (W-M) internal diffusion model to obtain kinetic information on mineral and colloid surface adsorption, which was subsequently used to infer the adsorption mechanism. The kinetic models are provided below [**?**, **?**]:

Pseudo-first-order model:
Pseudo-second-order model:
Weber-Morris model:

where q and q are the unit adsorption capacity for Sr^{2+} by the sample at time t and at adsorption equilibrium, respectively ($mg \cdot g^{-1}$); k_1 is the pseudo-first-order rate constant (d^{-1}); k_2 is the pseudo-second-order rate constant ($g \cdot (mg \cdot d)^{-1}$); k d is the W-M model rate constant ($d^{-1}/^2$); and C is the influence constant of boundary layer thickness on adsorption ($mg \cdot g^{-1}$).

G. Adsorption Model

The adsorption data obtained at different Sr^{2+} concentrations were fitted using the Langmuir, Freundlich, Temkin, and Dubinin-Radushkevich (D-R) isotherm models. The Langmuir isotherm model assumes single-layer adsorption and that the attraction between molecules decreases sharply as the distance between molecules increases. The Freundlich isotherm model is based on adsorption with an uneven distribution of sites on the adsorbent surface. The Temkin model assumes that adsorption energy decreases linearly with increasing surface coverage [**?**]. The D-R isotherm model can be used to calculate the average free energy of adsorption and determine the adsorption type. The models are expressed as follows [**?**, **?**]:

Langmuir:
Freundlich:
Temkin:
D-R:

where C_0 and C are the mass concentrations of Sr^{2+} in the initial solution and at adsorption equilibrium, respectively ($mg \cdot L^{-1}$); m is the mass of the sample; g is the gravitation constant; and V is the sample volume in mL used in the adsorption experiment.

In the D-R equation, E denotes the energy required for 1 mol of nuclide to react with the adsorbent surface. The parameters are defined as:

$$q_e = \frac{k_F C_e^{1/n}}{1 + k_L C_e}$$

$$q_e = q_m \frac{k_L C_e}{1 + k_L C_e}$$

$$q_e = k_T \ln(f) + k_T \ln(C_e)$$

$$\ln(q_e) = \ln(q_m) - \beta\varepsilon^2$$

$$E = \varepsilon = RT \ln\left(1 + \frac{1}{C_e}\right)$$

where q is the maximum adsorption capacity for Sr^{2+} of the sample ($mg \cdot g^{-1}$); k is the adsorption constant of the Langmuir model; k_F and 1/n are the adsorption constants of the Freundlich model; k_T and f are the constants of the Temkin model (k_T is related to adsorption heat; f is the maximum binding energy of adsorption); R is the molar gas constant ($8.3145\ J \cdot (mol^{-1} \cdot L^{-1})$); T is the absolute temperature (K); β is a constant related to the adsorption energy ($mol^2 \cdot kJ^{-2}$); is the Polanyi adsorption potential ($kJ \cdot mol^{-1}$); and E is the average free energy of adsorption ($kJ \cdot mol^{-1}$).

Results and Discussion

A. Characterization

1. XRD and FT-IR The mineral compositions of the samples were determined by XRD and FT-IR analyses. Figure 2 Figure 2: see original paper shows the XRD patterns of the samples. The predominant mineral components of granite are quartz, albite, potassium feldspar, and biotite, with albite and K-feldspar together constituting approximately 70% of the mass percentage of the granite minerals. Bentonite contains montmorillonite as the main mineral component as well as secondary minerals such as quartz, illite, and albite (Figure 2(b)). The interplanar spacing of the bentonite colloid at the (001) plane differs slightly from that measured for the soil sample (1.52 nm). The diffraction curve of the bentonite colloid contains only a montmorillonite peak, indicating that secondary minerals were removed by centrifugation and dialysis [**?**].

FT-IR analysis of the surface functional groups revealed that granite mainly consists of Si- and Al-based functional groups, including O-H, Si-O, Al-O, and Si-O-Si, which are similar to those present in bentonite [**?**, **?**]. The bentonite colloids and bentonite have similar primary functional groups with minimal differences in the corresponding peak shapes and positions in the FT-IR spectra. This suggests that colloid formation has a negligible effect on the surface functional groups and chemical bonds of bentonite.

2. Zeta Potential and Particle Size Distribution The Zeta potential of the geological materials and bentonite colloids at different pH values is shown in Figure 3 Figure 3: see original paper. Granite reaches the zero-point charge at a pH of approximately 5.30, whereas bentonite and bentonite colloids have negative zeta potentials across the pH range of 3-11, with bentonite colloids having a slightly lower potential than bentonite. The negative charge of bentonite arises from both the permanent charge of the structure and variable edge charge [**?**]. The colloid preparation process can affect the deprotonation of -SiOH and -AlOH groups, resulting in bentonite colloids with slightly lower potential than bentonite. However, the charges of bentonite and bentonite colloids were similar, indicating that the contribution from edge charge was small.

Figure 3(b) shows the average size and size distribution of the colloidal particles determined by dynamic light scattering. The colloidal particle sizes ranged

from 100.9 nm to 433.5 nm with an average of 209.1 nm, suggesting relatively good colloidal stability. The polydispersity index (PDI) is a crucial parameter for characterizing particle size uniformity in a system. The bentonite colloid sample had a PDI of 0.217 in ultrapure water, indicating excellent dispersion.

3. TEM and SEM-EDS Analysis The TEM images of the samples are shown in Figure 4 [Figure 4: see original paper]. Granite exhibits a massive shape with a dense internal texture. Both bentonite and bentonite colloids display similar sheet morphologies with irregular edges, and the layered structure of the colloid is more pronounced, confirming the XRD results. In addition, the TEM images illustrate that the prepared bentonite colloid has good dispersion, and the particle size is approximately 200 nm, which is consistent with the particle size distribution test results.

The surface morphologies of the samples before and after adsorption were characterized by SEM-EDS. Figure 5 [Figure 5: see original paper] shows that granite samples have a fragmented distribution with unevenly sized layered fragments, and the sample surface is smooth with angular features. There is no significant difference in the morphology of granite before and after adsorption, although adsorption makes the surface rougher and more fragmented, possibly due to immersion and agitation. The EDS results for samples after adsorption indicate an increase in the quantity of adsorbed Sr^{2+} on the granite surface; however, there is no discernible distribution pattern for Sr^{2+} or direct correlation between the distributions of Sr^{2+} and other ions. The nonuniform particle size of bentonite indicates a distinct flaky structure with numerous irregular pores and cavities that facilitate the migration and diffusion of nuclides [**?**]. After adsorption, the bentonite surface curls and becomes rough, presumably due to drying after adsorption [**?**].

The immersion and dialysis of bentonite generates bentonite colloids with a distinct layered morphology [**?**] with clearer boundaries and more regular distribution than that of the soil samples. Adsorption does not considerably change the morphology of the colloids but makes the layered structure denser, more compact, and agglomerated, with less distinct interlayers. The EDS results presented in Figure 6 [Figure 6: see original paper] show that O, Si, Al, and Mg remain the main elements in the colloidal samples after simulated nuclide adsorption. The Sr^{2+} ions are adsorbed unevenly on the colloids, mainly accumulating in the layered stacking regions, indicating the presence of different adsorption sites on the colloids, with stacking edge sites possibly having stronger affinity for Sr^{2+} [**?**]. The distribution of Sr on the colloids appears correlated with that of Ca, suggesting that Ca in the colloids might participate in Sr adsorption via an ion exchange reaction between Ca and Sr. Bentonite colloids primarily consist of montmorillonite, and Missana et al. [**?**] observed similar phenomena for Sr adsorption onto Na-montmorillonite.

B. Adsorption Behavior of Sr^{2+}

1. Adsorption Kinetics The results of the kinetic experiments are presented in Figure 7 [Figure 7: see original paper]. The adsorption capacities of the three geological materials for Sr^{2+} decrease in the order: bentonite colloid (BC) > bentonite (Bt) > granite (Gt). The adsorption equilibrium time for Sr^{2+} at five days is notably shorter for Gt than for BC and Bt, possibly due to differences in the number and specificity of surface adsorption sites among the materials. The pseudo-second-order kinetic equation fits the adsorption data for Gt with a high correlation coefficient ($R^2 = 0.998$), suggesting that Sr^{2+} adsorption by Gt is primarily chemical [**?**]. The fitting curve for the Gt data based on the W-M model has three distinct regions corresponding to surface diffusion, internal diffusion, and equilibrium, indicating that intraparticle diffusion is not the sole rate-controlling step for Gt adsorption.

In contrast, BC rapidly reaches adsorption equilibrium within 1 h, with q increasing from 2.136 mg/g to 13.989 mg/g, indicating a considerably greater adsorption capacity than that of Bt. This result is attributed to the larger specific surface area and lower surface charge of BC compared with Bt, leading to an increased number of adsorption sites and enhanced ability to capture Sr^{2+} ions. Both BC and Bt data were most accurately fitted by pseudo-second-order kinetics, suggesting a chemical adsorption process involving surface complexation between functional groups and ion exchange [**?**]. Fitting the adsorption data of BC and Bt using the W-M equation revealed a two-stage process, unlike the adsorption on Gt. The fitted BC and Bt curves do not pass through the origin, implying that Sr^{2+} adsorption is primarily determined by the adsorption sites and that the activation energy for adsorption and internal diffusion plays a minor role in the process [**?**]. Overall, the adsorption capacity of bentonite surpassed that of granite, suggesting potential synergy between the two materials in blocking nuclide diffusion. However, the strong adsorption capabilities of bentonite colloids introduce considerable uncertainty and risk factors for long-term repository disposal.

2. Effect of Nuclide Concentrations The concentrations of nuclides carried in groundwater fluctuate with natural precipitation and water flow, which can deteriorate the blocking performance of barrier materials. Figure 8 [Figure 8: see original paper] shows that BC has the highest adsorption potential for Sr^{2+}, whereas the adsorption capacities of Gt, Bt, and BC for Sr^{2+} all increase with Sr^{2+} concentration, gradually tending toward equilibrium. This phenomenon may be attributed to the low probability of Sr^{2+} binding to sites at low concentrations, with the probability increasing with Sr^{2+} concentration, which facilitates capture of Sr^{2+} by adsorption sites. Consequently, the saturation of binding sites decreases, resulting in a slower increase in adsorption capacity and gradual approach to equilibrium [**?**].

To further explore the adsorption mechanism, we used the Langmuir, Freundlich, Temkin, and D-R isotherm models to fit the adsorption data at different Sr^{2+}

concentrations. Compared with the Freundlich and Temkin models, the Langmuir adsorption model fit the Gt, Bt, and BC data with higher correlation coefficients of 0.996, 0.991, and 0.989, respectively, indicating that Sr^{2+} is mainly adsorbed as a surface monolayer [**?**]. Within the experimental concentration range, the maximum adsorption capacities of Gt, Bt, and BC for Sr^{2+} were 1.229 mg/g, 4.407 mg/g, and 30.303 mg/g, respectively, and were positively correlated with the specific surface areas of the materials. The fit of the Gt, Bt, and BC data to the Temkin model also showed high linearity, suggesting strong intermolecular interactions between the adsorbent and adsorbate [**?**]. As the Freundlich constant, 1/n, is less than 1, adsorption occurs easily. In the D-R equation, E denotes the energy required for 1 mol of nuclide to react with the adsorbent surface. Table 4 shows that Gt, Bt, and BC have E values above 16 kJ/mol; therefore, the adsorption mechanism of Gt, Bt, and BC is a chemical process [**?**], which is consistent with the kinetic fitting results.

3. Effect of pH The pH of the environment strongly influences mineral surface potential and ion species distribution, which affects ion adsorption on minerals [**?**, **?**]. Figure 9 Figure 9: see original paper shows the impact of pH on Sr^{2+} adsorption. The adsorption of Sr^{2+} by Gt, Bt, and BC is impeded by both acidic and alkaline environments. The adsorption capacities of BC, Bt, and Gt peak at pH 6, displaying values of 13.303 mg/g, 2.014 mg/g, and 0.733 mg/g, respectively. This phenomenon may be attributed to the combined effects of strontium species distribution, zeta potential, and colloidal stability. Figure 9 Figure 9: see original paper shows that Sr^{2+} is the primary strontium species under acidic conditions, where competition between excess H^+ and Sr^{2+} for adsorption sites hinders adsorption [**?**]. Consequently, adsorption is generally inhibited in acidic environments. As pH increases, complexes form between OH^- and Sr^{2+}, such as $Sr(OH)^+$, which hinder adsorption. Therefore, the adsorption of Gt, Bt, and BC is impeded in alkaline environments. The poor stability of bentonite colloids in acidic environments results in a greater inhibitory effect on BC than on Gt or Bt [**?**, **?**].

Zeta potential affects the affinity of the adsorbent for nuclides. The pH at the zero point of charge (pH_{ZPC}) for Gt is approximately 5.30. At pH values below 5.30, the Gt surface is positively charged, which impedes Sr^{2+} adsorption via electrostatic repulsion. Conversely, at pH values > 5.30, the Gt surface becomes negatively charged, facilitating adsorption. Given that the permanent structural charge of Bt and BC is negative, the zeta potentials of these materials remain negative within the experimental pH range and decrease with increasing pH, indicating that the affinity of Bt and BC for Sr^{2+} increases with pH. However, this behavior does not alter the inhibition of adsorption by Bt and BC under alkaline conditions. This result can be explained by the minor role that charge attraction plays in adsorption by Bt and BC. The pH of groundwater near disposal facilities is typically approximately 6.70. Around this pH, BC demonstrated high Sr^{2+} adsorption activity. Therefore, considering the potential effect of colloids on Sr^{2+} migration during disposal is important.

4. Effect of Ions The complex disposal environment for radioactive waste can result in groundwater containing numerous ions that affect nuclide adsorption by colloids and barrier materials. Preliminary analysis of the groundwater solution showed that the primary anions and cations near the disposal site were CO_3^{2-}, SO_4^{2-}, Fe^{3+}, Ca^{2+}, Mg^{2+}, and K^+. Given the potential of Sr^{2+} to form insoluble precipitates with anions such as CO_3^{2-} in groundwater, various electrolytes (KCl, $FeCl_3$, $CaCl_2$, and $MgCl_2$) were used to investigate Sr^{2+} adsorption behavior in the presence of cations. Figure 10 [Figure 10: see original paper] shows that coexisting cations inhibit Sr^{2+} adsorption by silicate minerals. These inhibitory effects were consistent for BC, Bt, and Gt, with effectiveness decreasing in the order $Fe^{3+} > Ca^{2+} > Mg^{2+} > K^+$. BC was inhibited the least due to its high unit adsorption capacity, whereas Gt displayed the most sensitivity to coexisting cations. Typically, high-valence cations compete more effectively for adsorption sites than low-valence cations [**?**], which accounts for Fe^{3+} having a more pronounced inhibitory effect than K^+.

The difference between the inhibitory effects of Ca^{2+} and Mg^{2+} arises from the difference in the radii of the corresponding hydrated ions. The smaller the radius of a hydrated ion, the more strongly the ion competes with Sr^{2+} for adsorption. Given the same positive charge, the hydrated ion radius of Ca^{2+} (0.200 nm) is smaller than that of Mg^{2+} (0.310 nm), leading to a greater inhibitory effect for Ca^{2+} than Mg^{2+}.

C. Migration Behavior of Sr^{2+}

1. Effect of Bentonite Colloid on Migration Batch adsorption experiments confirmed the robust adsorption capacity of BC for Sr^{2+}, motivating us to perform dynamic column experiments to further investigate the influence of BC on Sr^{2+} migration in the granite system. Figure 11(a) [Figure 11: see original paper] shows a breakthrough curve in which neither the peak values of BC-Sr comigration nor Sr migration alone reach 1, indicating retention of Sr^{2+} in the granite column. The longitudinal dispersion length (D) measured during Sr-BC comigration was $4.27\$\times10^{\{-4\}}.Penetrationofthegranitecolumncommencedat18h,whichwasconsiderablyshorterthan70hforSrmigrationalone,indic$ transport. This effect can be attributed to colloidal adsorption and inherent migration. Both bentonite colloids and granite exhibit negative charges across a broad pH range, resulting in repulsion between the colloids and granite during nuclide transport. Static adsorption experiments revealed that the adsorption of Sr^{2+} by BC is robust, establishing BC as a carrier for accelerating Sr^{2+} transport. Many studies [**?**, **?**, **?**, **?**] have corroborated that bentonite colloids strongly promote the migration of Eu(III), Pu(IV), U(VI), and other nuclides.

Retention of Sr^{2+} in the granite column was observed in the migration experiments. To investigate the influence of colloids on Sr retention in granite, experiments were conducted on the adsorption/desorption of Sr^{2+} on BC and Gt. As shown in Figure 11(b) [Figure 11: see original paper], the adsorption results indicate that the BC-Gt system has strong adsorption capacity for Sr^{2+}. Under

alkaline conditions, the adsorption capacity of the BC-Gt system is considerably greater than that of the Gt system alone because the stability of BC is enhanced in an alkaline environment [**?**]. The desorption experiments revealed that Gt has a lower adsorption capacity for Sr^{2+}, with most sites being reversible [**?**], resulting in a low retention rate (15.1% to 35.6%) for Sr^{2+} in Gt after desorption, which is consistent with observations from the migration experiments. The retention of Sr^{2+} on Gt was slightly higher for the BC-Gt system, attributed to the small number of colloids attached to the granite surface. Notably, acidic conditions considerably increased Sr^{2+} retention, which can be attributed to the reduced stability of BC under acidic conditions [**?**], leading to sedimentation and adhesion with Gt, which weakens desorption. Overall, fractures in granite significantly increase the risk of Sr^{2+} leakage into the biosphere, and a small amount of colloid adhering to the granite surface in the repository may slightly enhance Sr^{2+} retention in granite. However, colloid promotion was not observed to have a significant impact on Sr^{2+} retention in granite during dynamic migration experiments. This indicates that the influence of colloids on Sr^{2+} retention in granite should be considered alongside multiple factors such as flow rate, concentration, and pH, which are crucial for safety assessment of nuclear waste disposal facilities.

2. Comigration of Bentonite Colloids and Polynuclides As shown in Figure 12(b) [Figure 12: see original paper], the adsorption capacity of bentonite colloids for nuclides in multicomponent systems increased in the order $Co^{2+} < Ni^{2+} < Sr^{2+} < Cs^{+}$, indicating that BC has the strongest affinity for Cs^{+} in a multicomponent system. Analysis of the breakthrough curve revealed that the detection time for Sr^{2+} in the multinuclide comigration system surpassed that of the BC-Sr system. This delay may arise from interactions between numerous positively charged ions and bentonite colloids, mitigating electrostatic repulsion and reducing colloidal fluidity to promote bentonite agglomeration. These agglomerates were retained in the column due to size exclusion effects. Similar outcomes have been reported in studies on coexisting ions, where fluctuations in ion content were attributed to external interference. In particular, the coexistence of ions with the same valence state causes steric hindrance, and filtering exerts more pronounced inhibitory effects on colloidal transport [**?**]. The effluent recovery rate increased in the order $Co^{2+} < Ni^{2+} < Sr^{2+} < Cs^{+}$ due to the nuclide adsorption capacity of BC. Nuclides adsorbed on BC can be retained in granite columns, diminishing nuclide recovery rates in the effluent. Differences in affinities and binding capabilities of the colloids for different simulated nuclides resulted in Sr^{2+} and Cs^{+} eluting faster than Co^{2+}, which had a more pronounced elution curve. Nuclides with stronger affinity for BC were more affected during migration. The penetration times of nuclides in the multielement system increased in the order $Sr^{2+} < Cs^{+} < Ni^{2+} < Co^{2+}$. Hence, considering the role of bentonite colloids is important for predicting Sr^{2+} migration in safety assessments of repositories.

Conclusion

In this study, the impact of bentonite colloids on Sr^{2+} migration in granite was investigated in terms of adsorption capacity by conducting a combination of static adsorption and dynamic column experiments. The influence of the multinuclide system on Sr^{2+} migration was analyzed. The findings provide fundamental data and rational recommendations for safety assessment and establishment of migration models for nuclear waste disposal facilities.

The static adsorption experiments demonstrate that bentonite colloids have considerably greater Sr^{2+} adsorption capacity than bentonite and granite, with maximum adsorption rates of 30.303 mg/g, 4.407 mg/g, and 1.2285 mg/g for bentonite colloids, bentonite, and granite, respectively. The adsorption behavior of Sr^{2+} conformed to both the Langmuir isotherm and pseudo-second-order kinetic models. Fitting the Sr^{2+} adsorption data with the W-M model revealed a two-stage distribution, indicating that single-layer chemical adsorption is controlled by site activation energy. The distribution of Sr^{2+} on the colloid surface is nonuniform, and the mechanism of Sr^{2+} adsorption may involve ion exchange with Ca. Colloids have optimal Sr^{2+} adsorption capacity in neutral environments. However, cations in groundwater inhibit Sr^{2+} adsorption onto colloids, with inhibition efficacy decreasing in the order $Fe^{3+} > Ca^{2+} > Mg^{2+} > K^{+}$. Colloids maintain substantial adsorption potential even at high Sr^{2+} concentrations.

Dynamic migration experiments revealed that the presence of colloids influences the retention of Sr^{2+} in granite, consistent with findings from adsorption/desorption experiments. However, the addition of colloids considerably accelerated the diffusion rate of Sr^{2+} in granite, reducing the breakthrough time from 70 h to 18 h. The presence of Co^{2+}, Ni^{2+}, and Cs^{+} in a multinuclide system diminished the ability of colloids to promote Sr^{2+} migration. The effluent recovery rate increased in the order $Co^{2+} < Ni^{2+} < Sr^{2+} < Cs^{+}$ due to the nuclide adsorption capacity of BC. This result confirms that the stronger the colloidal adsorption capacity for a nuclide, the greater the impact of colloids on Sr^{2+} migration. In conclusion, the presence of colloids increases the risk associated with Sr^{2+} migration in disposal facilities. Therefore, the influence of colloids on Sr^{2+} migration must be considered in safety assessments.

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