Sorption of strontium and fractal scaling of the heterogeneous media in a candidate VLLW disposal site

Jinsheng Wang · Rui Zuo · Yanguo Teng · Qinhong Hu · Zongjian Sun

Abstract Because of the deposit and accumulation from the debris flow, the heterogeneous geological characteristics is obvious for a candidate very low level waste (VLLW) disposal site, with the grain size ranging from tens of microns to 75 cm. Therefore, it is challenging to directly measure the sorption capacity of the media and the distribution coefficient of some radionuclides, such as strontium. We have studies the correlation of the particle mass content with different grade size and the sorption capacity, which is important in the modeling of radionuclide migration in the heterogeneous disposal site. A total of three deep pits and five shallow trenches were excavated, and 21 solid samples were collected for laboratory experiments. The grade and percentage of the different-sized particles were obtained, and the fractal dimension (D) of the media was calculated from the results of sieved experiments. Steady state sorption time and sorption isotherm of strontium was determined in the heterogeneous media, and sorption and distribution of strontium in the heterogeneous media were evaluated by the relationship between the mass percentage and distribution coefficient (K_d) of the fine-particle media, which was comprised of selected particles with a diameter less than 1 mm, and the correlation on the K_d and D was regressed fit. The results indicated that fractal dimension bounded from 2.39 to 2.62 in the media, and K_d values of strontium ranged between 119 and 126 in the fine-particle media, and corresponding value was 11 and 43 in the original media. The correlation between K_d and D was approximately linear.

Keywords Heterogeneous media · Waste disposal · Fractal dimension · Distribution coefficient · Strontium

Introduction

Near surface geological disposal is widely used for disposing the very low level radioactive waste (VLLW). On the process of safety of an underground VLLW repository, the detailed understanding of the hydrogeological feature and sorption characteristics of radionuclides in variously porous media is of great importance. In the subsurface system, radionuclide transport is governed by two main classes of processes, water flow and sediment erosion, transport and deposition [1], so it is essential to study the characteristics of the hydroeological condition of the repository field site and the sorption behavior of the nuclide.

The homogeneous fine-particle media, like clays, is of particular importance for the use as backfill materials in waste repository design because of their high sorption capacities. In addition, argillaceous formations are particularly suitable as host rock formations, and ensure the radioactive nuclide not to migrate to the environment in the performance period of the disposal repository. However, the subsurface media in general are heterogeneous and often anisotropic [2]. Therefore, to establish reliable hydrogeological parameters in the heterogeneous media is a complex task and cannot be achieved on the basis of a few in situ borehole tests or a limited laboratory test programs on drill cores. Furthermore, due to high costs,

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conventional geotechnical projects do not allow an extensive core testing program regarding the relationship between intrinsic rock properties (such as porosity, microcrack density, permeability and foliation) and the physicochemical in situ conditions (such as effective stress and formation fluids).

Therefore, a series of analysis methods of parameter in heterogeneous media were attempted to develop and apply [3–5]. In these methods, fractal theory and approach was well used for dealing with the scaling of irregular and heterogeneous systems. The fractal models are available for the fragmentation of classical aggregates, aggregates with fractal pore space and fractal surfaces, and they offers a new opportunity for modeling the fragmentation process [6]. Fractal models can also describe hierarchical systems and complex natural phenomena, and are suitable to model soil structure and soil hydraulic properties [7]. For example, Xu et al. [8] studied the correlation between the pore-size distribution and the grain-size distribution, and found that the fractal dimension of the grain-size distribution could be used to determine the unsaturated hydraulic conductivity, instead of the fractal dimension of the pore-size distribution. Millán et al. worked with 140 PSDs (particle-size distributions) to study the relationship between scaling exponents and soil texture (clay and/or sand content), and compared the goodness-of-fit of traditional Weibull and normal distributions, and the proposed piecewise fractal function. They concluded that PSDs could present different behavior depending on soil texture [9]. Lu et al. [10] employed an indicator simulation approach to generating large-scale facies distributions, and a monofractal model, fractional Brownian motion to generate the permeability increments within facies.

The geometry of fractals and the mathematics of fractal dimension have provided useful tools for a variety of scientific disciplines, among which is chaos [11]. However, in a geological radioactive waste disposal site, the sorption of radionuclide on the media is among the principal aspects to be evaluated for the repository performance. Radionuclide migration in porous media can be influenced by the distribution of grain-sizes in heterogeneous media. The distribution coefficient (K_d) is the key parameter for modeling of nuclide migration and evaluation of nuclear environmental safety, because it represents the extent of radionuclide migration in porous media.

Strontium is an important component of radioactive waste. It has two important isotopes: ^{90}Sr that emits β radiation with a half-life of 29 years and ^{85}Sr that is a γ emitter with a half-life of 64.8 days. The behavior of strontium isotopes in the soil, which may be discharged to the environment as a result of nuclear weapons testing or incidents occurring at reprocessing plants, is of considerable interest. Nuclide ^{90}Sr, with its relatively long half-life,

is considered to be the more critical isotope of strontium element, which has a propensity to be strongly retained within the living organisms, mainly in the bones as a source of long-term radiation of bone marrow [12, 13]. In the geological systems, the mobility of strontium depends on its adsorption and desorption behavior, it is mainly adsorbed onto clay minerals, organic matter, as well as oxides of iron, manganese and aluminium [14–17].

Results of various researches have been reported in regard to the strontium adsorption behavior on many kinds of media, such as kaolinite, bentonite and chlorite-illite and some natural clays involving these minerals [18, 19], the ratio of mineral mixtures has already been considered [13]. During the last 30–40 years a lot of sorption experiments with Sr have been performed in different nuclear waste disposal programs, it has been described in the former articles by authors [20, 21], here, we focused on the relation of sorption of strontium and heterogeneous media.

Some publications have been published to describe the micro-pore media sorption and the fractal analyse. For example, the adsorption process that depended mainly on grain size of the solid and the type of soil had been tested [22], and Rudzinski et al. found that there was a functional relationship between the differential distribution of adsorption energies and the differential distribution of pore sizes on such "partially correlated" surfaces. They also showed that the differential pore size distribution reduces to the classical fractal pore size distribution in the limit of very small pore sizes, or when the fractal dimension D approaches 3 [23]. Lee et al. [24] also studied the effects of the grain size on the exchanging cation on the pore structure and surface fractality of montmorillonite. However, the effect of the heterogeneous media in field sites on strontium adsorption is hard to measure and control, it has generally been considered as a defined value like in the homogeneous media, the various law or trend of the nuclide sorption with the variety of the medium structure often was ignored in the calculated process. In a word, the relation of the fractal dimension of the media and the distribution coefficient of nuclide is not clear.

In China, all of the VLLW is required to be disposed in the area near the radioactive waste production; it is called the principle of "vicinity" [25]. In the area of the deposit and accumulation from the debris flow in the Southwest of China, it is difficult, and yet necessary, to select the location of candidate repository. The site has the characteristics of abundant rainfall, ecological sensitivity, proximity to the river, and complex hydrogeological condition. Among these factors, the worst one is the heterogeneous nature of porous media, with the grain sizes ranging from 75 cm to $n \times 10^{-2}$ mm.

In the heterogeneous VLLW disposal site, we develop a simple attempted approach to describing the correlation of

the fractal analysis of the media and the strontium sorption. This approach reduces to the classical fractal approach in the limit of small pore radii, and it is shown that there exists a functional relationship between sorption and surface heterogeneities. By applying that relationship, it is found that the generalized pore size distribution developed here from the fractal approach becomes the generalized adsorption distribution. In that way a bridge is built between the distribution coefficient of the nuclide sorption and the fractal heterogeneous surfaces of the media.

Experimental approach

The VLLW disposal site

The candidate VLLW disposal site is located at the central part of the alluvial-pluvial fan in the Southwest of China, which is on a narrow but long accumulation of the old debris flow from the northeastern mountain. The stratum age is Q_3, deduced from the relative height (4–8 m higher than the present riverbed) and original geological data. The front of the alluvial–pluvial fan is at the interchange of two streams while the repository is designed on the end close to the mountain (as shown from Fig. 1), the width of the top front is about 20 m. The upper reaches of the disposal site is valley as the channel of the debris flow, but the lower reaches is the accumulation area with the maximum width of about 200 m. The thickness of the fan body is about 10–20 m, with a gradual trend of increasing thickness from the front to the end.

From top to the bottom, there are four divided layers of the fan body. A thin slope washing layer with the thickness of 3–5 m is in the surface, is mixed with clay and gravel form the weathering bedrock. The depth of the second layer is 1–3 m with the lithology of loose larger gravel with a maximum diameter of 0.35 m. The third layer is mixed with the tight gravel and clay ata thickness of 4–10 m. The bottom layer is composed of the sand-mixed crushed stone with a diameter of 0.5–0.75 m. Below the bottom layer, the bedrock of tight Permian sandstones, which could be considered as the water-impermeable layer. All of the information of the layers could explain that some alluvial or pluvial actions frequently occurred in the geological

Fig. 1 The VLLW disposal site location and the sampling points
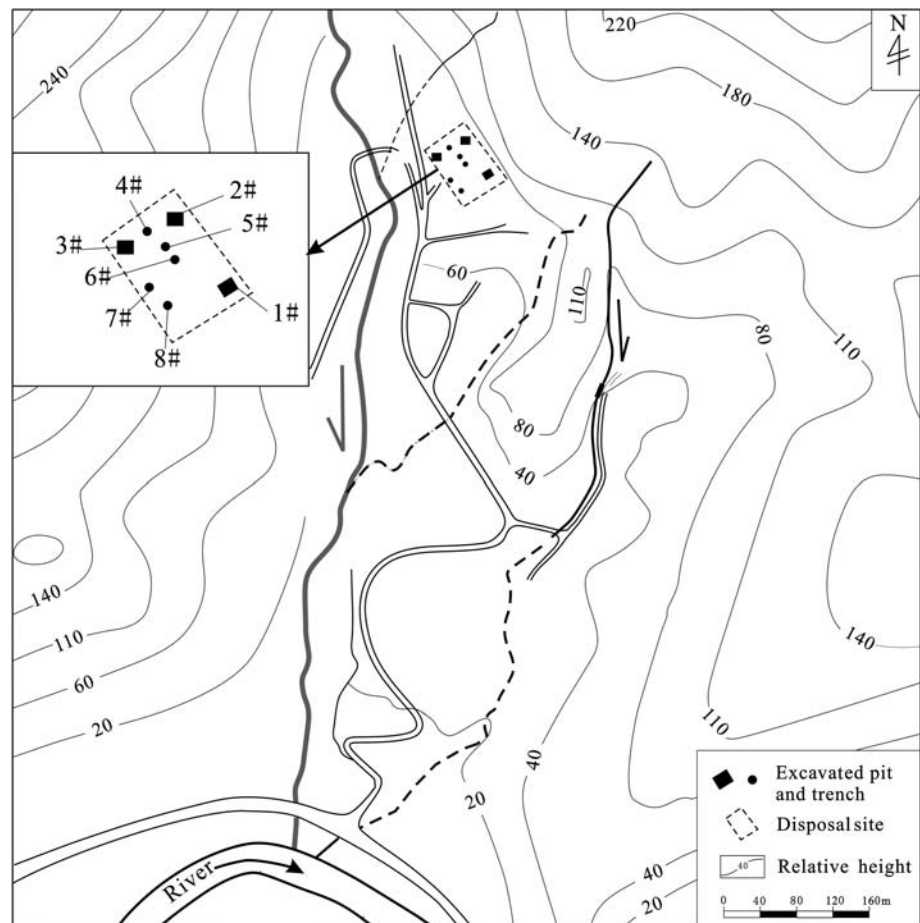
history. In the fan area, the groundwater table is in the third layer with the approximate depth of 10 m below the ground surface.

Samples

For the VLLW disposal site, because of the deposition and accumulation from the debris flow, the heterogeneous character is obvious. Furthermore, the thickness of the alluvial–pluvial layer is generally about 10 m while the maximum depth is less than 20 m, so the drilling pit and trench is suitable for the geological survey, and collection of groundwater samples. Three deep pits and five trenches were designed in the field disposal site. The pits, numbered as 1#, 2# and 3#, on the opening of 3×3 m, were required to dig and excavate until the groundwater table, supporting structures were emplaced to protect the stability of the pits. For the five trenches, serial number of 4#,…, 8#, the depth of 2–3 m was desired for the samples. All of the sites of the pits and trenches were shown in Fig. 1.

All of the groundwater and soil samples were collected from the candidate VLLW disposal site. For the excavated pit 1# with the depth of 9.6 m, six deposit soils were sampled at the depth of 1.5 m (thereafter named as 1–1), 2.8 m (1–2), 4.5 m (1–3), 5.7 m (1–4), 7.3 m (1–5), and 9.2 m (1–6), and the pit 2#, the sampling sites were at 1.5 m (2–1), 2.4 m (2–2), 3.6 m (2–3) and 4.8 m (2–4), about the pit 3#, the sites were at 2.1 m (3–1), 3.8 m (3–2), 5.3 m (3–3), 7.1 m (3–4), 8.5 m (3–5) and 9.3 m (3–6). For the trenches 4–8#, only one representative sampling was operated in every trench. All of the groundwater samples were sampled out of shallow groundwater from the excavated pits in experimental field at June 2005. Groundwater samples were collected following "Technical Specifications for Environmental Monitoring of Groundwater in China" [26], the results for groundwater and soil samples were presented in Table 1.

Theory

Distribution coefficient (K_d)

Distribution coefficient (K_d) represents the distribution ratio of solute in the liquid and solid phases. The process of distribution and sorption is dynamic and complicated in the groundwater and geological anisotropic media. For a quantitative evaluation of solute migration in geological media, all of the distribution processes is broadly defined as sorption, which is represented by K_d.

The distribution coefficient, K_d, is calculated by means of the following formula:

Table 1 Result of the groundwater and soil analysis

Groundwater data		Soil data	
Constituent	Data (mg/L)	Element	Abundance (%)
CO_3^{2-}	0	Al_2O_3	14.30
HCO_3^-	119.72	Fe_2O_3	12.63
Cl^-	1.40	CaO	3.03
Ca^{2+}	52.07	MgO	2.92
Mg^{2+}	6.91	K_2O	2.00
SO_4^{2-}	62.4	Na_2O	2.59
$K^+ + Na^+$	3.08	P_2O_5	0.76
HPO_4^{2-}	<0.07	MnO	0.12
F^-	0.19	TiO_2	2.25
Total hardness	158.47	FeO	0.66
pH	7.6		
Electrical conductivity (μs/cm)	330		

$$K_d = \left(\frac{C_0}{C} - 1\right)\frac{V}{M} \tag{1}$$

where K_d is the distribution coefficient (mL/g); C_0 and C are the initial and the steady state sorption concentrations in the liquid phase (mg/L); V is the volume of the liquid (mL); M is the mass of the fine-particle soil (g).

Fractal dimension (D)

Fractal scaling is valid at microscopic scale for several types of geological porous media. To investigate the impacts of fractal scaling upon hydraulic properties of porous media, a fractal representation of a porous medium is developed. The fractal dimension of the pore-size distribution can be obtained from the slope of the linear regression in the plane of the $\lg(W/W_T)$ and $\lg\overline{R_i}$. The fractal dimension of the pore size distribution is deduced and calculated as:

$$\frac{W(r < \overline{R_i})}{W_T} = \left(\frac{\overline{R_i}}{R_{max}}\right)^{3-D} \tag{2}$$

where r is the different diameter of the sieve, $\overline{R_i}$ is the average of the diameters of the adjacent order sieves (R_i and R_{i+1}), W_T is the total mass of the sieved sample, R_{max} is the maximum diameter of the sieved particle.

Then, the relative curve of the $\lg(W/W_T)$ and $lg\overline{R_i}$, and b that is the slope of the fitting line could be determined, and the fractal dimension D is:

$$D = 3 - b \tag{3}$$

The value of fractal dimension spans a large range from 1.0 to 3.0 [27, 28].

Experimental procedure

Sample sieving

All of the 21 deposit soil samples were oven-dried at 105 °C for 2 h, and were cooled to room temperature. Following the method "standard for soil test method in China" [29], these samples were then sieved by passing 60, 40, 20, 10, 5, 2, 1, 0.5, 0.25, 0.1, and 0.075 mm sieves to obtain the mass distribution of different particle diameters. Although soils are complicated assemblage of different particles that usually stick together, no additional attempt was made to determine the presence of small soil particles aggregated to a huge particle, because it is too difficult to control, and nowadays, no detailed operation could be found in the existence publications.

Batch tests

Being representative heterogeneous media, the mixed soil of the unsaturated zone at the VLLW disposal site was comprised of fine clay, gravel and crushed stone, so it is difficult to get an accurate K_d value. We tried to determine the approximate K_d value of the actual media by the relation of the measured K_d of the fine-particle with diameter <1 mm from screen separation and the fractal dimension of the porous media.

Batch tests determined the sorption and partition of Sr under different particle diameters of 1–0.5, 0.5–0.25, 0.25–0.1, 0.1–0.075, and less than 0.075 mm, and the initial Sr concentration is 10 mg/L. The solid–liquid ratio was 1:2, and temperature was 20 °C.

Reagent chemical of $Sr(NO_3)_2$, with the stable isotope of the strontium, was used for the experiment. Owing to the same basic chemical properties between the stable isotope and the radioactive isotope, their sorption in the soil followed the same law. Since strontium element is a long-lived nuclide, the concentration change as a result of radionuclide decaying could be neglected in the short-term experiment, stable strontium can be substituted for radiostrontium in batch tests to determine K_d [30]. The concentration of Sr^{2+} was determined by ICP-AES (inductively coupled plasma atomic emission spectroscopy with Perkin-Elmer 3300 DV, the detection limit of Sr is 0.00004 mg/L).

In the process of strontium sorption reaction, maybe some other reaction, e.g., precipitation reactions, would be existed. Some experiments have found that, in solution system, the precipitation reaction of Sr^{2+} easily appeared under the condition of CO_3^{2-} [31]. Hwang et al. [32] experimented that $SrCO_3$ precipitation was occurring from pH 6.5 when the nanofiltration operation was done in an open carbonate system with an equilibrium dissolved CO_2 concentration of 10^{-5} M. In these tests, because of little constituent of CO_3^{2-} (as shown at Table 1), the precipitation of strontium in the solution system has been ignored.

Results and discussion

Fractal dimension

According to experimental data of the particles diameters from sieving, and following the formula (2) and (3), the fractal dimension D in the unsaturated porous media could be calculated. The percentage of the different diameter particle and the fractal dimension D of each deposit soil sample were shown at Table 2.

Sorption characteristics

The batch-test samples were divided into Test A, B and C. The aim of these tests was to obtain the steady state sorption time of the sorption of strontium in the media (Test A), the sorption isotherm (Test B), and the distribution coefficient of Sr in the media under different initial conditions (Test C).

Steady state sorption time (Test A)

Five soil samples, chosen by sieving with particle sizes of 1–0.5, 0.5–0.25, 0.25–0.1, 0.1–0.075 and less than 0.075 mm from sample 1–2, were used to determine the changing trend and steady state sorption time of the K_d value of Sr with a long period of shaking time (21 days); the testing time was chosen at 2nd, 4th, 7th, 9th, 14th and 21st day. All sorption experiments were performed in duplicate, and the uncertainty is 6%.

Figure 2 showed the relation of the sorption distribution coefficient of Sr and the contact time of the solid and the liquid, which showed the time for Sr sorption to reach steady state.

The results showed that Sr distribution coefficient initially increased with an increasing contact time, and tended to stabilize after a certain time. Figure 2 showed a distinct sorption inflection point at about 7 day's contact time. From the results, it could be seen that the K_d value obviously bounded before the steady state sorption time of 7 days, after which the sorption of Sr arrived the steady state. Therefore, for the subsequent experiments, shaking contact time would be longer than 7 days in order to assure the steady state. From a study on steady state sorption time of Sr in urban clay solution, a minimum time period of more than 6 days was necessary to reach apparent steady state was also observed by Reala et al. [33]. In addition, other results indicated there was different viewpoint of the steady state sorption time in different media [34, 35].

Table 2 Distribution of the soil particle diameters and fractal dimension

| Sample site		The percentage of the different particle diameter (mm)												D	r^2
		>60	60–40	40–20	20–10	10–5	5–2	2–1	1–0.5	0.5–0.25	0.25–0.1	0.1–0.075	<0.075		
1#	1–1	1.50	8.19	17.17	21.87	17.37	14.80	1.88	7.63	3.52	2.93	1.98	1.16	2.410	0.966
	1–2	3.05	11.48	8.18	15.43	12.00	12.16	2.35	11.80	7.83	6.47	4.22	5.01	2.615	0.942
	1–3	6.03	4.77	12.97	15.98	11.90	11.51	2.04	12.59	7.77	6.39	3.21	4.83	2.602	0.944
	1–4	9.93	10.84	25.33	17.21	11.25	9.67	1.49	6.38	3.00	2.52	1.54	0.85	2.398	0.966
	1–5	3.14	4.48	13.24	18.17	15.52	15.59	2.09	10.61	5.70	5.36	4.14	1.96	2.509	0.926
	1–6	3.81	4.59	9.61	16.62	18.11	17.40	2.47	6.06	4.91	7.54	6.14	2.73	2.564	0.916
2#	2–1	7.34	10.01	26.78	18.11	14.86	7.11	1.31	4.45	3.78	2.61	2.27	1.37	2.460	0.974
	2–2	7.96	8.60	18.33	17.03	14.66	13.23	2.27	7.37	4.58	2.33	2.14	1.50	2.451	0.970
	2–3	4.50	6.10	19.68	19.59	13.93	10.62	2.34	9.74	3.93	4.01	3.66	1.90	2.503	0.951
	2–4	22.76	3.42	38.71	11.70	5.97	5.78	2.69	3.89	1.86	1.42	0.74	1.06	2.412	0.991
3#	3–1	3.98	5.46	24.12	23.06	13.40	9.92	2.65	5.67	4.30	3.41	1.94	2.09	2.484	0.982
	3–2	2.03	7.81	28.10	23.47	14.63	9.89	1.30	4.29	2.58	2.73	1.78	1.37	2.432	0.985
	3–3	5.48	11.30	17.64	19.78	14.24	11.27	1.21	6.21	3.65	3.65	3.07	2.49	2.526	0.978
	3–4	2.64	10.14	21.72	20.91	14.91	11.49	1.60	6.44	3.24	2.63	2.16	2.12	2.482	0.988
	3–5	7.02	5.66	21.44	20.56	13.73	12.37	1.41	6.49	3.12	2.98	3.28	1.94	2.497	0.974
	3–6	4.80	13.53	19.76	23.68	12.10	10.15	1.09	4.31	2.34	3.99	2.27	1.97	2.496	0.980
4#		1.69	2.32	14.15	24.33	12.60	17.47	2.74	5.60	5.25	5.43	5.41	3.02	2.556	0.922
5#		2.62	4.35	16.86	19.67	14.56	18.92	2.04	7.63	4.54	3.47	3.23	2.11	2.491	0.966
6#		3.54	5.47	16.40	18.73	13.94	17.68	2.21	6.70	5.34	4.61	3.93	2.45	2.525	0.941
7#		3.09	5.59	15.31	21.60	17.35	15.53	2.85	4.53	3.15	4.75	4.78	1.47	2.488	0.933
8#		4.40	2.15	25.62	17.11	13.84	12.75	1.48	6.68	4.47	4.73	4.66	2.10	2.528	0.944
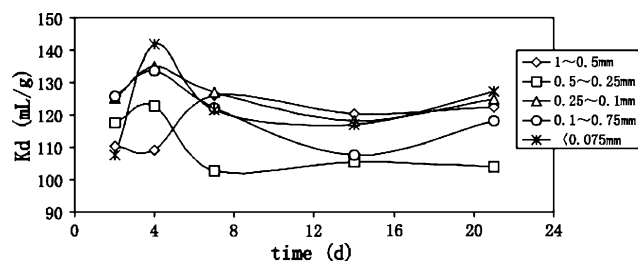
Fig. 2 Steady state sorption time of Sr sorption

Sorption isotherm (Test B)

Sorption isotherm batch tests were conducted with the following conditions: initial concentration of the Sr^{2+} at 2, 10, 20, 40, 80 and 200 mg/L, separately; mass of the grain particle is 2.5 g with the grain size less than 1 mm (fine-particle media); the ratio of the liquid and solid of 1:10; and shaking contact time of 7 days.

Figure 3 showed the sorption isotherms of Sr onto fine-particle media, a variation order can be seen from the sorption isotherms.

The Freundlich sorption model is an empirical relationship describing the sorption of solutes from liquid to solid surfaces; the equation of the Freundlich sorption isotherm is expressed as formula (4a) or (4b):

$$C_s = kC_{eq}^{\frac{1}{n}} \tag{4a}$$

or:

$$\ln C_s = \frac{1}{n}\ln C_{eq} + \ln k \tag{4b}$$

where C_s is the amount of solute adsorbed per mass unit of solution, C_{eq} is the solute concentration in the supernatant after sorption and centrifugation, N and k is the empirical constants.

According to experimental results, the relation of sorption per unit grain and concentration of Sr per unit solution was obtained as shown in formula (5). The obtained N value is 0.518, R^2 was 0.9838, showing a strongly non-linear sorption of Sr in the fine-particle media.
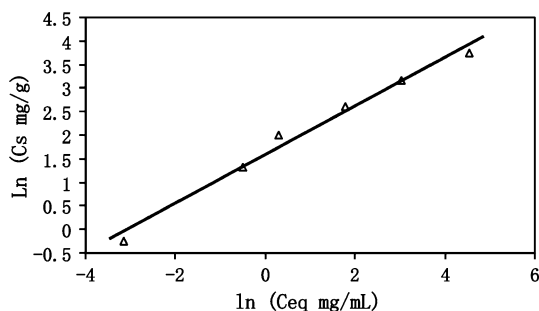

Fig. 3 Sorption isotherm of strontium on fine-particle

$$\ln C_s = 0.5184\ln C_{eq} + 1.5857 \tag{5}$$

Fitted isotherm parameters for strontium were presented in Fig. 2; the Freundlish model fit the sorption of Sr well. Other researchers also found that the behaviour of strontium in batch experiments is best characterized by Freundlich-type adsorption isotherms under the conditions studied [36] and the concentration dependence data on adsorption of Sr(II) were well fitted to the Freundlich adsorption isotherm [37].

The deduced K_d value of Sr in the heterogeneous media (Test C)

Test C contained two series of experiments named C_1 and C_2. In test C_1, based on the percentage of the sieving tests on the solid samples, three groups of batch experiments were operated using 5 g solid sample from the sample 1–1, 1–2, 1–3, which was mixed according to the ratio of the mass of the different diameter particles (1–0.5, 0.5–0.25, 0.25–0.1, 0.1–0.075 and less than 0.075 mm, respectively). The mother liquid was 10 mL $Sr(NO_3)_2$ at the concentration of 10 mg/L. Here, the distribution coefficient was noted as $K_d(C_1)$.

In test C_2, the K_d value from another three experiments was determined by the same solid samples 1–1, 1–2, 1–3 in test C_1. Every sample, with the mass of 5 g, was divided in 6 single experiments according to the particle sizes of 1–0.5, 0.5–0.25, 0.25–0.1, 0.1–0.075 and less than 0.075 mm. The distribution coefficient was noted as $K_d(C_2)$, and was calculated by the formula (6):

$$K_d = \Sigma K_{dn} \cdot X_n \tag{6}$$

where n is the grade of the chosen particle size, X_n is the percentage of the mass of the n grade particle and the total mass which included entire particle from the collected sample, K_{dn} is the corresponding distribution coefficient of the n grade particle.

Experimental conditions and the results of K_d value of test C_1 and C_2 were listed in Table 3.

The $K_d(C_1)$ could be considered as the measured value by the experiments, while the $K_d(C_2)$ was computed by formula based on the experimental data. As seen from Table 2, the difference between $K_d(C_1)$ and $K_d(C_2)$ of the same sample was very small. Therefore, the calculation methods of distribution coefficient by the mass percentage and the corresponding K_d value was feasible.

Based on the above result that the sorption contribution of the large-sized particles could be ignored, the distribution coefficient of the actual heterogeneous site could be obtained by the K_d value from fine-particles that had been measured by the batch tests. The calculated formula (7) to describe the approximate K_d value of the field site is:

Table 3 The K_d value and the relative coefficients from test C_1 and test C_2

Test	Sample	Mass of the solid sample (g)					$K_d(C_1)$
C_1	1–1	5					121.6
	1–2	5					120.7
	1–3	5					120.0

Test	Sample	1–0.5	0.5–0.25	0.25–0.1	0.1–0.75	<0.075	$K_d(C_2)$
C_2	1–1	2.21	1.02	0.85	0.58	0.34	121.2
	1–2	1.67	1.11	0.92	0.60	0.71	120.9
	1–3	1.81	1.12	0.92	0.46	0.69	122.0

Table 4 The distribution coefficient of Sr in the fine-particle (K_{d1}) and in the field site

Sample		Percentage of the mass of the fine-particle in the total (%)	K_{d1}	K_{d2}
1#	1–1	17.2	119.2	20.5
	1–2	35.3	122.4	43.3
	1–3	34.8	121.5	42.3
	1–4	14.3	118.9	17.0
	1–5	27.8	120.9	33.6
	1–6	27.4	126.1	34.5
2#	2–1	14.5	121.8	17.6
	2–2	17.9	119	21.3
	2–3	23.2	121	28.1
	2–4	9.0	119.8	10.7
3#	3–1	17.4	121.7	21.2
	3–2	12.8	122.5	15.6
	3–3	19.1	123.3	23.5
	3–4	16.6	121.5	20.2
	3–5	17.8	122.6	21.8
	3–6	14.9	125.1	18.6
4#		24.7	120.7	29.8
5#		21.0	121.5	25.5
6#		22.0	120.5	26.5
7#		18.7	125.7	23.5
8#		22.6	123.7	28.0

$$K_{d2} = K_{d1} \cdot X_1 \tag{7}$$

where, K_{d1} is the distribution coefficient of Sr in the fine-particle, which was the diameter less than 1 mm in this work, X_i is the percentage of the mass of the fine-particle possessing in the total mass, and K_{d2} is the approximate distribution coefficient of Sr in the heterogeneous media. The calculated results from this method were shown at Table 4.
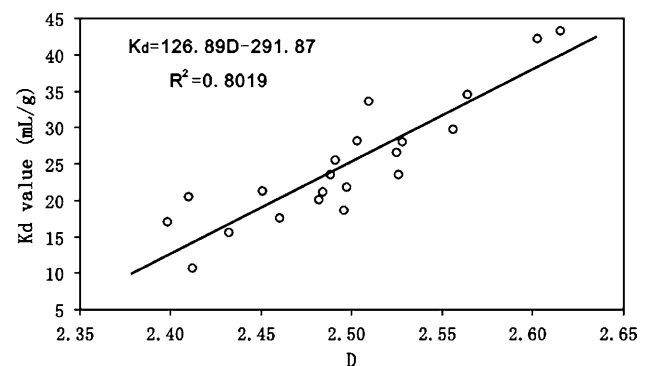
The relation of K_d and D

Heterogeneous character and random distribution of different size particle is the property of the heterogeneous media, and it is difficult to find out the orderliness. The fractal dimension, which is based on the mass content, not only reflects the gradual changing of the media, but could explain the difference of the texture. From the theory of hydrogeological parameter measurements, the fractal dimension could discover the rule and relation between the spatial framework of the heterogeneous media and the hydraulic conductivity and dispersivity [38]. However, the sorption capacity of heavy metals or nuclides is described as distribution coefficient, which depends on the particle size in the media. For the same material, the capacity of sorption is increasing by the gradual decreasing of the solid grains size. Therefore, some relation may exist between the distribution coefficient and the fractal dimension of the mass. In this work, because the VLLW disposal site is obviously heterogeneous media, where the diameter of the grains ranges from 75 cm to $n \times 10^{-2}$ mm, as shown from the result of the sieved experiments, the field-scale distribution coefficient is hard to measure directly. The relation and expression of K_d and D is a significative study for the sorption capacity of nuclide in the heterogeneous media.

According to the above fractal dimension from the sieved experiments and distribution coefficients from the batch tests, the relative curve was described in Fig. 4. The regression method was applied to fit the expression, as shown in formula (8).

$$K_d = 126.89D - 291.87 \tag{8}$$

Equation 8 depicted a relative character about the distribution coefficient and fractal dimension, with the correlation coefficient of 0.8019.

The results showed the general correlation of distribution coefficient of Sr with the fractal dimension of grain size. Some reasons were inferred about the phenomena: (1) the adsorption is a predominantly non-selective surface

Fig. 4 The correlation of K_d and D

process and depends on the specific surface area of the sediment, which is strongly related to the grain size; and (2) the fractal dimension has a close correlation with the grain size, it reflected the diversification of the surface texture of the grain size and the specific surface area and total pore volume. So the interrelated variety trend of the sorption of Sr and fractal dimension was found. Future work will focus on the relation of the surface areas of the various particle size fractions and fractal dimension.

Conclusion

A total of 21 deposit soil samples were collected in the candidate VLLW disposal site, among them, 16 samples were in the different depth from three deep excavated pits; the others obtained from trenchs at the same site. Sieved experiment and batch test in the fine-particle ($d < 1$ mm) was applied to study the fractal character and sorption. The results showed that the fine-particle has a percentage of about 10–20%, and the fractal dimension based on the mass content ranges 2.39–2.61. About the sorption characteristics, the steady state sorption time of strontium sorption is 7 days, and the Freundlich isotherm described the sorption well. The distribution coefficient of Sr ranged from 118.9 to 126.1 in the fine-particle, while it bounded from 10.8 to 43.2 by the deduction method of mass percentage. A regression method was applied to fit the correlation between K_d and D, with an approximately linear relation obtained.

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