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The study of migration velocity of Cs-135 and Se-79 in intact rock

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Abstract: The migration characteristics of highly mobile nuclides in natural barrier are the key concerns in the long-term safety assessment of geological disposal facilities. This paper focused on typical granite in Beishan area for geological disposal of high-level radioactive waste, and established a computational model for the release and migrate of nuclides after closure of repository by GoldSim. The migration characteristics of Cs-135 and Se-79 were analyzed by Monte Carlo stochastic simulation. The results show that the migration characteristics of Cs-135 and Se-79 in intact rock are direct influenced by hydraulic conductivity and hydraulic gradient, the migration velocity are 2.47×10^{-4}m/a and 9.61×10^{-4}m/a when the hydraulic conductivity correspond to 5.74×10^{-9} m/s. The analysis of the results shows that the migration characteristics can provide feedback guidance for the site selection and design of the disposal repository.

1. Introduction

The safety disposal of high-level radioactive waste(HLW) is an inevitable requirement for the protection of people and environment[1]. At present, the feasible method for disposal of HLW is deep geological disposal. Granite is a kind of suitable host rock for geological disposal[2-3], and it is also the main host rock in Beishan, Gansu province, which is the preferred preselected area for disposal of HLW in China. In general, the nuclides will migrate into the host rock as soon as the failure of engineered barrier[4].Considering the characteristic of containing random micro fractures, the hydraulic conductivity values are extremely low in intact rock, and it plays a vital role in the process of retard nuclides.

The processes involved in the migrate of nuclides in host rock are extremely complex, accompanied by the coupling effects of advection, diffusion, adsorption and precipitation. And the migration characteristics of nuclides vary greatly, as well as the impact on the long-term safety of the repository. Finland, Sweden, France, Canada, Japan and other countries have found that long-lived nuclides such as Cs-135, Se-79 and Tc-99 are generally the key sources of radioactive effect from repository[5-9]. It is of great significance to carry out the study on migration characteristics of such nuclides in host rock for site characterization. In this paper, the long lived nuclides of Cs-135 and Se-79 with similar distribution coefficients are selected for the research of migration characteristics in intact rock of granite in Xinchang sub-area.

2. Introduction of the Xinchang sub-area

The Xinchang sub-area is located in the middle of the Beishan area (Figure 1[10]), with a distance to Jiayuguan city about 135 km, which is a potential sub-area for HLW repository. The Xinchang granite intrusion is a 22 km long and 7 km wide rock block, intruded into the east-west striking Proterozoic

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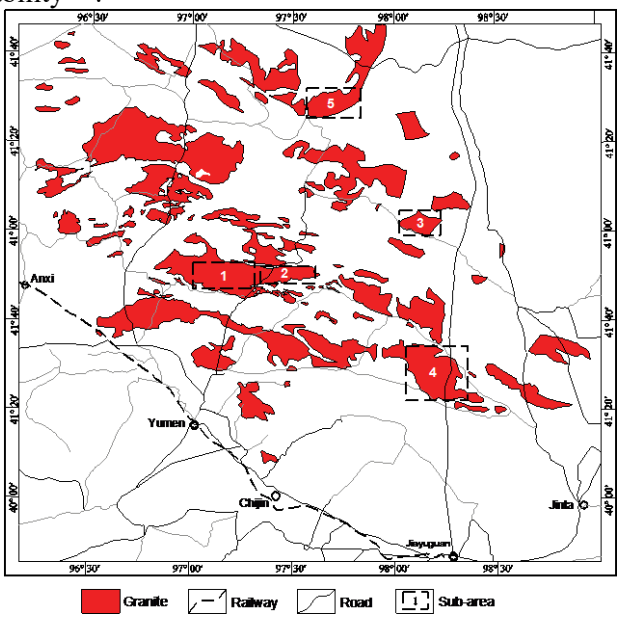
metamorphic rock belts. The major rock types include gneissic biotite monzonitic granite and biotite granodiorite with an age around 260 million years. The hydrogeological investigation show that for most of the test intervals within intact rock or fracture zones, the hydraulic conductivity values are very small, generally less than 1.0×10^{-8}m/s, and mainly concentrating between 1.0×10^{-12} m/s and 1.0×10^{-10}m/s, the hydraulic gradient of groundwater is generally less than 1%, indicating that the host rock of the site have an extremely low permeability[10].



1-Jiujing sub-area; **2-Xinchang sub-area**; 3-Yemaquan sub-area; 4-Shazaoyuan sub-area;5-Suanjingzi sub-area.

Figure1. Location of the Xinchang sub-area in the Beishan area

In this paper, the hydraulic conductivity values are obtained from the field hydraulic test results in deep vertical boreholes in Xinchang sub-area (BS06,BS17,BS18,BS19, BS32,BS33). The hydraulic conductivity values of intact rock between 200~600m in boreholes are shown in Figure 2 (excluding the hydraulic conductivity values of water conducting fractures). It is revealed that the hydraulic conductivities of 90% test intervals are lower than 1.0×10^{-9}m/s.

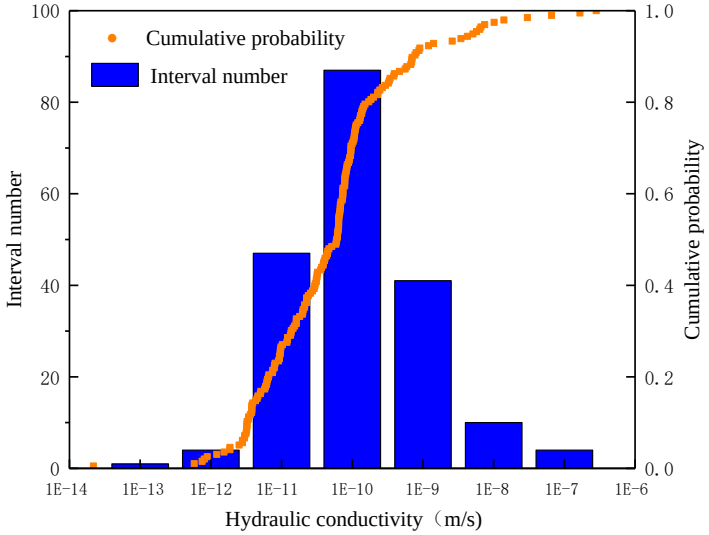


Figure 2. Hydraulic conductivity distribution of intact rock at a depth from 200 to 600 meters

3. Simulation process

Based on the preliminary concept of HLW geological disposal in China [11] and the characteristics of

Xinchang sub-area[10,12], the normal evolution scenario of the nuclides release and migrate after closure is constructed by GoldSim. Then, the migration characteristics of nuclides in intact rock are studied by the method of scenario and parameter uncertainty analysis. In this study, it is assumed that all containers fail at 1,000 years after repository closure, and the calculation extends to one million years after. The spatial extent of the assessment model is from the repository to the biosphere including waste form, corrosion products, buffer, EDZ, intact rock, fractured rock, fault, and the interface of geosphere and biosphere (Figure 3). The safety indicator is the far field release rate of nuclides, which represents the activity of nuclides migrating out of the natural barrier and entering the biosphere, and the unit is Bq/a.

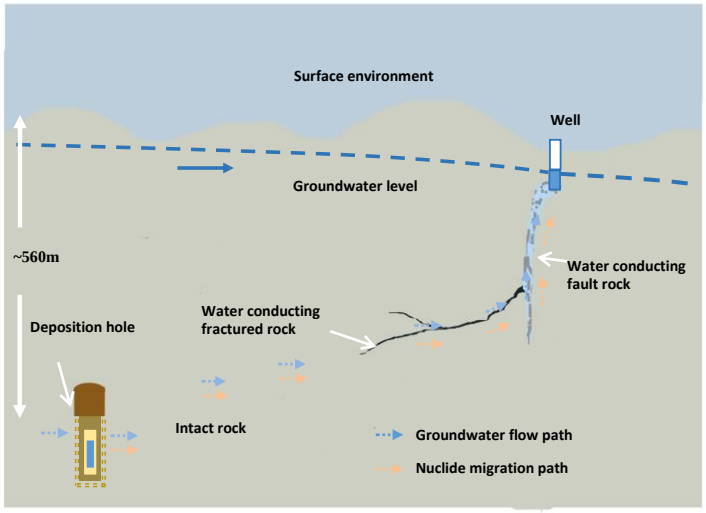


Figure 3. Illustration of radionuclide transport pathway for Xingchang sub-area

The nuclide inventories, effective diffusion coefficient, distribution coefficient, solubility data reference to related data from JAEA [9], whose disposal concept similar to the preliminary concept of HLW geological disposal in China (The key parameters of Cs-135 and Se-79 are shown in Table 1), The data about corrosion products are taken from EPRI[13], the characteristic parameters of buffer are taken from JAEA[9]and Liu [14]. The migration process of nuclides in host rock adopts the one-dimensional flat model. It is assumed that the hydraulic gradient of the entire migration path is 0.3% and the longitudinal dispersion length is assumed to be 10% of the transport length. Other hydrogeological parameters are taken from the latest results from the Xinchang borehole sampling[10] and from Chen [15].

Table 1. The key parameters of Cs-135 and Se-79

Nuclide	Half-life [a]	Solubility [mol/L]	Distribution coefficient K_d for Corrosion products [m^3/kg]	Distribution coefficient K_d for buffer[m^3/kg]	Distribution coefficient K_d for granite[m^3/kg]
Cs-135	2.30×10^{6}	Highly	0.01	0.01	0.05
Se-79	2.95×10^{5}	3.00×10^{-9}	0.001	0.001	0.01

The main processes modeled include advection-diffusion in fractures, matrix diffusion, and nuclides decay. The governing equation expressing nuclide transport in the matrix under steady-state groundwater flow conditions are as follows[15]:

$$R_n^m \frac{\partial C_n^m}{\partial t} = \frac{\partial}{\partial \omega} D^m \frac{\partial C_n^m}{\partial \omega} - R_n^m \lambda_n C_n^m + R_{n-1}^m \lambda_{n-1} C_{n-1}^m \quad (1)$$

Where:t:time[s]; n:nuclides; m:matrix(intact rock); R_n^m:retardation coefficients for nuclide n in the matrix m; λ_n:decay constant[s^{-1}]; θ^m:matrix porosity; D^m:effective diffusion coefficient[m^2/s]; C_n^m:nuclide concentration in fractures and in the matrix[mol/m^3]; ω:perpendicular distance into the matrix from the fracture surface[m]; t:time[s].

4. Results and discussion

4.1 The migration velocity of Cs-135 and Se-79 in intact rock

It is assumed that the thickness of intact rock around the deposition hole is 10m and 50m respectively, the far-field release rate of Cs-135 and Se-79 are statistically analyzed (Figure.4). The results show that the far-field release rate of Cs-135 and Se-79 decrease with the increase of the thickness of intact rock. The far-field release rate decrease almost four orders of magnitude as the intact rock increased five times. Then, the migration velocity of Cs-135 and Se-79 in intact rock are studied by Monte Carlo stochastic simulation.

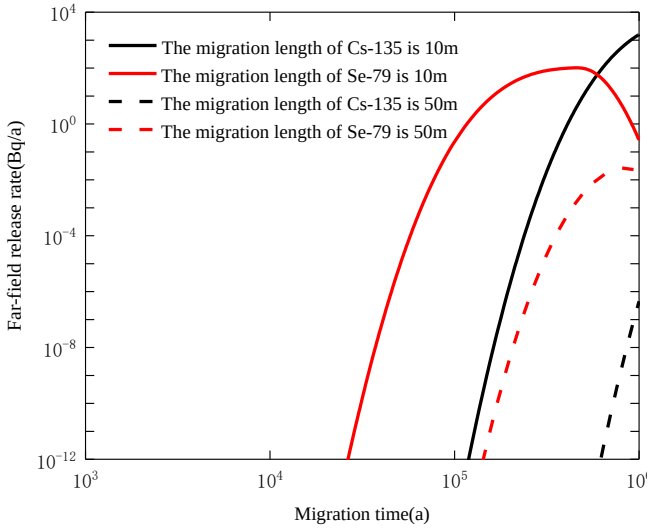


Figure 4. The far-field release rate of Cs-135 and Se-79 in intact rock

As shown in Figure 2, the hydraulic conductivities of 95% test intervals correspond to 5.74×10^{-9} m/s. In this study, the 50 random sampling between 1m to 100m of intact rock are made by Monte Carlo stochastic simulation, then input parameters of thickness of intact rock and simulated respectively. The far-field release rate of Cs-135 and Se-79 varied with the thickness of intact rock are statistically analyzed (Figure 5A). The results show that the migration time of Cs-135 and Se-79 have a linear relationship with the thickness of intact rock, and the migration velocity of Cs-135 and Se-79 in intact rock are about 2.47×10^{-4}m/a and 9.61×10^{-4}m/a respectively (see Figure 5A and Table 2), which can ensure that the nuclides stay within the site for hundreds of thousands of years. Consider an extreme case, when the hydraulic conductivity of the intact rock is set as the maximum value of 5.59×10^{-7} m/s from the field hydraulic test results in deep vertical boreholes, the results reveal that the migration velocity of Cs-135 and Se-79 in intact rock are about 8.85×10^{-2} m/a and 0.37 m/a respectively (see Figure 5B and Table 2). The calculation results show that the nuclides may be isolated in the host rock for ten thousands years when the width of the repository is 3km.

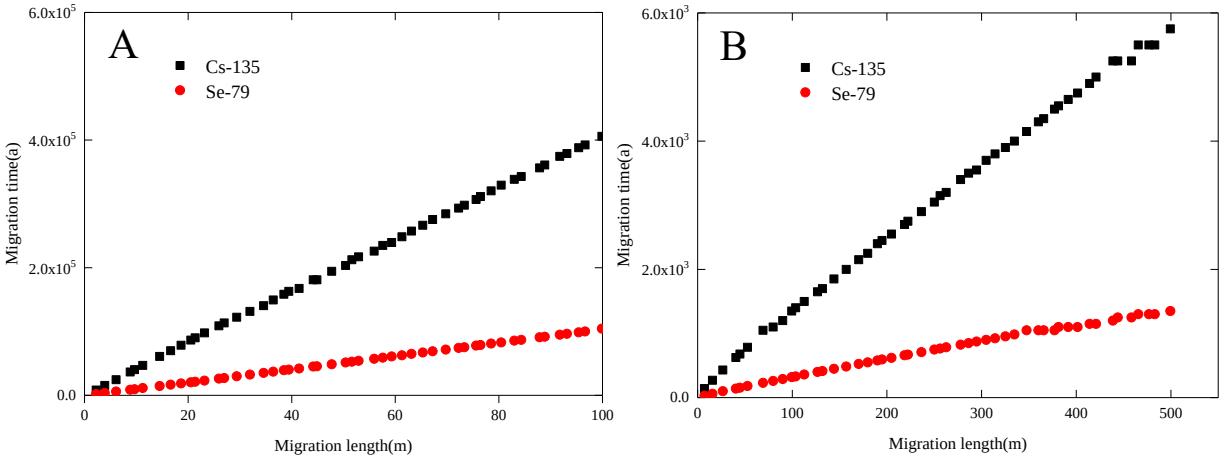


Figure 5. The migration velocity of Cs-135 and Se-79 in intact rock
(Figure 5A. represent intact rock hydraulic conductivity of 5.74×10^{-9} m/s; Figure 5B. represent intact rock hydraulic conductivity of 5.59×10^{-7} m/s)

Table 2. The migration velocity of Cs-135 and Se-79 in intact rock

Hydraulic conductivity (m/s)	Nuclide	Migration velocity(m/a)	Correlation coefficient R^2
5.74×10^{-9}	Cs-135	2.47×10^{-4}	0.9998
	Se-79	9.61×10^{-4}	0.9999
5.59×10^{-7}	Cs-135	8.85×10^{-2}	0.9988
	Se-79	3.70×10^{-1}	0.9957

4.2. Factors affecting migration velocity of Cs-135 and Se-79 in intact rock

After container failure, nuclides will migrating toward the biosphere through groundwater. The migration velocity of nuclide is direct related to the groundwater flow, while groundwater flow depends on hydraulic conductivity and hydraulic gradient. The following research focus on the nuclides migration influenced by hydraulic conductivity and hydraulic gradient of intact rock.

It is assumed that the thickness of the intact rock is 10m, the hydraulic gradient is 0.003. And the hydraulic conductivity is set between 1.00×10^{-9}m/s and 1.00×10^{-7}m/s, 50 random sampling of hydraulic conductivity are made by Monte Carlo simulation method and simulated respectively. The relationship between migration velocity and hydraulic conductivity are statistically analyzed(Figure 6A). It is assumed that the thickness of the intact rock is 10m, the hydraulic conductivities is 5.74×10^{-9} m/s. And the hydraulic gradient is set between 0.001 to 0.003, 50 random sampling of hydraulic gradient are made by Monte Carlo stochastic simulation and simulated respectively. The relationship between migration velocity and hydraulic gradient are statistically analyzed(Figure 6B).

Figure 6 shows that the migration velocity of Cs-135 and Se-79 are in line with the negative exponential function as well as the hydraulic conductivity and hydraulic gradient. The migration velocity increase or decrease correspondingly with the hydraulic conductivity and hydraulic gradient. And, Cs-135 is more sensitive than Se-79.The simulation results in Fig. 6 also show that when the hydraulic conductivity of the intact rock is 5.74×10^{-9}m/s and the hydraulic gradient is 0.003, the safety of the disposal repository can be guaranteed when the thickness of intact rock reaches 10m, indicating that the typical granite in Beishan area is favorable for geological disposal of HLW.

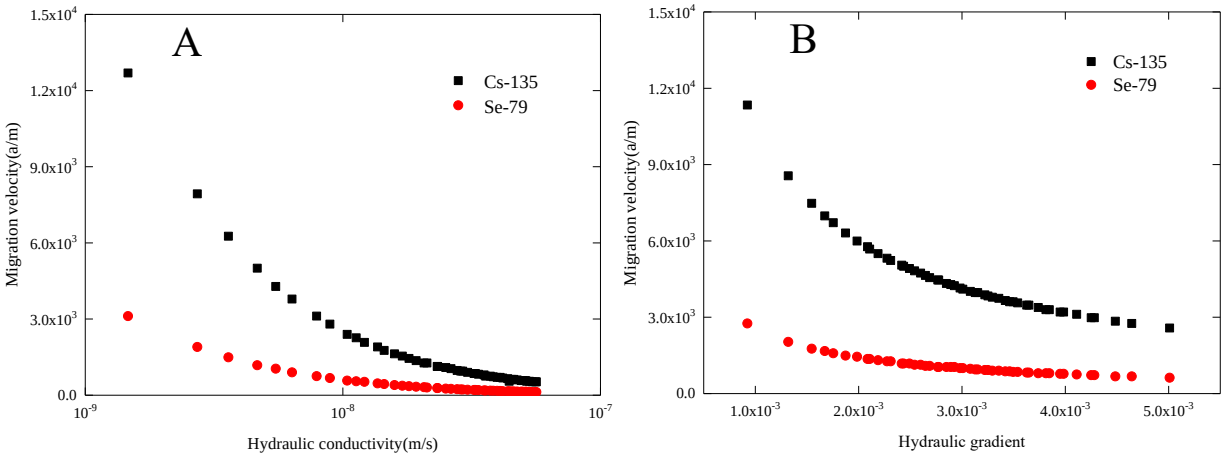


Figure 6. The migration velocity of Cs-135 and Se-79 vary with hydraulic conductivity and hydraulic gradient

The current results of site investigation revealed that the typical intact rock of granite in Beishan area have favorable characteristics for waste disposal, such as low hydraulic conductivity and low hydraulic gradient, which can effectively support the long-term stability of the engineered barrier and retard the nuclides release. However, the water conducting fractures are unfavorable to long-term safety of granite disposal repository. The distribution of water conducting fractures in host rock is of remarkable randomness, and it is faced with great uncertainty to master the distribution characteristics of fractures within the site. Therefore, it should be paid more attention to identify and analyze of the uncertainty of the distribution of water conducting fracture in site investigation, as well as the optimized of the conceptual design of the disposal repository optimized according to the uncertainty analysis results , so as to give consideration to the functional settings of engineered barriers and natural barriers for the safety disposal of radioactive waste.

5.Conclusion

Based on the actual needs of geological disposal research and development, combined with the existing research basis in Beishan area, the migration characteristics of Cs-135 and Se-79 in intact rock are studied by Monte Carlo simulation method. The main conclusions are as follows:

(1)The migration time of Cs-135 and Se-79 have linear relationship with the thickness of intact rock, the migration velocity of Cs-135 and Se-79 in intact rock are about 2.47×10^{-4}m/a and 9.61×10^{-4}m/a when the hydraulic conductivity correspond to 5.74×10^{-9} m/s, which can ensure that the nuclides stay within the site for hundreds of thousands of years.

(2)The migration velocity of Cs-135 and Se-79 are in line with the negative exponential function as well as the hydraulic conductivity and hydraulic gradient, which increase with the decrease of the hydraulic conductivity and hydraulic gradient.

Acknowledgments

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