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Multi-parameter Experimental Adsorption Effect of ^{239}Pu on Phyllitic Slate and Clay

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Abstract

The paper studies the adsorption features of ^{239}Pu in phyllitic slate and clay media through static experiment method. The two different geo-media were collected from a site for disposing very low level waste (VLLW) as experimental samples. Experimental results have shown that an adsorption equilibrium time of ^{239}Pu in phyllitic slate and clay is approximately 10 days, and the adsorption coefficient (K_d) values of phyllitic slate and clay are about 6000ml/g and 2700ml/g, respectively. The experimental result of particle size effect indicates when adsorption of plutonium in particle samples increases, the K_d values of these two geo-media increase while the particles decrease in size. And the experimental impact on aqueous phase pH value reveals that K_d of ^{239}Pu increases at higher aqueous phase pH value, illustrating stronger adsorption capacity. Experimental data are also measured through the curve fitting, K_d values are thus obtained theoretically.

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Keywords: ^{239}Pu; phyllitic slate; clay; K_d value

1. Introduction

Studies on the adsorption and migration behavior of radionuclide in natural geological-mass and artificial medium is essential during the process of selecting and constructing radioactive waste disposal site. Plutonium, one of the important adsorbing radionuclide in high level radioactive wastes, has

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motivated scholars to carry out research on its adsorption behaviors. Plutonium is known to exhibit mainly five oxidation states, 3+ through 7+, in aqueous solutions. Among all the valence states, Pu(Ⅳ) is the most stable, and heptavalent plutonium ions only exists in alkali solution, and Pu(V) remains stable in solutions at pH values range from 2 to 6. Plutonium ions may exist in the forms of Pu^{3+}, Pu^{4+}, $[PuO_2]^+$, $[PuO_2]^{2+}$ at different valences. Scholars from China Institute of Radiation Protection have carried out field tests and simulation experiments to study the migration characteristics of ^{237}Np, ^{238}Pu, ^{241}Am and ^{90}Sr in loess, aquifer, engineering barrier material, cement, metamorphic cement solidified block, cement mortar powder-soil[1-6]. These experiments enhanced our understanding of migration characteristics of radionuclide. However, the migration characteristics of ^{239}Pu has been little reported in the literature, to the best of our knowledge. ^{239}Pu is an important isotope of plutonium and byproduct of nuclear reactor operations and nuclear bomb explosions. It may interact with the underground water and surrounding geological materials, and trigger chemical reactions such as oxidation-reduction, disproportionation, hydrolysis reaction, precipitation-dissolution, adsorption and desorption etc. These reactions may lead to changes of the nuclide forms in the underground water, and thus influence the adsorption and migration of plutonium[7-12]. Therefore, it is necessary to further investigate the adsorption dynamics of ^{239}Pu under different geological conditions and aqueous environments.

This paper investigates the adsorption characteristics of nuclide in phyllitic slate and soil (mostly clay) gathered from a site for disposing very low level waste (VLLW). In particular, we focus on 1) the adsorption equilibrium time of ^{239}Pu in the samples; 2) the experimental impact of particle size change on adsorption capacity; and 3) the experimental impact of aqueous phase pH value change on plutonium adsorption by analyzing K_d (adsorption coefficient) values of ^{239}Pu under different conditions. The curve fitting to experimental data is carried out and an analysis of the adsorption characteristics of ^{239}Pu under different conditions is conducted.

2. Theory and experiments

2.1. Adsorption coefficient

Adsorption coefficient K_d (adsorption ratio) is an "equilibrium" distribution of some nuclides between the solid phase and liquid phase. Using the static method, the adsorption coefficient can be defined as following:

$$K_d = \frac{(A_0 - A_t)/\mathrm{m}}{A_t/V} \qquad (1)$$

This experiment dopted the static experiment method, which was the so called batch type method. The materials were placed in a container, then radioactive carrier was added. Meanwhile, the same volume of radioactive GSCC liquid was prepared in order to measure the radioactivity A_0 (min^{-1}) of tracer liquid which was not in contact with the materials. After the container was oscillated for a certain period of time, the source material underwent centrifugation. Constant temperature was maintained during the whole experiment. Through this method, the radioactivity A_t (min^{-1}) of solution was measured after adsorption in liquid phase. Thus, the adsorption ratio (distribution coefficient), adsorption quantity and aqueous nuclide concentration at adsorption equilibrium were obtained. For the purpose of calculations, Eq. 2 is used in this paper, which is equivalent to Eq. 1:

$$K_d = \frac{C_0 - C_t}{C_t} \bullet \frac{V}{m} \qquad (2)$$