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Research on adsorption and desorption behavior of Pu on bentonites for buffer backfill material

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Abstract

To the chemical behavior of Pu on backfill material for high-level radioactive waste repository. Adsorption and desorption of Pu on Natural bleaching earth, Na-bentonite, GMZ-bentonite, Ca-bentonite, acid-Ca-bentonite and alk-Ca-bentonite are discussed in this paper. Data analysis shows that adsorption abilities of Ca-bentonite, alk-Ca-bentonite，Ca-bentonite are greater than GMZ-bentonite，Natural bleaching earth, Na-bentonite, and in which Na-bentonite is relatively minimal. Desorption abilities of Na-bentonite, acid-Ca-bentonite are greater than alk-Ca-bentonite, Ca-bentonite, GMZ-bentonite, Natural bleaching earth, and in which GMZ-bentonite and Natural bleaching earth are relatively minimal. When the pH of the water phase is 1 or 13, the adsorption ability of Ca-bentonite is changed obviously. One reason is that the degree of surface complex reaction has been changed. Another reason is that the compositions and structure of Ca-bentonite have been also changed after acidizing or alkalosis by H^+ or OH^- in the water phase.

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Keywords: *239Pu；Bentonite；Adsorption；Desorption；*

1. Introduction

Nuclear waste repository contains multiple barrier systems that are mainly divided into engineered barrier and natural barrier. Engineered barrier includes high-level radioactive waste form, packaging containers, and buffer/back-fill materials, etc. Bentonite, internationally recognized as the ideal buffer/back-fill material of underground

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repositories for high-level radioactive, is the last line of artificial barrier materials and plays an important role in the conduction and dissipation of the decaying heat from radioactive waste, and can keep the security and stability of the geological repository effectively. In the studies and researches from Chinese scholars by years, it is found that GMZ-bentonite mineral deposit in Xinhe County of Inner Mongolia, where bentonite has some characteristics such as high montmorillonite content, large cation exchange capacity, and large specific surface area, is suitable as a preferred supply base of buffer/back-fill materials in china. Pu is the most important element in transuranic elements and it has 20 kinds of isotopes, in which ^{239}Pu is the most important. The radioactive element ^{239}Pu is an important raw material in atomic energy industry and can be used as a fission agent of nuclear fuels and nuclear weapons. Meanwhile, nuclide ^{239}Pu is one of the important key radio nuclides in the radioactive waste and it mainly comes from spent fuels of nuclear reactors and waste water produced by past nuclear tests. It has a high specific activity, a large heat release rate, a long half-life(T1/2=2.4×104(year)), a high biotoxicity and it also has a unique redox chemical property and multiple valence coexisting in the same system. So ^{239}Pu is always a main attention object of geological disposal of radioactive waste and environmental safety assessment, in particular, it plays an important role in the geological disposal safety of high-level radioactive waste and environment assessment. Currently, some study results about adsorption behavior of Pu in various types of clay have been achieved. Adsorption and desorption behaviors of ^{239}Pu in Natural bleaching earth, sodium bentonite, GMZ-bentonite, calcium bentonite, acidified calcium bentonite and alkalized calcium bentonite are discussed in this paper.

1 Experimental methods and principle theories

1.1 Bentonite samples

Four kinds of soil samples, including Natural bleaching earth, sodium bentonite and calcium bentonite in Lingshou Shijiazhuang and GMZ-bentonite in Inner Mongolia, are chosen as research objects, whose oxide components are shown in table 1.

Table 1 Chemical composition of bentonite samples

Samples	*w*/%									
	Al_2O_3	SiO_2	P_2O_5	CaO	K_2O	TiO_2	Fe_2O_3	MgO	Na_2O	MnO
Natural bleaching earth	6.32	33.66	0.06	25.55	2.00	0.13	1.15	8.00	1.35	0.06
Na-bentonite	10.20	64.30	0.02	6.30	2.07	0.16	1.51	2.15	0.24	0.09
Ca-bentonite	12.14	19.05	0.09	29.69	1.34	0.23	1.92	14.25	0.20	0.05
GMZ-bentonite	12.14	44.65	0.13	13.20	1.24	0.45	4.10	2.10	0.23	0.23

At first, we add HNO_3 solution at pH = 1 into calcium-based soil dropwise. Soil sample reacts with HNO_3 and instantly a lot bubbles are generated. Next, we continue to add HNO_3 solution until no more bubbles generated. After soil has been soaked in HNO_3 solution for about 48h, acidified calcium soil is obtained through being filtered, being washed by distilled water, and being dried for 3d at 80℃. The above operations are repeated by using the same amount NaOH solution at pH=13 and alkalized soil calcium is obtained. All the soil samples are milled by a 200 mesh sieve.

1.2 Experimental methods

1.2.1 Adsorption experiments

Each of six soil samples is weighed 0.08g and is put in a centrifuge tube of 10mL. Original solution of 5mL is added into it and is stood for 24 hours. And then, we add standard Pu solution of 20 μL(1600Bq/mL) in it. The centrifuge tube is shocked in the vibrator until the adsorption equilibrium, then it is removed from vibrator, and is done centrifugation for 1 hour (speed of 5000r/min). 4mL supernatant is taken to prepare a sample and its α activity