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Removal of plutonium from contaminated soil by chemical leaching

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Abstract

Due to the high cost and technical challenges for the treatment and disposal of heavy plutonium(Pu) contaminated soils, it is deserved to develop efficient methods to remove Pu from contaminated soil in order to reduce its concentration and the volume of the radioactive waste required to be deposed with high standard. Inorganic acids, organic acids and oxidants were screened deliberately for removal Pu from one kind of Pu contaminated desert soil. The leaching process was investigated for the dependence of the decontamination efficiency with batch wise leaching. Furthermore, the decontamination efficiencies of Pu from the leachate were studied by flocculation deposition. The removal efficiencies increased with the augment of liquid-solid ratio, temperature, stirring time and the maximum Pu removal efficiency of 90% specific activity was gained by sulfuric acid. The inorganic and organic flocculants are capable to remove Pu in the leachate completely with decontamination factor more than 5×10^3 , and the weight of final residue less than 10% of the initial contaminated soil and the remaining specific activity of Pu-239 in the treated leachate less than 0.07Bq/L, which fulfills the requirement for direct discharge.

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1. Introduction

The global or regional soil radioactive contamination originated from the research and development of nuclear weapons, accident of nuclear power station or application of isotopes [1]. Plutonium (Pu) is one of the most concerned contaminated actinide nuclides due to the extreme radiotoxicity, highly chemical toxicity and relatively long half life ($T_{1/2}$, ²³⁹Pu=24100a, ²⁴⁰Pu=6560a). After it released into the environment by human activity, Pu can be intake to the human organism through the alimentary, respiratory canal or wound and then concentrated in the bone

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tissue and kidneys thus forming sources of local irradiation [2]. After released into soil, Pu combined with the main soil constituent like montmorillonite, illiete and organic matter, which lead to the extraction of Pu a high challenge work. Consideration of the high cost and technical challenges for the treatment and disposal of heavy Pu contaminated soils, it is deserved to develop efficient methods to remove Pu from contaminated soils in order to reduce its concentration and the volume of the radioactive residue required to be deposed with high standard geological disposal. Pu has the most complicated chemical behavior in all elements and the complex reaction made the completely extraction of Pu difficult by biological or physical remediation technologies. As one of traditional remediation technologies, chemical leaching can remediate the contaminated soil by the dissolution, extraction and separation of nuclides effectively [3]. During the leaching process, the separated nuclides were transformed to the appropriate form for successive disposal or recycle, and the rapid separation especially suit for the emergent case, like NPPs accident. To break the combination of Pu with soil constituent, acids, oxidant and complexant always used for resolution and separation of Pu from soil. But during the process of Pu removed from contaminated soil, the loss of soil constituent must be highly concerned for the ecological consideration. Thus, the integrated utilization of acids or reagents should be deliberately selected for fulfill the requirement of Pu removal and soil ecological protection. Inorganic acids like HNO₃, H₂SO₄, HCl and HClO₄ have been successful used for digestion and determination of Pu in soil. Organic acids like CH₃COOH, citric acid and EDTA possess the prominent property of oxidation and coordination with Pu, which could improve the extraction efficient. Meanwhile, the introduction of oxidant would break Pu complex in soil, which not only benefit the extraction of Pu from soil but also for the extraction of Pu from leachate. Meanwhile, the decontamination efficiency of process was dominated by the leaching technologies which influence the transfer of Pu in liquid-solid system. In this present paper, inorganic acids, organic acids and oxidants were screened deliberately for extraction of Pu from one kind of desert contaminated soil and the leaching technologies also were investigated attentively. Furthermore, the removal efficiency of Pu from the leachate was studied by flocculation deposition and the decontamination factor for total process was evaluated.

2. Experimental

2.1. Materials and reagents

All the chemicals were of analytical grade and without further purification prior to use. Dowex 1×4 strong basic anion exchange resin (60-80 mesh), production of Dow Chemical, American; the ultrapure water with resistivity of 18.2 M Ω ·cm was obtained from a Milli-Q water purification system, Millipore, American; the ultrapure acid was prepared by a DET2800 purification system, Berghof, German; QM-3SP4 soil ball grinding mill, Instrument Factory of Nanjing University, China; SevenEasy pH meter with a LE420 pH electrode, Meteller Toledo, American; Pu in all samples was determined by a X series II type quadrupole ICP-MS (ICP-QMS, Thermo Fisher, American) after purified by Dowex 1×4 anion exchange resin in $6\sim8$ M HNO₃. Polyaluminum chloride (PAC), polymeric ferric sulfate (PFS), anionic polyacrylamide (APAM) and cationic polyacrylamide (CPAM) were purchased from British chemical industry co., LTD, Shanghai, China.

2.2. Preparation of soil

Desert soil was collected and spiked with PuO_2 powder, homogenized and sieved through a 200-mesh nylon sieve. The humidity of soil was determined by drying the soil in oven at 105 °C overnight, and the pH, content of total dissolved salt and carbonate were also determined.

2.3. Batch leaching experiment

In a typical batch leaching experiment, 1g of soil was added into a 50ml centrifugal tube and 10:1 aliquot of leaching reagents was added. After stirring for 48h at room temperature (20°C), the two phases were separated by centrifugation for 30min with speed of 4000 rpm. The concentrations of Pu in the supernatant liquor (leachate) and residue were determined by ICP-QMS after pre-treatment or digestion. The residue was dry at 105°C overnight and weighed for calculation of the mass loss (S_1) of soil during the leaching process, which was defined as the following eqn:

$$S_{\rm l} = \frac{m_0 - m_1}{m_0} \ 100\% \tag{1}$$

Where, m_0 and m_1 represent the mass of soil before and after leaching respectively. The removal percent of Pu (R_p)

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