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Solubilization of Pu-239 in low-level radioactive contaminated soil by the addition of microbial leaching solution of *Acidithiobacillus ferrooxidans*

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Abstract

Researches on microbial solubilization of traces of radionuclide plutonium in contaminated soil with *Acidithiobacillus ferrooxidans* were conducted. When *A. ferrooxidans* was cultured in medium containing sulfur and iron, 92.93% of the Pu-239 in solution was removed during bioleaching. In addition, the changes of pH value, concentrations of Fe²⁺, Pu-239, organic matter, nitrogen and phosphorus were periodically analyzed. The results showed that bioleaching affected the combination forms of Pu-239 existing in the soil. The percentage of Pu-239 exists in exchangeable state increased significantly because the carbonate state, iron and manganese oxidation state and residual state transformed into exchangeable state during bioleaching. At the same time, the percentage of organic state kept steadily. The changes of form into exchangeable state which can improve the leaching rate effectively. The results showed that the bioleaching process can be used to remove Pu-239 from contaminated soil.

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Keywords:

1. Introduction

Since July 1945, the United States conducted the first nuclear test in New Mexico desert, a total of eight countries around the world had conducted more than 2,000 times of nuclear weapons tests (including atmospheric tests). At the same time, about 30 nuclear explosion test sites have been established. Nuclear legacy pollutants have become a

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serious threat to ecosystem [1-4]. The United States, Russia and other nuclear-armed countries have investigated and studied the effect of plutonium to environment in nuclear test sites [5-8].

There are 20 different plutonium isotopes, while ^{239}Pu need to be specially considered because ^{239}Pu has unique properties [9-10]: i. Easy to decay; ii. Half-life is 24,100 a, and it can decay into ^{235}U whose half-life is 7 billion years. At the same time, ^{239}Pu has the characteristics of radiation toxicity, high activity and high heat energy [11]. And it will generate internal irradiation, causing serious consequences when the radioactive nuclide came into our body. Broadly, the technologies used to remedy soils contaminated by ^{239}Pu can be classified into two categories: physical and chemical technologies. The physical technology includes high gradient magnetic separation (HGMS), electrodynamics decontamination and in situ verification, while chemical method mainly includes washing decontamination and heap leaching [12]. In comparison to the traditional physical and chemical treatment methods, using in situ stabilization techniques cannot only lower the cost and the secondary pollution, but also provide a practical way for the treatment of radioactive soil, and a lot of experiments have been carried out on this subject [13].

A. ferrooxidans was firstly separated by Hinkle and Colmer from acid mine drainage in 1947 [14]. Since then Temple conducted further researches on the strain [15]. The short rod bacterium, belonging to acidophilus chemoautotroph strain, obligate aerobic, suitable growth pH value of 2.0-2.5, suitable growth temperature for 28-35°C. Because it can obtain the energy through oxidation of Fe^{2+} , sulphur, and metal sulfides, and it can take oxygen as the final electron acceptor to make a strong acid environment [13], and it was widely used in bioleaching of sulfide ore, coal biological desulphurization and environmental pollutions treatment fields [16-19]. But solubilization of radioactive plutonium with respect to soil has not been reported so far. Also most of the earlier studies only concentrated on the distribution of plutonium in soil. Hence the present study was designed based on these facts to assess the applicability of *A. ferrooxidans* for the removal of ^{239}Pu from low-level radioactive contaminated soil.

2. Materials and methods

2.1. Contaminated soil

Soil contaminated with ^{239}Pu was collected from Lop Nur. Firstly, the roots of plants and rubble need to be removed, and all through the sieve of 2.0 mm. Then, in order to provide for a single cumulative sample, all the soil samples need to be mixed completely. Finally, the soil were dried for 48 h at 110°C in thermoelectric thermostat drying box, then stored in jars [20-22]. Before the bioleaching, in order to reduce its volume, the soil samples need through grille screen, dual plate sieve and spiral separator. According to our experiments, particle size had an influence on the soil sample specific activity and when the particle size is larger than 0.25 mm the activity is far lower than the standard of China's provisional regulation. So soil samples used to mix with the *A. ferrooxidans* only when particle size less than 0.25 mm [23-25].

Representative physic-chemical properties of soil samples were determined, including particle size of soil, changes of pH, changes of content in organic matter, nitrogen and phosphorus and the activity of ^{239}Pu and the changes of the combination forms of ^{239}Pu is also determined.

2.2. Culturing of *A. ferrooxidans*

A. ferrooxidans was separated from acidic soil independently in this experiment, which has been preserved in China Center for Type Culture Collection for long-term (Accession number: CCTCC M208130, *Acidithiobacillus ferrooxidans* HT-3). *A. ferrooxidans* was pre-cultivated in a modified 9K liquid medium contains the following compositions in 1.0 l distilled water: 2.50 g of $(\text{NH}_4)_2\text{SO}_4$, 2.50 g of KH_2PO_4 , 0.50 g of $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$, 0.25 g of $\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$ and 40.40 g of $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$. Then added *A. ferrooxidans* culture medium 150 ml to a conical flask; added 20 ml liquid of *A. ferrooxidans* active bacteria to culture medium, culture at 28°C in thermostat oscillator.

2.3. Preparation of culture medium

The culture medium was rich in iron and sulfur and contained the following compositions in 1.0 l distilled water: 3.45 g powdered sulfur, 3.85 g of pyritic ash, 2.50 g of $(\text{NH}_4)_2\text{SO}_4$, 2.50 g of KH_2PO_4 , 0.50 g of

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