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Microbial mobilization and immobilization of plutonium

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Abstract

Microorganisms present in transuranic (TRU)- and mixed-wastes and in contaminated soils can bring about the transformation of plutonium (Pu) and affect its stability and mobility. Microbial dissolution of Pu is due to the production of sequestering agents, enzymatic oxidation of Pu(IV) to Pu(V) and Pu(VI) under aerobic conditions, or reduction of Pu(IV) to Pu(III) under anaerobic conditions. Immobilization is due to reduction of Pu(VI) or Pu(V) to Pu(IV), bioaccumulation extracellularly or intracellularly, and bioprecipitation. Disproportionation reactions of Pu(V) can also generate soluble Pu(VI) and insoluble Pu(IV) species. Plutonium is transported predominantly as colloids but microbes under appropriate conditions can affect the formation or destabilization of colloids. In this paper, the abundance of microbes in Pu contaminated soils and radioactive wastes and the mechanisms microbial dissolution and immobilization of Pu are reviewed.

Keywords: Plutonium; Bacteria; Oxidation; Reduction; Mobilization; Immobilization

1. Introduction

The presence of Pu in TRU and mixed wastes and in contaminated soils is a major concern because of the potential for migration from the waste sites and long-term contamination of the environment. Plutonium may exist in several oxidation states (III, IV, V, VI) and as various chemical species (salts, organic complexes, colloids) having a very complex chemistry and environmental behavior. The Pu in TRU waste may be present in various forms, such as oxide, coprecipitates, ionic, inorganic-, and organic-complexes depending on the process and waste stream. The various chemical species and oxidation states of Pu complicate assessing its environmental behavior. The toxicity and the long half-lives of its isotopes are the primary causes for concern. Soil pH, the presence of organics, redox conditions, mineralogy, and microbial activity affect the chemical speciation of Pu. Chelating agents present in TRU and mixed wastes form a strong complex with Pu(IV) and affect Pu mobility. In addition to the radionuclides, the TRU waste consists of a variety of organic and inorganic compounds in particular nitrate and sulfate. In the absence of oxygen, microbes can use nitrate and sulfate present in the waste as electron acceptors.

2. Microbiology and solution chemistry of plutonium

Plutonium(IV) is the most common and stable form in the environment. Soluble forms of Pu(V) and Pu(VI) are found in water and in marine environment [1]. Under appropriate conditions, microorganisms could affect the chemical nature of Pu by altering the speciation, solubility and sorption properties and thus could increase or decrease the concentrations of Pu in solution. Plutonium in the waste may be present initially as soluble, colloidal or insoluble forms and, after disposal, may be converted from one to the other by microorganisms [1–5]. Although the geochemical interactions of Pu have been extensively studied little is known of the biochemical mechanisms of Pu transformation in the environment. An increase in microbial activity will affect the redox and Pu oxidation state. Microbes can solubilize Pu due to production of organic acids such as citric acid and sequestering agents such as siderophores and transport Pu inside the cells. Decrease in Pu concentration in solution was attributed to an increase in biomass and removal of Pu by biosorption or bioaccumulation and bioprecipitation [1,4,5]. Biotransformation of Pu-organic complexes should result in the degradation

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Table 1
Abundance of microorganisms in Pu-contaminated soils and radioactive wastes

Sample source	Plutonium concentration	Microorganisms detected	Reference
Soil			
Nevada Test Site, area 13 (soil pH 8.4–9.2)	NA	Bacteria 2.9 ± 0.3 to $8.0 \pm 0.7 \times 10^6$/g Fungi $3.1 = 0.7$ to $24.6 + 2 \times 10^3$/g	[6]
Los Alamos (LANL), TRU waste shallow burial site TA-54, area C	NA	Bacteria Aerobes: 2.3×10^6/g Anaerobes: 3.3×10^6/g Actinomyctetes Detected with depth, not counted Fungi 4.9×10^4/g	[7]
Sediment/water			
Rocky Flats Plant Pond B-1 sediment (total Pu 81.5 mCi)	1000 dpm/g	Bacteria Aerobes: $3–300 \times 10^6$/ml Anaerobes: $0.2–2.0 \times 10^6$/ml	[8]
Water	NA	Aerobes: $1.1–340 \times 10^3$/ml Anaerobes: $2–7.8 \times 10^2$/ml	
Waste—solids			
^{239}Pu contaminated waste from a steel burial drum	NA	Bacteria Aerobes: 33–9500/sample Fungi: 46 CFU/sample	[7]
Waste—leachates			
Maxey Flats, Ky			
Trench 19S: ^{238}Pu	170,000 pCi/L	Bacteria	[3]
Trench 19S: ^{239,240}Pu	210,000 pCi/L	Aerobes: 2.2×10^2 CFU/ml Anaerobes: 3.2×10^2 CFU/ml	
West Valley, NY			
Trench 8: ^{238}Pu	160,000 pCi/L	Aerobes: 1.4×10^3 CFU/ml	[9]
Trench 8: ^{239,240}Pu	340 pCi/L	Anaerobes: 7.6×10^2 CFU/ml	

NA: not available.

of the organic ligand and precipitation of Pu. Microorganisms can play a major role in the generation and destabilization of colloidal Pu and thus affect their mobility in the environment.

2.1. Microorganisms in Pu contaminated sites

Microorganisms have been detected in low-level radioactive wastes, TRU wastes, Pu-contaminated soils, and in waste-repository sites slated for disposal of nuclear waste [3,4]. The abundance of microorganisms in Pu contaminated soils and wastes are summarized in Table 1 [3,6–9]. Low-level radioactive wastes, and TRU wastes contain low levels of Pu in addition to other radionuclides and organic compounds [3]. ^{238,239,240}Pu (gross alpha activity 1.7×10^5 pCi/L) was detected in leachate samples collected from the low-level radioactive-waste disposal sites: West Valley, NY, and Maxey Flats, KY [2,10,11]. Several aerobic and anaerobic bacteria were isolated from the leachate samples; among them, were *Bacillus* sp., *Pseudomonas* sp., *Citrobacter* sp., and *Clostridium* sp. The radioactivity and the organic chemicals present in the leachate were not toxic to the bacteria, which metabolized them, thereby producing-tritiated and carbon-14 methane [3,9]. Viable metabolically active microbes were detected at the LANL TRU waste burial site containing ^{239}Pu contaminated soil and flammable waste [12]. In general, Pu concentrations in the range of 10^{-7} to 10^{-3} M seem to affect most of the microorganisms studied [4,5,13–16].

2.2. Mechanisms of microbial dissolution and immobilization of plutonium

Plutonium exists in various oxidation states and the ones of concern are III, IV, V and VI. Electron donors and acceptors influence microbial activities and their presence could significantly affect the extent of dissolution and precipitation, particularly under anaerobic conditions. A slight increase in microbial activity (respiration) can alter the redox potential and reduce Pu(VI) to Pu(V) since the redox potential between Pu(VI) and Pu(V) is very small (Fig. 1). Reduction of Pu(V) to Pu(IV) either by bacterial action or by disproportionation reactions can occur. Although reduction of Pu from higher to lower oxida-

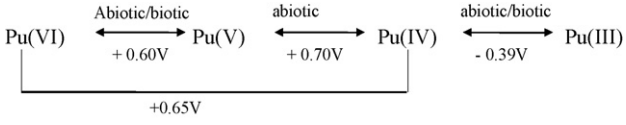

Fig. 1. Plutonium oxidation states and redox potentials at pH 8 [17].

tion state has received much attention microbial oxidation of lower valence Pu to higher state although possible has not been demonstrated yet.

Dissolution of Pu is brought about by direct enzymatic or indirect non-enzymatic actions of microorganisms. Direct enzymatic action includes reduction from higher to lower oxidation state or vice versa. Although, the enzymes involved and the biochemical mechanisms have not been elucidated, it should be similar to that of metal reduction reported in other microorganisms. Indirect non-enzymatic action includes dissolution by metabolic products, lowering of the pH and Eh of the medium and production of reductants such as Fe(II), Mn(II) and sulfides.

2.3. Reductive dissolution of Pu(IV) to Pu(III) by bacteria

Plutonium is present in the environment mostly as the oxides and hydroxides of Pu(IV) that have low solubility. The mobility and bioavailability of Pu(IV) is limited by its solubility. The solubility products of Pu(IV) oxyhydroxide are estimated to be $10^{-56.8}$ and $10^{-57.8}$. The solubility of plutonium(III) hydroxide is much greater, $K_{sp} = 10^{-22.6}$. Consequently, the reduction of Pu(IV) oxyhydroxides to Pu(III) is expected to increase the solubility of plutonium in the environment. The persistence of the reduced soluble Pu(III) is affected by its stability, as well as by its interactions with organic and inorganic materials. Rusin et al. [18] showed that an iron-reducing bacterium *Bacillus* sp. solubilized up to 90% hydrous PuO_2(s) in about 7 days under anaerobic conditions in the presence of nitrilotriacetic acid (NTA). In these studies, Pu(III) was present as a Pu–NTA complex. However, without NTA, only 40% of Pu was solubilized suggesting that a strong complexing agent is needed to keep the Pu in solution. Little dissolution of PuO_2 was observed in sterile culture media or in the presence of non-iron-reducing bacteria, such as *Escherichia coli*.

We investigated reductive dissolution of Pu(IV) by the anaerobic bacterium *Clostridium* sp. Addition of 1×10^{-7} M ^{242}Pu(IV)-nitrate had no effect upon the growth and metabolism of glucose of *Clostridium* sp. Plutonium added to the bacterial growth medium (uninoculated control) resulted in its precipitation and was removed by 0.4 μm filtration. Speciation calculations showed that Pu most likely existed as $Pu(OH)_4$ at pH 6.2 due to hydrolysis and polymerization. The growth of the bacterium lowered the Eh of the medium from +50 mV to −180 mV, and the pH from 6.2 to 2.8, concomitant with the production of acetic and butyric, acids and carbon dioxide (225 μmol). After 14 h of growth, 70% of the Pu passed through a 0.4 μm filter and 55% passed through a 0.03 μm filter suggesting a significant portion of Pu was solubilized. Solvent extraction by thenoyltrifluoroacetone (TTA) confirmed a decrease in the polymeric form of Pu and an increase in the soluble fraction, suggesting the presence of Pu^{3+}. XANES analysis of Pu at L_{III} edge showed that the sample inoculated with *Clostridium* sp., the absorption edge position had shifted to lower energy at 18.064 keV compared to Pu(IV) nitrate (18.068 keV). This shift of −4 eV is similar to that observed for the trivalent Pu oxidation state and confirms the presence Pu(III) (Fig. 2). The Eh of the medium was low and the CO_2 concentration high, thus favoring the reduction of Pu from the tetravalent to the trivalent state. These results suggest that under appropriate conditions Pu can be reduced to Pu(III) by anaerobic bacteria (Francis et al., in preparation).

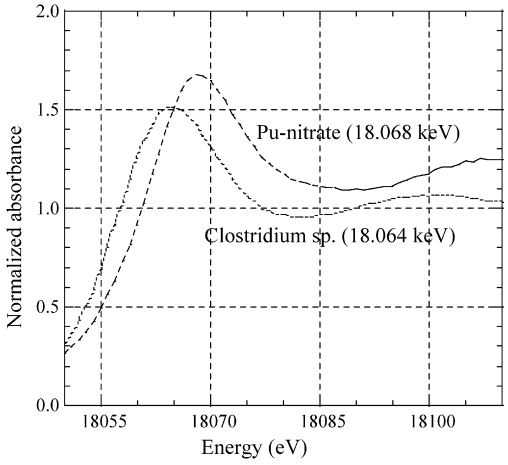

Fig. 2. XANES analyses of Pu before and after bacterial action (Francis et al., in preparation).

2.4. Dissolution of Pu by metabolic products

Dissolution of Pu by indirect actions of microorganisms is brought about by their production of organic acids, such as citric acid, extracellular metabolites, and siderophores [19]. As there are chemical and biochemical similarities between Pu(IV) and Fe(III), and between Th(IV) and Pu(IV), iron-sequestering agents could be important in the complexation of Pu and other actinides, thus increasing their solubilization and bioavailability. *Pseudomonas aeruginosa* isolated from a Pu-contaminated pond at Rocky Flats capable of bioaccumulation of uranium, also elaborated several chelating agents for thorium and uranium when grown with these metals [20]. Microorganisms grown in iron-deficient medium are known to elaborate specific iron chelators. For example, dissolution of plutonium dioxide was enhanced in the presence of Desferal, a polyhydroxamate chelate produced by microorganisms [12]. Desferrioximine and enterobactin isolated from *E. coli* solubilized hydrous plutonium(IV) oxyhydroxide [21].

Microorganisms grown in the presence of plutonium produced complexing agents [13]; these agents may transport plutonium into the cells [5,22]. Thus, the potential exists for dissolution of actinides in wastes by microorganisms, thereby increasing their bioavailability and mobility. For example, microorganisms grown in the presence of plutonium produced complexing agents, such as citric acid and several unidentified compounds capable of dissolving and mobilizing Pu in soils [23]. Several bacteria and fungi grown in the presence of Pu produced extracellular Pu complexes that increased the concentration of Pu in soil-column eluates relative to controls. Elution through soil effectively removed positively charged Pu complexes. The increased mobility of Pu in soil resulted from the formation of neutral and negatively charged Pu complexes [23]. Biotransformation of Pu-organic complexes should result in the degradation of the organic ligand and precipitation of the actinide.

2.5. Mechanisms of microbial immobilization of plutonium

Immobilization or precipitation of radionuclides is due to changes in the Eh of the environment, enzymatic reductive precipitation (reduction from higher to lower oxidation state), biosorption, bioaccumulation, biotransformation of radionuclide-organic and -inorganic complexes, and bioprecipitation reactions.

Immobilization of Pu by microbes may be due to indirect action by changing the Eh of the environment, and facilitating abiotic precipitation of Pu by reduction from higher to lower oxidation state, biosorption by bacteria, and bioprecipitation reactions [4,15,24–26]. The chemical form and the type of the association of Pu with the bacteria have not fully been elucidated. Reductive precipitation of Pu(VI) to Pu(IV) by resting cells of microorganisms have been observed [1,24]. Enzymatic reduction of soluble Pu(VI) and Pu(V) to Pu(IV) by cell suspensions of *Shewanella putrifaciens*, *S. oneidensis* and *Geobacter metallireducens* under anaerobic conditions have been reported [1]. Sorption studies of Pu(VI) with *Bacillus sphaericus* showed that Pu(VI) was reduced with increasing contact time to Pu(V). The cell bound Pu(V) was released into solution due to its weak complexation and it disproportionated to Pu(IV) and Pu(VI). The oxidation of Pu associated with bacteria was confirmed by XANES and TTA extraction [24,25]. Characterization of Pu associated with the cells by EXAFS spectroscopy showed that Pu(VI) was primarily bound to the phosphate groups on the cell surface but no carboxylate complexation was observed [25].

2.6. Immobilization of plutonium by biosorption and/or bioaccumulation processes

In nature, bacteria interact with metal ions to immobilize and concentrate them, eventually generating minerals. Microbial biofilms bind significant quantities of metallic ions, and also serve as templates for their precipitation. These processes have received considerable attention because of their potential for bioremediating radionuclide-contaminated sites and wastestreams. Although the biochemistry of the interactions of toxic metals and uranium with bacterial cell walls, extracellular biopolymers, and microfossil formations has been extensively studied, we have limited information on microbial immobilization of Pu [4]. Of particular concern is the long-term stability of the immobilized actinides that may undergo subsequent remobilization.

Keith-Roach et al. [27] showed minimum Pu (and Am) concentrations in water column correspond to maximum biomass and proposed that the data to be in support of a bioaccumulation process (e.g., biosorption or bioprecipitation). Kauri et al. [28] investigated the ability of five strains of bacteria isolated from soil in Japan that had been contaminated with Pu by fallout for more than 40 years to bind low concentrations of Pu^{4+} during their growth. Although Pu was associated with all the bacterial strains studied, the association varied with the type of bacterial isolate indicating differences in the mechanisms of binding. For example, the interactions were examined between of ^{241}Pu nitrate and *Halomonas* sp. isolated from the Waste Isolation Pilot Plant (WIPP) site and *Acetobacterium* sp., isolated from alkaline groundwater at the Grimsel Test Site, Switzerland, at pH 5.0. Both cultures were grown to late log-phase, washed in the appropriate electrolyte, and diluted to an OD of 0.4. *Halomonas* sp. biosorbed 9% (0.17×10^{-9} M) of the total ^{241}Pu in solution (1.8×10^{-9} M) [29]. *Acetobacterium* sp. biosorbed 7% (0.11×10^{-9} M) of the total ^{241}Pu in solution at the highest concentration tested (1.5×10^{-9} M) [29]. On a dry-weight basis, *Acetobacterium* sp. sorbed 145 ng ^{241}Pu g^{-1} dry cells, and *Halomonas* sp. sorbed 351 ng ^{241}Pu g^{-1} dry cells. The extent of biosorption depends upon the form and chemical speciation of the Pu species present, with only a fraction of it being bioavailable in these studies.

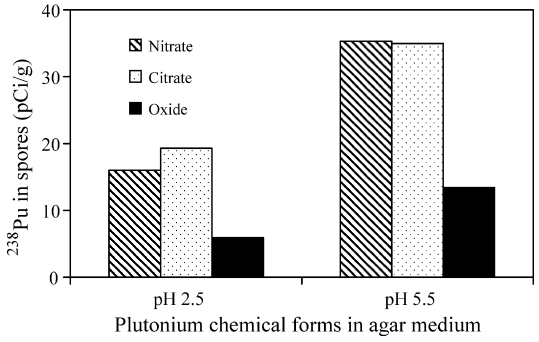

Fig. 3. Dissolution and uptake of ^{238}Pu by *Aspergillus niger* [22].

Giesy et al. [30] investigated the effect of naturally-occurring organics on Pu uptake by the fresh-water bacterium *Aeromonas hydrophila* and by the alga *Scenedesmus obliquus*. The organic matter was concentrated from the waters of Skinface Pond, near Aiken, South Carolina, and separated by membrane ultrafiltration into four nominal diameter-size fractions (*F* I > 0.0183; 0.0183 > *F* II > 0.0032; 0.0032 > *F* III > 0.009; *F* IV < 0.009 μm). Each fraction was introduced into cultures of *S. obliquus* and *A. hydrophila* at the concentrations found in nature. ^{237}Pu uptake was determined in log phase cultures after 6 h incubations. The initial Pu concentration in each flask was 1.1×10^{-4} μCi/ml $^{237}Pu(NO_3)_4$. Fractions I and II reduced ^{237}Pu uptake by *S. obliquus*, *F* IV increased uptake, and *F* III had no effect. ^{237}Pu uptake by *A. hydrophila* was no different in the presence of *F* I, *F* II, or *F* III than in tryptic-broth medium alone, whereas *F* IV increased its ^{237}Pu uptake.

The sorption of ^{239}Pu from aqueous nitrate medium was studied using the fungus biomass *Rhizopus arrhizus*. The biosorption of ^{239}Pu was maximal at pH 6–7, and this fungal biomass appears to be a promising sorbent treating radioactive effluents from the nuclear industry [31]. Plutonium uptake by *Aspergillus niger* mycelium its transport to spores have been reported. Studies were carried out on the influences of different chemical forms and concentrations of ^{238}Pu in culture medium at pH 2.5 and 5.5 on the uptake and transport of Pu by the mycelium to the spores of the common fungus *A. niger* (Fig. 3). When Pu was added to the culture medium as dioxide microspheres, or as Pu-nitrate, or Pu-citrate, it was transported to the spores; there was an almost linear relation between its transport and concentration. Raising the pH of the culture medium from 2.5 to 5.5 generally increased

the transport of all three chemical forms. At Pu concentrations of 224 pCi/g, at both pH 2.5 and 5.5, the transport of Pu to the spores was approximately three-fold greater from the nitrate- or citrate-form as from the dioxide microspheres. The derived transport factors indicated that Pu was concentrated in the mycelium and then further transported to the aerial spores of this fungus. A new, simple technique for collecting spores was developed to prevent cross-contamination of the spores with mycelial fragments, and by direct contact with the Pu-containing agar medium. The specific activities of the spores grown at pH 5.5 generally were at least twice those of the spores grown at pH 2.5. The uptake of Pu dioxide was approximately 33% of that from the nitrate- and citrate-forms at both pH levels. These findings suggest that this common soil fungus may solubilize soil-deposited Pu and render it more bioavailable for higher plants and animals [22]. If a similar process occurs in Pu-contaminated soils, it could be an important link in transferring of soil-deposited Pu to humans and would also explain the apparent time-dependent increases in the uptake rate of Pu by plants grown in contaminated soils [22].

3. Role of microbes in the mobilization and immobilization of Pu from contaminated soils

Chemical characterization of Pu at contaminated sites shows that its environmental form varies according to the site and the waste stream. For example, at Rocky Flats, CO, the predominant form appears to be as PuO_2(s); at the Nevada Test Site Pu was associated with mineral colloids; while, at Oak Ridge, TN it is associated with organic matter and at Mayak Production Association, Urals, Russia, bound to iron oxide colloids [32–35]. Plutonium is generally considered to be relatively immobile; however, its transport, albeit at very low concentrations, has been observed at several DOE sites including Rocky Flats (RF), Los Alamos National Laboratory (LANL), and Nevada Test Site (NTS), and Mayak Production Association, Urals, Russia [32–36]. Plutonium in surface waters at the Rocky Flats Environmental Technology Site has been shown to be associated with a 10,000 Da organic macromolecule as colloids [37]. However, the microbial effects on the stability and mobility Pu colloids have not been evaluated.

Studies with Pu contaminated soils from NTS site show that Pu and Am are remobilized due to enhanced aerobic or anaerobic microbial activity and that the type of addition of carbon source affected the rate and extent of dissolution (Dodge and Francis, unpublished results). Studies are under way to determine the microbial mechanisms of dissolution of Pu from contaminated soils.

4. Conclusion

Microorganisms can affect the solubility and mobility of Pu by oxidation–reduction reactions, by the production of sequestering agents, and by bioaccumulation and bioprecipitation processes. Although the physical, chemical, and geochemical processes affecting dissolution, precipitation, and mobilization of Pu have been investigated, we have only limited information on the interactions of microorganisms with Pu and the effect of microbial processes on the mobilization and immobilization of Pu. Fundamental understanding of the mechanisms of microbial transformations of various chemical forms of Pu under various microbial process conditions, such as aerobic, anaerobic (denitrifying, fermentative, and sulfate reducing) and repository relevant conditions would help to assess the microbial impact on long-term behavior of Pu during on-site storage, shallow land-burial, and disposal in deep geological formations as well as management of contaminated sites.

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