

Microbial Biogeochemistry of Uranium Mill Tailings

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I. Introduction	113
II. Sulfur Cycle	114
III. Iron/Manganese Cycle	115
IV. Uranium Reduction	117
V. Nitrogen Cycle	118
A. Nitrification	118
B. Denitrification	119
VI. Fungi	120
VII. Biosorption	121
VIII. Conclusions	124
References	125

I. Introduction

Uranium mill tailings (UMT) are the crushed ore residues from the extraction of uranium (U) from ores. Except where ores have a high carbonate content, the commercial extraction of U from ores generally involves leaching with sulfuric acid (H_2SO_4). The effluent (termed *raffinate* or *barren solution*) and tailings from the mill are discharged as slurry to a waste-retention pond or to mined-out underground workings for disposal. As the ores typically are low grade, essentially all of the tonnage of ore processed at the mill is disposed of as tailings. The present inventory of UMT in the United States is about 240 million tons. The final product of the milling process is a U concentrate called *yellowcake*. U extraction efficiencies at commercial mills range from about 65% to 95% (Mudd, 2000). Because solubilization of ^{226}Ra from ore minerals in both sulfuric acid and sodium carbonate is low, the tailings remain a radiological hazard. Safe management of tailings has been the focus of regulatory and environmental research attention since the 1950s.

Among the radioactive wastes associated with the nuclear fuel cycle, UMT are unique in terms of their volume and their limited isolation from the surficial environment. For this latter reason, their management and long-term fate has many interfaces with environmental microbial communities and processes.

II. Sulfur Cycle

Because of the H_2SO_4 used to extract U from the ores at acid-leach mills, ^{226}Ra is believed to occur in UMT as a sparingly soluble sulfate phase, probably coprecipitated with Ba and other alkaline earth sulfates (Goulden *et al.*, 1998) and lead sulfate (Steger and Legeyt, 1987). Sulfate-reducing bacteria have been identified in U heap leaching piles (Schipper *et al.*, 1995) and UMT (Hard *et al.*, 1999). Fortin and Beveridge (1997) note that sulfate reduction in sulfidic mine tailings is most likely limited by the supply of organic carbon. In UMT, organic carbon supplies may include solvents used in the milling circuit and discharged to the tailings impoundment (such as kerosene) and carbonaceous ores such as lignites. Cover and codisposal practices at UMT such as the use of a wood chip cover (at the Dawn U mill in eastern Washington; U.S. Department of Energy, Office of Environmental Management, 1999) and of municipal waste (Helling *et al.*, 1998) can add bioavailable carbon sources to the underlying UMT. Sewage sludge and other organic amendments may also be used during decommissioning activities to promote establishment of a vegetative cover and to limit oxygen penetration and pyrite oxidation.

Laboratory studies by Landa *et al.* (1986) showed enhanced leaching of ^{226}Ra from acid-leach UMT by the action of sulfate-reducing bacteria. Recent studies by Martin *et al.* (2003) have shown the process to be operative in water-covered (>2 m water depths), acid-leach/lime-neutralized UMT at Elliot Lake, Ontario, Canada. Despite the input of oxygen from photosynthesis by a dense, standing crop of mat-like submerged aquatic vegetation (*Chara* sp.; “muskgrass”), anoxic conditions occur at the tailings water interface as a result of oxygen consumption by microbial respiration of decomposing biomass. Sulfate-reducing conditions are evident at about 4 cm above the benthic boundary and persist into tailings. The flux of Ra and Ba into the overlying water is much higher in the regions associated with *Chara* as compared to shallow regions (<1.5 -m water depths), where only sparse filamentous vegetation is present. Indeed, the observed increases in ^{226}Ra concentrations in the tailings pond water over a 5-year period at this site seem to be related to the gradual increase in the spatial extent and density of this benthic flora.

Martin *et al.* (2003) suggest that vegetation control measures (such as water lowering to take advantage of ice scouring of the pond bottom during the winter) should be considered to minimize ^{226}Ra release from the submerged tailings into the overlying pond water. It is of interest to note that living *Chara* is known to concentrate radium (Ra) strongly

from water (Smith and Kalin, 1989). Thus, the presence of *Chara* not only indirectly influences redox conditions in the underlying tailings but also probably directly affects the flux of ^{226}Ra to and from them.

Just as sulfate-reducing microorganisms may enhance Ra release, sulfur-oxidizing microorganisms may diminish it by raising aqueous sulfate concentrations, thereby suppressing the solubility of compounds such as Ba(Ra)SO_4 . Laboratory studies by Pérez and Torma (1985) showed that both ^{226}Ra release from acid-leach UMT in aerobic flasks and percolation columns decreased (compared to sterilized controls) in systems inoculated with *Thiobacillus ferrooxidans*. These studies have implications for the weathering of pyritic UMT and the modeling of ^{226}Ra release for long-term radiological assessments of UMT disposal (see, e.g., Fernandes *et al.*, 1996). In addition to their inferred occurrence in UMT, discrete Ba(Ra)SO_4 particles may be released to streams from settling ponds used to treat U mine drainage waters (Essed, 1981). Such materials may be deposited in aquatic environments where the action of sulfate-reducing or sulfur-oxidizing microorganisms may be important. As an example of the latter, Willett *et al.* (1994) showed that aerobic incubation of acid-leach UMT with acid sulfate soils typically resulted in a decrease in the fraction of the total ^{226}Ra that was exchangeable, indicating immobilization as an insoluble sulfate phase.

$^{210}\text{Polonium}$ (^{210}Po ; half life = 138 days) occurs further down the ^{238}U decay chain. As a member of group VI A of the periodic table, the chemistry of Po resembles that of sulfur. Under sulfate-reducing conditions in groundwater aquifers (Harada *et al.*, 1989) and phosphogypsum piles (Burnett *et al.*, 1995), ^{210}Po may be released to interstitial waters by the action of sulfate-reducing bacteria. The process is likely operative in UMT as well. Once solubilized, ^{210}Po may migrate as an aqueous species, coprecipitate as a metallic sulfide if sulfide concentrations are high (Burnett *et al.*, 1995), or be subject to microbially mediated volatilization, possibly as dimethyl polonium (Hussain *et al.*, 1995; Momoshima *et al.*, 2001).

III. Iron/Manganese Cycle

UMT tend to be high in iron and manganese because of native concentrations in ores, iron introduced to processed ore by abrasion of iron rods in the grinding circuit, and additions of MnO_2 and ferric salts to the leaching circuit as oxidants for tetravalent U. Selective leaching trials have shown iron and manganese oxides to be important hosts of ^{226}Ra in acid-leach UMT (Landa, 1982; Somot *et al.*, 1997). The

dissolution of these oxides by the action of the Fe(III)-reducing bacteria can release Ra to contacting waters (Landa *et al.*, 1991). Data presented in the study by Martin *et al.* (2003) described above indicate that iron reduction occurring in the tailings at the shallow site may be contributing Ra and Ba to the water column, although not in large amounts. In the case of Mn^{2+} oxidation, Mathur and Dwivedy (1988) showed that precipitation of hydrous oxides of manganese by *Arthrobacter* sp. was able to remove more than 95% of the ^{226}Ra in UMT pond effluent.

Arthrobacter is a genus of particular interest. Miller *et al.* (1987) showed a striking preponderance of the *Arthrobacter* sp. in sandy UMT sampled at the Edgemont, South Dakota; Maybell, Colorado; and Rifle, Colorado, sites. As part of this investigation, Updegraff *et al.* (1986) provided additional characterization of the nutritional requirements of these bacteria; earlier work by Updegraff and Douros (1972) had shown members of the genus *Arthrobacter* to also be predominant in U ore samples collected at Uravan, Colorado. This apparent association with U-bearing materials is intriguing. Suzuki and Banfield (2001) showed an isolate from the Midnite U mine in eastern Washington state that closely resembles (based on sequencing of 16S RNA) *Arthrobacter ilicis* to be both resistant to U toxicity (80 ppm test incubation) and to incorporate U intracellularly; the authors also suggest that the isolate might have possible utility in bioremediation of actinide-contaminated soil and water.

Siderophores are low-molecular weight, iron-chelating ligands synthesized by plants and microorganisms under iron-limited conditions. Their ability to bind actinides has received considerable attention, and indeed siderophore analogs may find engineering applications in nuclear fuel reprocessing (Renshaw *et al.*, 2003). Kalinowski *et al.* (2004) have investigated the role of siderophores in the leaching of U from uraniferous, pyritic shales (previously mined in Sweden as a U ore). Materials from abandoned ore piles with about 30 years of in-place, postmining weathering were examined in short-term laboratory experiments. With the sulfuric acid leaching that had occurred in these materials during the 30-year weathering period, the starting material of the lab trials approximates UMT. In the presence of aerobic cultures of siderophore-producing *Pseudomonas fluorescens*, U leaching rates increased about 50-fold compared with uninoculated controls (pH rose from 4.5 to 9.3 over 8 days with *P. fluorescens* growth). For unweathered ore, the increases in leaching rates in the presence of *P. fluorescens* were about 1000-fold; the differences in U leaching rates likely reflect that the weathered material has only residual U in resistate phases present. This work indicates that siderophores may be of

significance in the mobilization of U from ore and mill tailings under aerobic, neutral-to-alkaline conditions.

IV. Uranium Reduction

Lovley *et al.* (1991) demonstrated the enzymatic reduction of soluble U(VI) to U(IV), which precipitated as uraninite (UO₂) (Gorby and Lovley, 1992). Since then, a large body of work has been carried out documenting the range of microorganisms involved, physiological constraints on the process, and applications to bioremediation of U-contaminated groundwater by *in situ* and pump-and-treat operations. These studies have recently been reviewed by Lloyd and Macaskie (2000). UMT sites have been the focus of many of the field studies looking at both natural and accelerated bioremediation.

The presence of indigenous bacteria capable of U(VI) reduction was verified in groundwater sampled at three UMT sites (both acid- and alkaline-leach mills) studied by Abdelouas *et al.* (2000). The addition of organic carbon (optimally as ethanol) and phosphate (as sodium trimetaphosphate) was required to initiate the reduction of the U present in the groundwater downgradient of the mills. Investigations on sediment and water from the U-contaminated aquifer at the Shiprock, New Mexico, UMT site by Finneran *et al.* (2002) also indicate that the addition of a carbon source (optimally, acetate or glucose) would stimulate the rate of U removal by microbial reduction.

A detailed discussion of microbial communities in aquifers affected by UMT is beyond the scope of this review. Most attention with respect to passive or engineered removal of U from such groundwaters has focused on iron-reducing and sulfate-reducing bacteria (Anderson and Lovley, 2002; Chang *et al.*, 2001; Holmes *et al.*, 2002). Fermentative bacteria, such as species of *Clostridium*, may also be involved in U reduction at these sites (Selenska-Pobell, 2002; Suzuki *et al.*, 2003).

Where microbial reduction of U(VI) occurs in anaerobic portions of aquifers, uraninite is deposited. In later years, these subsurface zones may be contacted by oxygenated groundwater. Thus, the possible reoxidation of biogenic uraninite is a concern in terms of the long-term success of bioremediation of U-contaminated aquifers. Suzuki *et al.* (2002) induced anaerobic conditions in sediment–water mixes from the Midnite U mine in Washington state and showed that the average particle size of the biogenic uraninite produced by native microflora was only about 1.5 nm. Such small particles may be prone to rapid reoxidation, although studies by Abdelouas *et al.* (1999) on groundwater and sediment from the UMT site at Tuba City, Arizona, indicate that

preferential oxidation of other aquifer components, such as the iron sulfide mackinawite, may limit reoxidation of biogenic uraninite. Another contaminant mobility issue noted by Suzuki *et al.* (2002) is that facilitated transport of these U(IV) nanoparticles may be possible unless they are flocculated by solutes in the groundwater or physically trapped by extracellular polysaccharides.

The initial studies of microbial U reduction focused on soluble U(VI) species. Recent studies have shown that U(VI)-sorbed species and the U(VI) lattice components in minerals may also be reduced to U(IV). For example, Haas *et al.* (2002) showed that U(VI) sorbed to kaolinite was readily reduced by *Shewanella putrefaciens*, but that the reduction of U(VI) sorbed on montmorillonite was much less effective. Fredrickson *et al.* (2000) demonstrated that *S. putrefaciens* could reduce the U(VI) oxide metaschoepite ($\text{UO}_3 \cdot 2 \text{H}_2\text{O}$) to the U(IV) oxide uraninite. Thus, microbial reduction of U should be considered a possible reaction at multiple stages in groundwater flow path investigation and simulation at UMT sites, even when sorption or coprecipitation occur. In addition to enzymatic U(VI) reduction by bacteria, abiotic, surface-catalyzed U(VI) reduction by Fe(II) has been reported by Liger *et al.* (1999). Surfaces such as nanoparticulate hematite can serve as both the sources of Fe(II) (by reductive dissolution) and the sorption surfaces for U(VI) at oxic/anoxic transition zones in aquifers at UMT sites and other settings. The authors suggest that at near-neutral pH and in the absence of excessive desorption of U(VI) by the formation of strong aqueous carbonate and hydroxy carbonate complexes, this reaction may be kinetically competitive with enzymatic reduction.

V. Nitrogen Cycle

A. NITRIFICATION

Ammonia is used in large quantities at U mills to precipitate the extracted U as ammonium diuranate (yellowcake). Nitrate-containing reagents were generally used in limited quantities, yet one sees nitrate contamination in surface and groundwaters at many UMT sites. The nitrates appear to have formed by microorganisms using the abundant ammonia as a substrate. Populations of both autotrophic, ammonia-oxidizing bacteria (AAO) and denitrifying bacteria have been reported at UMT sites. Using molecular methods, Ivanova *et al.* (2000) have characterized the population of AAO in groundwater sampled in the contamination plume downgradient from the UMT at Shiprock, New Mexico. (Uranium milling ceased at this site in 1968; U.S. Department of Energy, 1998). Groundwater at this site had concentrations of up to

7800 mg L⁻¹ nitrate and 630 mg L⁻¹ ammonia. Ivanova and coworkers estimated the AAO population size by measuring the concentration of the functional gene-marker encoding the ammonia monooxygenase (*amoA*), the enzyme mediating the first step in nitrification. A good correlation ($r = 0.76$) was found between the nitrate concentration and the quantity of *amoA* gene present in the groundwater, supporting the view that the AAO mediate nitrification in this system.

In addition to its inherent role as a nutrient and groundwater contaminant, nitrate present within the tailings can influence radionuclide release from these materials. Anaerobic, nitrate-dependent, microbial oxidation of ferrous iron can increase pyrite oxidation (Landa *et al.*, 2000; Straub *et al.*, 1996) within the tailings, resulting in increased U and Th leaching and suppressed Ra release from solid phases such as BaSO₄. In addition, transport of nitrate from the tailings can potentially decrease rates of U reduction in the subsurface by providing anaerobic bacteria with a more energetically favorable terminal electron acceptor than U(VI) (Ivanova *et al.*, 2000).

B. DENITRIFICATION

Longmire and Thomson (1992) have presented evidence for denitrification at the Maybell, Colorado, UMT site. In contrast to a dispersed chloride plume, nitrate concentrations decreased from two to four orders of magnitude immediately downgradient of the UMT impoundment. Microcosm studies using core materials from 10 to 40 m in depth slurried with groundwater that was amended to nitrate levels close to the maximum groundwater concentration observed at the site all showed rapid denitrification, with an average nitrate removal of 40% in 2 weeks. No remedial action is planned for the Maybell site, and denitrification is one of the geochemical processes being relied on here for natural attenuation of nitrates (U.S. Department of Energy, 2003). Barton *et al.* (1994) extended these studies and demonstrated with culturing techniques the presence of denitrifying bacteria in high-nitrate groundwaters at four UMTRA sites. Abdelouas *et al.* (1998) showed the presence of two species of denitrifying bacteria (*Psuedomonas aeruginosa* and *Psuedomonas stutzeri*) in groundwater sampled at the alkaline-leach, Tuba City, Arizona, UMT site. In anaerobic microcosm experiments with C and P amendments to optimize reaction rates, the nitrate concentration was lowered by denitrification from about 1400 mg L⁻¹ to less than 0.1 mg L⁻¹ in 10 days at 24°C.

In contrast to the Maybell site described above, the buildup of nitrate and lack of nitrite at Shiprock (Ivanova *et al.*, 2000) indicate that

denitrification is limited at that site. As nitrate is a contaminant of concern at many UMT sites, these contrasting results between the Maybell and Shiprock sites are of real interest. Recent landfill studies by Senko *et al.* (2002) have shown the intermediate products of denitrification (nitrite, nitrous oxide, and nitric oxide) were able to oxidize U(IV) and thereby mobilize previously reduced U. Environmental factors, such as availability of carbon, that might be controlling rates of denitrification in the subsurface sediments at UMT sites would appear to be an area of considerable importance when considering U mobility. To ensure long-term immobilization of U by microbial reduction of U(VI), the cycling of N needs to be considered and the removal of nitrogen species (initially introduced as NH_4^+) from the UMT system appears to be a desirable initial goal (Elias *et al.*, 2003).

As nitric acid is used to dissolve fuel rods in reprocessing plants, nitrates are a major contaminant of concern at these and other fuel cycle facilities. Thus, the linkage of the N cycle to U reduction has implications beyond UMT sites. The U.S. Department of Energy is currently sponsoring research by researchers at Stanford University that looks at pump-and-treat, fluidized bed reactors to promote nitrate removal from groundwater by denitrification (Gu *et al.*, 2003). The goal will be complete reduction of NO_3^- to N_2 , to avoid the oxidation of U(IV) by denitrification intermediate products noted by Senko *et al.* (2002). This aboveground treatment of the groundwater is planned as the initial step, before *in situ* biostimulation of U reduction at an Oak Ridge, Tennessee, field site with nitrate concentrations of up to 8 g/L.

VI. Fungi

Kalin and Stokes (1981) reported on the occurrence and metal content of macrofungi at abandoned UMT sites in the Elliot Lake and Bancroft areas of Ontario, Canada. The authors noted the consistent association of two of the fungi with balsam poplar roots and suggested that mycorrhizal associations might aid in the establishment of vegetation on UMT by enhancing nutrient uptake or by restricting uptake and translocation of heavy metals. Recent work by Rufyikiri *et al.* (2002) has shown that the arbuscular mycorrhizal fungus, *Glomus intradices*, can assimilate U from solution culture and translocate it to excised carrot roots. Future research may determine whether U hyperaccumulates in the arbuscular mycorrhizal-colonized roots, or whether enhanced U translocation to shoots occurs. This research has great promise in designing phytoremediation strategies for UMT and U-contaminated soils.

Zhdanova *et al.* (2003) have shown the tendency of some soil fungi to grow toward radioactive sources (“radiotropism”), colonize, and promote the dissolution of hot particles (HPs) from the Chernobyl reactor accident and the Semipalatinsk nuclear weapons test site. Bioaccumulation of radionuclides, in this case ^{137}Cs and ^{152}Eu , was demonstrated, even in the absence of hyphal contact with the HPs, indicating a role of exometabolites in the dissolution of the HP constituents. It has been suggested that mycorrhizal fungi may also increase the release of calcium from apatite in soils (Blum *et al.*, 2002). Stieff (1984) noted the presence of ^{226}Ra HP in UMT at Monticello, Utah. Landa *et al.* (1994) showed the presence of ^{210}Pb -bearing PbSO_4 HPs that precipitated from laboratory-scale U milling solutions. The radiotropic behavior of select soil fungi perhaps may have a role in determining the long-term physical and chemical stability of such sparingly soluble HPs in UMT piles and the surrounding depositional environments.

VII. Biosorption

Biosorption refers to the binding of metals by ligand interactions or ion exchange reactions to the living or dead microorganisms. Uranyl salts have a long history of usage as stains for the microscopic study of microbial cells: the uranyl ion (UO_2^{2+}) reacts with phosphate and amino groups to stain various cellular components (Bozzola and Russell, 1992). During the early 1980s, investigations by Tsezos and Volesky (1982), Strandberg and Shumate (1982), Tsezos and Keller (1983), and Galun *et al.* (1984) showed that fungal biomass had a high capacity for retention of U, thorium (Th), and Ra. Systems for U removal from bioleach (i.e., by action of sulfur-oxidizing bacteria)—heap leaching solutions (Denison Mines, Elliot Lake, Ontario)—by nonliving fungal biosorbents have been shown to be effective in bench-scale tests (Tsezos *et al.*, 1989). Immobilized cells of a metal-tolerant *Citrobacter* sp. have been used for U removal from acidic mine drainage waters at the ENUSA mine site in Salamanca, Spain (Basnakova *et al.*, 1998). In addition, biosorption undoubtedly is part of the contaminant-removal mechanism in constructed wetland-type, effluent treatment systems at U mills [see, e.g., Brierley and Brierley, 1981; Federal Ministry of Economics and Technology (Germany), 2000], and in U-contaminated plumes in shallow aquifers.

The high concentrations of Fe and Al in UMT effluent may limit the efficiency of such treatment systems. Iron (Galun *et al.*, 1984; Tsezos and Volesky, 1982) and aluminum (Tsezos *et al.*, 1997) have been shown to interfere with U sorption by fungal biomass (*Rhizopus*

arrhizus and *Penicillium digitatum*). A steric hindrance by colloidal hydrolysis products of Fe and Al seems to be the likely cause of the lowered U uptake (Tsezos, 1983). In addition, high silica in pH 4 bioleach solutions from the Denison Mines solutions that were subsequently contacted with *R. arrhizus* resulted in the precipitation of aluminosilicates, as well as aluminum hydroxides, on (and within) mycelia, which likewise inhibited U sorption. Hard *et al.* (1999) have shown sulfate-reducing bacteria isolated from U mining waste to also accumulate large quantities of extracellular aluminum, presumably as an amorphous hydroxy precipitate. Such biogenic precipitates can influence both radionuclide migration from UMT and biosorption of radionuclides in effluent treatment systems.

The use of waste materials and industrial byproducts can be a low-cost alternative to the dedicated production of biosorbents. Tsezos *et al.* (1987) showed pelletized, municipal sewage sludge to be an effective, high-capacity biosorbent of ^{226}Ra from U mining and milling wastewater (neutralized to pH 7.5) from Elliot Lake, Ontario. Smirnov *et al.* (1995) of the All-Russian Research Institute of Chemical Technology (ARRICT) have proposed the use of mycelial biomass (available as a waste byproduct from the commercial production of antibiotics) in a two-stage process for the treatment of UMT. In the first stage, acid from the sulfuric-acid extraction circuit of the U mill is leached with 1–3 M alkaline earth chloride or nitrate solutions (e.g., CaCl_2) to leach ^{226}Ra . This acidic leachate is then contacted with the mycelial biomass (the testing here used biomass from erythromycin production; *Streptomyces erythraeus*) to bind the Ra. Removal of Ra was enhanced by the addition of carrier barium, allowing the second-stage process to reduce the ^{226}Ra concentration to below the Russian discharge limit of 54 pCi/L.

An added benefit in terms of release of a more environmentally benign effluent was the pH neutralization achieved with acidic solutions by the ARRICT. Georgescu *et al.* (1999) have shown waste yeast from fermentation processes in the pharmaceutical industry to achieve significant neutralization of alkaline waste waters from U ore processing. A pH 10 effluent treated in a 10-cycle, flow-through column achieved 61% U removal (better than activated carbon) and a final pH of 7.12. Biosorption systems using waste biomass (such as yeast from the manufacture of pharmaceuticals, beer, and wine) offer the promise of economical bioremediation of effluents from both acid-leach and alkaline-leach U mills.

The ARRICT plan calls for disposal of the biomass after one cycle. Other proposed uses of mycelial biomass for removal of metals and radionuclides from aqueous effluents (e.g., U removal by *Penicillium*

biomass; Galun *et al.*, 1983) consider stripping the trapped constituent and regeneration of the biosorbent. Galun *et al.* (1983) actually found U uptake by *Penicillium* biomass to increase following U stripping with NaHCO_3 and reuse. Where reuse is a possible option, chemical or physical pretreatment of the biomass (e.g., with alcohol, boiling, or detergents; Gadd and White, 1989; Galun *et al.*, 1983) to optimize radionuclide uptake may be cost justified. Tailored biosorbents, which use recombinant DNA technology to enhance the radionuclide-binding properties of the outer cell membrane or cell wall (Barkay and Schaefer, 2001), will probably see increasing use in the coming years.

Salt extractions, similar to the ARRICT stage 1 above, received much attention in the 1960s–1970s (see Landa, 1980), but commercial-scale usage has not followed, presumably because of costs. An alternative, less costly use of the biomass proposed by the ARRICT team involves the mixing of the biomass directly with the UMT during disposal to bind ^{226}Ra and decrease subsequent leaching losses by infiltrating water. Such approaches may be of additional value in the retention of contaminants in the UMT other than ^{226}Ra ; for example, another *Streptomyces* (*Streptomyces longwoodensis*) has an exceptionally high U sorption capacity (440 mg U/g dry weight; Friis and Myers-Keith, 1986). Other biowastes such as brewers yeast (*Saccharomyces cerevisiae*) probably have utility in this regard; this material has excellent U sorbent qualities (Kuyucak and Volesky, 1988). The stability and diagenesis of biosorbent materials incorporated in UMT have not been investigated, and hence the effectiveness of such measures cannot be predicted over time. Also organic amendments of UMT can have consequences in terms of the creation of reducing conditions (see section II). The most likely future applications of biosorbents probably remain in terms of engineered, flow-through systems aimed at treating mill pond effluents and acid mine drainage from U mining, rather than direct usage with tailings solids.

The processes by which biosorption occurs are varied and can include binding of radionuclides to functional groups in the cell wall, binding of radionuclides to inorganic precipitates at the cell wall, and the incorporation of radionuclides inside of cells. Haas *et al.* (1998) have studied the bioaccumulation of U by the cyanobacterial lichen *Peltigera membranacea*; both intracellular U uptake localized mainly in organelles known as concentric bodies and the precipitation of acicular U and P-bearing nanocrystals on cell walls and within extracellular, gelatinous, polysaccharide exudates were observed. The ability of U to cross the cell membrane has been demonstrated in multiple studies with yeast, lichens, and bacteria (Haas *et al.*, 1998; Kuyucak

and Volesky, 1988; Malekzadeh *et al.*, 2002; Strandberg and Shumate, 1982). Initial fixation at cell surfaces is thought to initially occur by surface complexation reactions, with crystal growth following at these loci of nucleation. These precipitates may sometimes serve as sorbents for other metals in a process termed “microbially enhanced chemisorption” (Lloyd and Macaskie, 2000). Basnakova *et al.* (1998) have demonstrated the retention of Ni^{2+} by HUO_2PO_4 crystals previously deposited enzymatically on *Citrobacter* sp. Bioremediation systems based on this specific process would seem to be ideally suited to the treatment of effluents from the milling of the Ni-rich U ores of northern Saskatchewan.

Although most attention with respect to UMT has focused on U and Ra, Th remains an radioelement of interest with respect to the long-lived ^{230}Th ($T_{1/2} = 80,000$ years; parent of ^{226}Ra) in the ^{238}U decay series, and with respect to U–Th-bearing ores (e.g., the ores mined at Elliot Lake, Ontario). Much of the Th is solubilized in the acid-leaching circuit of the mill and during pyrite oxidation in tailings piles. Thus, biosorption of Th may represent a control technology of interest when dealing with UMT pond effluent and acid mine drainage from Th-bearing ores and tailings. Gadd and White (1989) examined the sorption of Th from simulated acid process streams by yeast and fungal biomass and noted the importance of Th chemical speciation in HNO_3 as a function of pH. Nitric acid solutions are good models of nuclear fuel reprocessing waste streams. In acid-milling UMT raffinates (pH ~ 2), the predominant Th species would be expected to be the uncharged Th sulfate complex $\text{Th}(\text{SO}_4)_2^0$ rather than Th^{4+} , which dominates in pure water (Langmuir and Herman, 1980). Thus, comparisons to the literature, even at comparable pH values, must be done with caution. The same caution holds true for U, where UO_2SO_4^0 would be expected to be the dominant uranyl species at low pH in raffinate and raffinate-affected environments, as opposed to UO_2^{2+} in pure water (Langmuir, 1978).

VIII. Conclusions

The interactions of microorganisms with UMT have been shown to be diverse and with significant consequences for radionuclide mobility and bioremediation. These radionuclides are associated with the U-decay series. Recent proposals (Kirk, 2004) have called for the use of UMT disposal facilities in the United States for the disposal of other forms of low-level radioactive wastes; such usage has apparently occurred in Germany, where the fission product technetium-99 has been

detected at a UMT disposal site (Koss, 1992). Mixed-waste operations such as these may require us to broaden our view of these microbial processes to consideration of other radionuclides.

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