

A REVIEW OF POTENTIAL BIOTECHNOLOGIES FOR REMEDIATION OF URANIUM MILL TAILINGS

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ABSTRACT

One environmental concern from the mining and milling of uranium is the byproduct called Uranium Mill Tailings (UMT), which are the residues remaining after the extraction of uranium from ore in the earth's crust. The principle concerns with uranium mill tailings are the release of radon and the potential contamination of surface and ground water from the closed or abandoned UMT stacks. These environmental impacts have led to the Uranium Mill Tailings Radiation Control Act (UMTRCA) and to investigations of remediation technologies for the UMT stacks. Remediation technologies range from physical remediation by capping the stacks with clay to potential application of biotechnologies. Biotransformation of trace metals in the environment has been demonstrated by processes such as mobilization by oxidation and reduction reactions, biosorption, and hyperaccumulation. Potential remediation strategies applicable to UMT include: microbial reduction of radium sulfate (insoluble) to radium sulfide (soluble), leaching and subsequent microbial oxidation or biosorption as a means to concentrate the radioactive material. Phytoremediation utilizes plants for *in-situ* hyperaccumulation of radionuclides, followed by harvesting of biomass as a means of limiting radon emission. Application of biotechnologies appears to be an alternative to physical and/or chemical remediation techniques, although has not yet been demonstrated on a full-scale basis.

INTRODUCTION

There is a total of approximately 2.2 million tons of recoverable uranium found in Australia, Canada, South Africa, the United States, Western Europe, and some African countries (1). Some primary environmental issues facing the front end of the nuclear fuel cycle are the treatment and disposal of uranium mill tailings (UMT). UMT are the mining and milling residues remaining after the extraction of uranium from ore in the crust of the earth. In the U.S., uranium mill tailings are regulated separately as a radioactive waste due to the large volumes of waste (currently over 140 Mm³ in the U.S.) and the elevated naturally occurring radioactive material that is present.

Radon-222, a radioactive decay product of uranium-238, is a major concern with UMT. Radon-222 is a radioactive noble gas that decays to several short-lived alpha-particle emitters, which can result in a dose to the lungs. In addition to the radon emissions, the UMT can be dispersed to the environment through erosion by wind and water. The Uranium Mill Tailings Radiation Control Act (UMTRCA) designates the U.S. Department of Energy (DOE) responsible for the remedial actions at inactive tailings stacks. The environmental impacts and the UMTRCA have led to investigations of remediation technologies for the UMT stacks ranging from physical remediation by capping the stacks with clay to application of biotechnologies.

Some of the potential biotechnologies include microbial oxidation and reduction as well as biosorption and phytoremediation. Studies involving the mobilization of insoluble radium compounds have been conducted under anaerobic conditions where the oxidation state of the radium compound is reduced. Investigations have also been conducted on the use of iron, manganese, and sulfur oxidizing bacteria as a means to solubilize uranium or precipitate thorium and radium. Biosorption experiments include extracellular uptake of radium from interaction with the cell wall or outer surface of the microorganism. Several biosorption studies have been conducted on the radionuclides, thorium and radium, with various microbial species. Finally, phytoremediation, which involves hyperaccumulation by plants, has been evaluated with several radionuclides. Phytoremediation experiments, both in situ and laboratory, have been conducted on uranium mill tailings. The general overall result of these biotechnologies was the decreased concentrations of radium-226, thorium-232 in UMT, or UMT effluents.

URANIUM MINING & MILLING PROCESS

Two of the initial processes of the nuclear fuel cycle are the mining and milling of uranium. Uranium is ubiquitous in the crust of the earth with the average abundance of 2.7 ppm. For economical mining, the uranium concentration in the ore is typically a little less than 1% by weight. The United States, primarily Colorado, Utah and Wyoming, mined approximately 2,170 metric tons (6%) of the world production of 35,692 metric tons of uranium in 1997 (2). The four primary processes used for uranium mining include underground, open-pit, *in-situ* leach, and by-product mining. Underground mining, which contributed to 5.5% of the U.S. and 39% of the world uranium production in 1997, is a process in which an ore deposit is mined that contains a hard stratum above it. Open-pit mining is a method utilized for shallow deposits. The world uranium production for open-pit mining in 1997 was 39%, and less than 0.5% in the U.S. The most used mining process in the U.S. for 1997 was *in-situ* leach mining, which consisted of 71.5% of the uranium production. This process consists of an ore deposit existing below the water table of a confined aquifer and is mined by the injection of a mildly alkaline solution into wells. The world uranium production by *in-situ* leach mining was 13% for 1997. The advantage of the *in-situ* leaching is the UMT by-product is avoided. By-product mining, mining of uranium along with other minerals such as gold, phosphate, and copper, resulted in 23% of the U.S. production in 1997, while the world uranium production by this method was 9% for the same year.

Milling is the process of extracting the uranium from the mined ore and is usually done very near the mining site. Figure 1 is a schematic of the first steps in the milling process. Milling begins with crushing and grinding the ore to a powder. Next the uranium is leached from the powder with sulfuric acid or alkaline carbonate solutions. The solid byproduct is called UMT. Uranium is a minor component of uranium mill tailings because the leaching phase removes on the order of 95% of the uranium from the ore. Other constituents of the UMT are thorium-230, thorium-232, radium-226, and radium-228 (3). Sulfuric acid is preferred over hydrochloric and nitric acid for the acid extraction process because it is cheaper and less corrosive in dilute solution than the other two acids. Alkaline leaching is preferred over acid leaching if carbonate minerals are present as a major component due their high alkalinity and large buffering capacity. The uranium in the dilute and impure leachate is then concentrated and purified by ion exchange or solvent extraction to produce a high-grade uranium product. In the ion-exchange process, a strong base is the mobile phase while a quaternary-ammonia type resin is the immobile phase

contained in a column. The two solvent extraction processes used are AMEX, which uses amine, or DAPEX, which uses alkyphosphoric acid. The final major stage of the milling process is chemical precipitation, which recovers the uranium from the aqueous solution to produce a product known as "yellow cake", which is 70-90 % U_3O_8 (1).

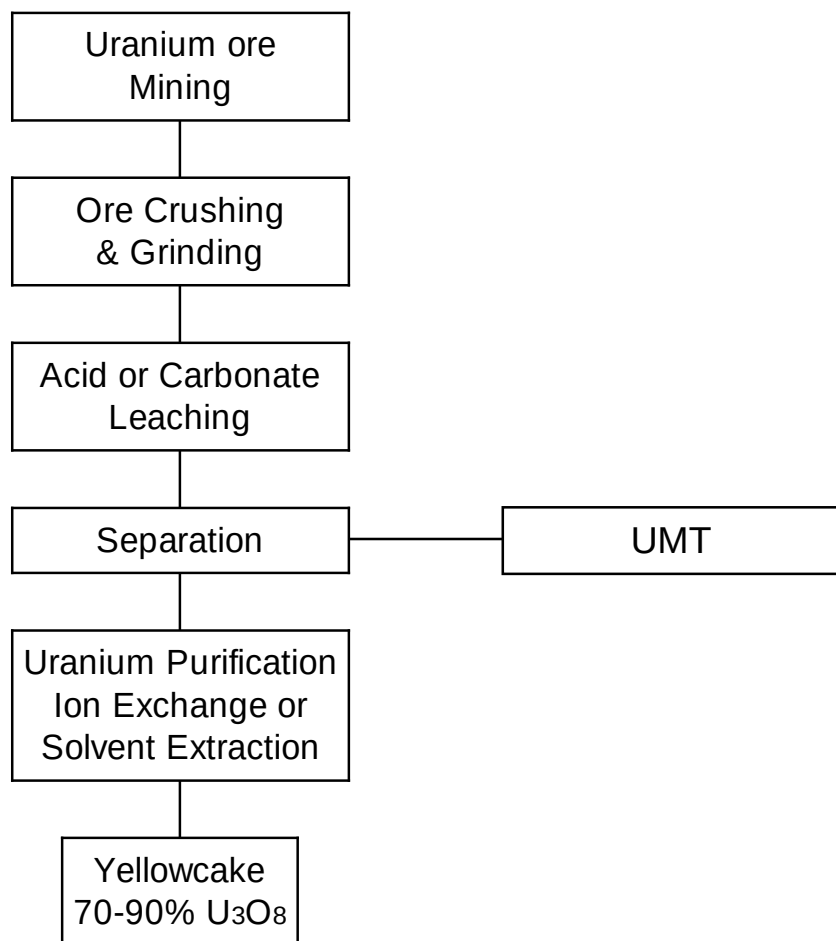


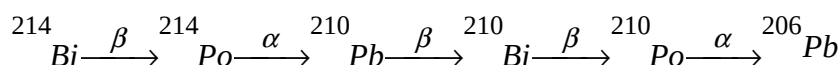
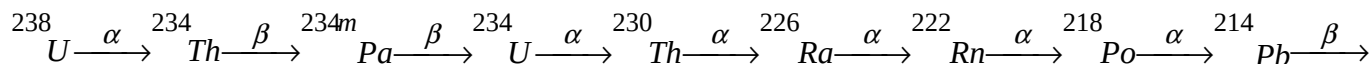
Fig. 1: Schematic of a typical milling process

RADIUM & THORIUM CHEMISTRY

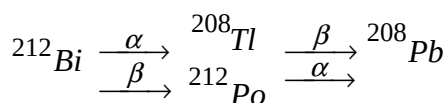
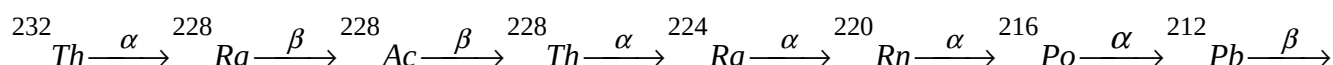
Uranium-235, 238 and thorium-232 are long-lived naturally occurring radioactive elements that are ubiquitous in the earth's crust. Radionuclides of radium and thorium (other than 232) are decay products of these three decay series. The uranium decay series (Fig. 2a, parent radionuclide uranium-238) and thorium decay series (Fig. 2b, parent radionuclide thorium-232) both contribute to human exposure. Radon-222, decay daughter of uranium-238 dominates the typical background dose to humans. Because of the long half-lives of radium-226 ($t_{1/2} = 1599$ yr.) and thorium-230 ($t_{1/2} = 75400$ yr.), removing the radium from the UMT will solve the radon emission problem for hundreds of years, but by removing the radium and thorium will curtail radon-222 emissions for thousands of years.

Radium, the precursor of radon, is an alkaline earth element that is ubiquitous in the crust of the earth. It has only one oxidation state (+2) in solution, and the divalent ion is not easily

complexed due to its highly basic characteristic. Radium forms water-soluble bicarbonate and chloride complexes. Radium sulfate is the most insoluble of the alkaline earth sulfates and the most insoluble radium compound known, but soluble in concentrated sulfuric acid. Radium sulfate can be chemically or biochemically reduced to radium sulfide, which is soluble in water and dilute acids (4).



(a)



(b)

Fig. 2: a) Uranium decay series and b) Thorium decay series

Thorium in the crust of the earth typically exists as an oxide or phosphate mineral in an oxidation state of +4, which can be dissolved with difficulty in concentrated acid. Nitrate, sulfate, and chloride are some water-soluble salts of thorium. Thorium hydroxide, a highly insoluble compound, is capable of dissolving in aqueous solutions containing complexants citrate or carbonate (5).

BIOTECHNOLOGIES

Bioremediation is a process that involves biodegradation and/or biotransformation of contaminants, with the intent of producing non-hazardous products. Extensive experience has been gained with bioremediation of numerous organic contaminants, including, for example, fuel hydrocarbons released from leaking storage tanks. With organic compounds, the most desirable end point of microbial activity is mineralization, yielding carbon dioxide, water, and new cell matter. With inorganic compounds, degradation is not an option. However, microorganisms do alter the chemistry and subsequent mobility of metals by a variety of mechanisms, including oxidation, reduction, release of chelating agents, and biosorption (6). A description of these phenomena in relation to radionuclides in UMT is given below.

Oxidation

Oxidation is the loss of one or more electrons from an atom or molecule. The ability of chemolithotrophic bacteria to oxidize inorganic compounds such as sulfides, manganese (+2), uranium (+4) and iron (+2) is well established. Oxidation of radionuclides in UMT, other than uranium, is less well understood.

Mobilization of radionuclides due to microbial oxidation reactions may be categorized as either direct or indirect. With direct oxidation, microorganisms use the element as an electron donor source, oxygen typically serves as the electron acceptor, and the carbon needed for biosynthesis is obtained from carbon dioxide. For example, a variety of thiobacilli can grow by aerobically oxidizing ferrous iron to ferric iron. Relatively little is known about the ability of chemolithotrophs to directly oxidize radium and thorium.

Indirect mobilization refers to processes involving oxidation of inorganic compounds other than the radionuclides. This often entails oxidation of sulfides to sulfuric along with oxidation of soluble ferrous iron to ferric iron. The acidic conditions allow the ferric iron to stay in solution, and the ferric iron can react with other metals to form a soluble complex. Such interactions are utilized in commercial leaching of low-grade copper ores. As shown below, however, limited success has been reported so far with indirect oxidative mobilization of radium and thorium.

Silver and Ritcey (7) tested the effect of *Thiobacillus ferrooxidans*, an iron-oxidizing aerobic chemolithotroph, on the mobility of radium-226, uranium and thorium in UMT from Rio Algom Quirke Lake Mine at Elliot Lake, Ontario, Canada. Three lysimeter boxes were filled with UMT and subjected to variations in sunlight and darkness, wind and precipitation similar to field conditions. Two of the lysimeters were inoculated with the thiobacilli, with one receiving a dose of organic solvents (kerosene, alamine, and isodecanol) used in uranium extraction, while the other did not. Hypochlorite was added to the third lysimeter to inhibit microbial activity (although it may have inadvertently acted as an oxidant). Iron sulfides in the two inoculated lysimeters were readily oxidized, leading to a gradual decrease in the pH of the leachate. However, no thorium and relatively little radium-226 was leached (over a simulated period of 20 years) from any of the lysimeters, regardless of the microbial activity in the two inoculated ones. In contrast, Silver and Ritcey (7) achieved more approximately 50% removal of radium-226 when the UMT were flushed with 2 M KCl, preceded and followed by washing with water. Similar results were obtained in another study by Silver and Ritcey (8) where lysimeters were set-up to test the effect of vegetation and inhibition of iron-oxidizing bacteria (using thymol) on the solubilization of radium-226. As above, the activity of iron-oxidizers did decrease the pH of the leachate, but had no impact on radium-226 removal. Neither did the vegetative cover. Another lysimeter was covered with a layer of compost as well as vegetation. Oxygen consumption at the surface due to microbial activity in the compost did prevent aerobic iron oxidation within the tailings almost as well as in thymol-treated lysimeter. This helped to prevent acid generation, but had no impact on the amount of radium removed.

Although no success has been reported so far with oxidative mobilization of radium and thorium, immobilization linked to oxidation of inorganic compounds is another process of potential interest. An example of indirect immobilization has been demonstrated by Mathur and Dwivedy (9) with manganese and radium-226. Both compounds were present in effluent from a UMT. When samples were inoculated with an *Arthrobacter* sp., the organism aerobically oxidized the manganese, resulting in precipitation of the manganese and coprecipitation of the radium-226 (9). Manganese (2+) and radium precipitation was 100% and 95%, respectively after 7 days of incubation. In contrast, essentially no removal was reported in sterile control samples.

Reduction

Reduction is the addition of electrons to an element or compound in a chemical reaction. Microorganisms can chemically reduce substrates to obtain energy to perform metabolic processes. Specifically, anaerobic sulfate reducing bacteria (SRB) utilize the sulfate ion as an oxidizing agent for the dissimilation of organic matter, resulting in the generation of H_2S and CO_2 (10). Similarly, anaerobic iron reducing bacteria have been used in controlled laboratory experiments to solubilize radium-226.

SRB have been shown to solubilize radium sulfate (insoluble in water) to radium sulfide (soluble in water). For sulfuric acid leached UMT, the radium present in the tailings will be primarily an insoluble sulfate precipitate. Experiments were performed with a pure culture of *Desulfovibrio desulfuricans* and a mixed culture of SRB that contained *Bacillus* and other unidentified bacteria, which was aseptically enriched from a South Dakota UMT site (10). The SRB in liquid growth medium were added to UMT in a glass flask. The flasks were subsequently incubated, sampled and analyzed for the number of microorganisms, culture contamination, and pH to determine whether the radium leaching was due to biosorption to the SRB cell walls (indirect effect), or an active biological process occurring only in the presence of live sulfate reducing bacteria cells (direct effect). These experiments concluded that the amount of radium-226 leached in the presence of both pure and mixed cultures of SRB is a factor of approximately 100 times greater than from the sterile system. Although SRB are capable of solubilizing radium in the presence of UMT, the specific mechanism (direct/indirect) for solubilization was not determined.

Under anaerobic conditions, microbial reduction of iron has resulted in an elevated release of radium-226 from UMT. Iron is present in uranium mill tailings as a result of the iron bearing minerals in the uranium ore, and the iron introduced from equipment in the grinding phase (11). *Alteromonas putrefaciens*, a facultative anaerobic bacterium, in a growth medium were added to UMT from a South Dakota site (11). When the Fe^{+2} accumulated and the O_2 was depleted, the bottles were flushed with a mixture of N_2 , CO_2 , and H_2 . The bottles were then incubated in the dark for a period of time and samples were taken from the medium and analyzed for dissolved sulfate, Fe^{+2} and radium-226. From this research, it was concluded that the radium-226 release from the inoculated tailings exceeded that of the sterile tailings by a factor of approximately 10 after 20 days, and the quantity of radium in solution in the inoculated samples tended to increase with time. From the data it was concluded that the leaching of radium-226 from the UMT is enhanced in the presence of *A. putrefaciens*, and that the enhancement is related with the reduction of Fe^{+3} in the tailings. The amount of radium released during a one-month incubation period of interest was approximately 3% of the total radium-226 in the UMT as compared to the SRB experiments where approximately 20 % was solubilized over the same time period.

Biosorption

Biosorption includes extracellular uptake of radionuclides from interaction with the cell wall or outer surface of the microorganism. These chemical and/or physical interactions may occur due to ion exchange, adsorption, complexation, crystallization and precipitation. Biosorption results vary with several factors such as composition of the cell wall, culture conditions, age of the culture, pH and temperature (12). Several biosorption studies have been conducted with various microbial species with thorium and radium.

Biosorption of radium can be an important removal mechanism for soluble radium from near neutral solutions. Tsezos et al. conducted experiments with radium chloride and different types of both dead and live biomass to determine the adsorption capabilities (13). Radium solutions with varying initial concentrations were contacted with different weights of dry *Penicillium chrysogenum*, *Rhizopus arrhizus*, *Aspergillus niger*, *Streptomyces niveus* and three activated sludge samples, and incubated for 24 hours. Radium uptake was occurred in each test. These experiments led to the conclusions that: 1) the adsorption of radium by biological adsorbents is feasible, and 2) the adsorption capacity is a function of radium solution equilibrium concentration and pH. *P. chrysogenum* was concluded to be an effective radium adsorbent (solid/aqueous phase partitioning coefficient, $K = 40 \text{ L/g}$, $\text{pH} = 7$). Of the biomass types investigated, significant uptake of radium did not occur until the pH was above 4, while the optimum uptake occurred between pH 7 and 10 (13). In a related set of experiments Tsezos et al. investigated the effect of the presence of complexing ions Ca^{2+} , Ba^{2+} , Mg^{2+} , Fe^{2+} , and Cu^{2+} (14). Radium-226 solutions were prepared in the same manner as the previous analysis, except for a variation in the concentration range of radium-226. The competing cations were introduced into the solution, with the solubility limits of the complexing ions not being exceeded. The complexing ions were tested both alone and in combinations at specific concentrations. This research determined that: 1) a pH range between 7-9 was optimum for radium biosorption under all conditions, 2) Ca^{2+} had a minor effect on radium sorption and 3) Ba^{2+} , Mg^{2+} and Fe^{2+} are significant competitors to radium for adsorption to *P. chrysogenum*. Tsezos et al. also determined for eluant type, eluant concentration, and solid to liquid ratio for five eluants ethylenediaminetetraacetic acid (EDTA), nitrilotriacetic acid (NTAA), triammonium citrate (TAC), HCl, and HNO_3 , optimum for removal of the radium from the biomass. The mineral acids and EDTA were concluded to be the most efficient radium eluants (15).

Radium adsorption to denitrifying bacteria has been investigated under laboratory conditions. Samples containing NO_3^- and radium-226 from Pond 3 at the Weldon Spring, MO site were investigated. After eight months the nitrate concentration was reduced from 21,500 mg/L to below 50 mg/L. The samples were centrifuged to remove the bacteria. It was then shown that the bacteria had adsorbed 98.6% of the radium originally present in the solution. One other radium adsorption experiment was completed with bacteria from the Y-12 Plant's Waste Coolant Degradation Reactor, which were aerobically grown in the presence of 270 pCi/L of radium-226. After one week's growth, the bacteria had adsorbed 89% of the radium (16).

The biosorption of thorium (+4) by *Citrobacter* sp. investigated in the presence of trivalent and hexavalent metal species (17). The cultures were harvested, and 5 g (wet) of cells were immobilized in a polyacrylamide gel (PAG). These immobilized cells were placed in columns and subjected to an upward flow of a citric buffer, metal nitrate containing lanthanum, uranium, and thorium, and ammonia acetate. The comparison of the residual metal in the column outflow and the metal content in the inflow solution determined the metal removal in the bioreactor. This experiment determined that the mixture of thorium and citrate buffer at particular molar ratios showed approximately 20% removal of thorium in the plug flow reactor. Another experiment was performed in a similar manner with an equimolar mixture of three metals, lanthanum, uranium, and thorium. This experiment concluded that the removal of thorium could be improved at very slow flow rates with manipulation of the composition of the solution by the addition of ammonium acetate and the reduction of the concentration of citrate (a tetravalent actinide chelating ligand) or the increasing of the substrate concentration. This test

demonstrated that the thorium removal was improved to 30%, while the amount of uranium and lanthanum removed was more than 62% and 85%, respectively (17).

REMEDIATION STRATEGIES

Bioremediation has been utilized to remove trace metals from copper, iron and uranium ores during mining operations. For a low-grade ore, it may be economically feasible to exploit direct and indirect microbial oxidation followed by leaching with a dilute sulfuric solution to remove the trace element from the ore. Application of microbial oxidation followed by leaching is common in copper and iron mining and has also been used commercially in uranium mining operations.

There are a number of possible means to use biotechnologies in the remediation of UMT (Fig. 3). A means of physical remediation would be to cap the UMT to reduce the radon emissions and reduce the effects of wind and water erosion. Microbial reduction of radium followed by leaching is one means utilize biotechnologies to remediate UMT. In the case where uranium is leached from the ore with sulfuric acid, the radium, as an insoluble radium sulfate, remains in the ore. By introduction or stimulation of sulfur reducing bacteria, the insoluble radium sulfate becomes soluble radium sulfide, which can be leached from the UMT. The resulting leachate could be treated via several biotechnologies: oxidation, reduction and biosorption. Leached radium sulfide could be oxidized back to radium sulfate where it would be allowed to precipitate out. Biosorption of uranium, thorium and radium compounds could result in a precipitate or radioactive materials that could be filtered out.

Phytoremediation is another means to reduce radon emissions and lower the potential impact of wind and water erosion. Plants that can grow on the UMT could concentrate radium within the plant cells. The bioaccumulation by plants is a consequence of the roots releasing exudates. The exudates solubilize or chelate elements and accumulate them into the plant cells. By harvesting the plants, the radium is removed from the UMT to a depth of up to 3-m (depending on the plant species). Assuming that the radioactivity has not accumulated in the roots, the plants can be harvested leaving the top of the UMT as a cap to the radium bearing UMT below. In addition, leaving a plant cover could lower the impact of wind and water erosion.

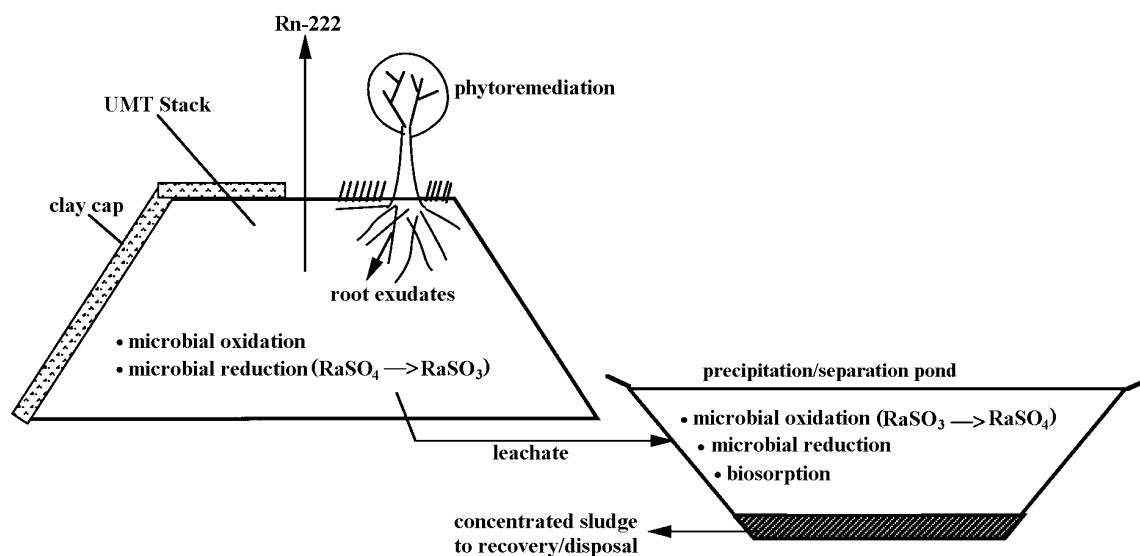


Figure 3. Schematic diagram of physical remediation and bioremediation.

Leaching

Leaching is a mechanism to separate the solubilized radioactive material from the bulk of the UMT. Biologically assisted leaching has been demonstrated for removing uranium from UMT as well as radionuclides from soil. The first step to bioleaching consists of biologically catalyzed metals being solubilized by, for example sulfur oxidizing bacteria. The metals, which contain soluble sulfate salts, are solubilized due to the acidity of the soil. The metal sulfates are then mobilized by a leaching process. The second step involves the collected leachate being further processed by the reducing bacteria, which results in precipitation of insoluble compounds. In an experiment reported by Gonzalez et al. (18), soil samples were air dried, homogenized, and analyzed for radioactivity, soil pH, particle size, microbial population size and composition, and water holding capacity. Thorium along with americium-241, and cobalt-60 were the primary radionuclides in this analysis being treated with sulfur oxidizing bacteria. In this procedure, the pH of the soil was reduced to less than 2, at which time the radionuclides are solubilized. These radioisotopes were then flushed with a known quantity of leaching medium several times. The initial two flushes were accomplished by saturating the soil and allowing free water to drain from the soil. The final flush was collected by applying a vacuum to remove free water. The results for this analysis concluded that there was approximately a 90% recovery in the leachate for americium-241, and a 5% recovery in the leachate for cobalt-60.

Phytoremediation

One of the newest bioremediation technologies is phytoremediation, in which vegetation is used for the in situ treatment of contaminated soils and sediments. Three mechanisms which plants can use to remediate organic and metal pollutants are: 1) direct uptake of the contaminants and accumulation of these substances in the plant's tissue, 2) releases of root exudates (including organic acids and enzymes) which stimulate microbial activity and biochemical transformations, and 3) the enhancement of mineralization in the root-soil interface (19).

In-situ and laboratory phytoremediation experiments have been conducted on mine tailings that are elevated in radium-226 to: 1) investigate the radium uptake of vegetation that has naturally grown on the UMT and 2) quantify uptake of radium-226 from uranium mill tailings by a grass, *Sporobolus airoides*, and a shrub, *Atriplex canescens* in a laboratory greenhouse (20). The uptake of radium-226 by naturally grown plant species showed a correlation between the radium-226 activity in the plants and the activity in the tailings/soil. These greenhouse experiments showed significant radium uptake by both the grass and shrub species grown on soil covered tailings. With minimal surface contamination and fertilization, the plants grown on soil covered tailings had significantly higher (5 to 300 times more) radium content than in control plants. Fertilization was shown to have an inverse effect on radium uptake by the plants since the fertilizer reduced the mobility of radium. The shrub species absorbed more radium than the grass due to better root penetration in the tailings (20).

In-situ phytoremediation experiments have been conducted on uranium mill tailings to investigate if: 1) the growth of native vegetation on tailings without using irrigation was possible, and 2) vegetation growing in tailings or soil covered tailings accumulated radioisotopes above background (21). The first phase of this experiment was conducted at three inactive tailings sites in New Mexico by placing varying cover materials on test plots. The sites were covered with either local clay soils or stony clay loam and sandstone rocks with boulders, or clay and sandstone rocks with boulders. These plots were then seeded with a mixture of native grasses

including *Agropyron smithii*, *Andropogon scoparius*, *Bouteloua curtipendula*, *Bouteloua gracilis*, *Eragrostis curvula*, *Sporobolus airoides*, *Artemisia tridentata*, *Atriplex canescens*, and *Melilotus officinalis*, and periodically observed for germination and survival. It was concluded that the stony clay loam and sandstone rocks with boulders provided the best substrate for the survival of grasses and weeds, while the local clay soils did not favor growth. The two native species, *M. officinalis* and *A. canescens*, had the highest mean germination rates in the stony clay loam and sandstone rocks with boulders cover. The plot with clay and sandstone rocks with boulders thrived on the sandstone rock treatment, and this treatment enhanced the establishment of weeds, grasses, and an invading perennial grass, *Sitanion hystrix*. The two native species that had the highest mean germination response of seeds for the clay and sandstone rocks with boulders treatment were *E. curvula* and *M. officinalis* (21). Dreesen et al. also conducted experiments to establish if elements were accumulating in the plants at 14 inactive sites in New Mexico, Colorado, and Utah. This research was conducted by the collection of aboveground samples of the predominant plant species such as *S. hystrix*, *S. airoides*, *Agropyron elongatum*, *A. canescens*, *Kochia scoparia*, and *Salsola kali*. At the various uranium mill tailings sites, it was noted that plants were growing in bare tailings or on shallow soil covered tailings. Tailings and soil samples were also collected at each site, and used to determine the extractable trace elements present. Radium-226 was determined on a total basis by gamma spectrometry in soils and tailings. It was concluded from this experiment that radium-226 was being taken up predominantly by the plant species, *K. scoparia* and *S. hystrix*. This investigation demonstrated the potential for contaminant uptake by plants growing in bare uranium mill tailings or tailings with shallow soil caps (21).

Phytoremediation in laboratory experiments has been investigated using UMT from the Rio Algom Quirke mill in Ontario, Canada (8). The initial steps of this experiment were previously mentioned in the oxidation section of this paper. After the application of water (equivalent to one year of rainfall), a seed mixture and fertilizer were applied to the tailings. In one case, grass was grown directly on the UMT and in another case the vegetation was grown on a 30-cm compost cap in addition to the UMT. Tap water and horticultural lamps were then applied to the lysimeters. Leachate was collected from each of the samples with recordings of the volume, pH, total iron and calcium, sulfur in the form of sulfate, and radium-226. It was concluded from this experiment that the establishment of vegetation directly on the tailings surface decreased but did not eliminate sulfuric acid generation. The concentration of the radium-226 was slightly lower for the fertilized and vegetated sample than for the bare or composted and vegetated samples. It was also determined from these tests that the solubilization of radium-226 was not influenced by the establishment of vegetation on the surface of the tailings, but that the radium-226 in the tailings was dissolved primarily as a result of its solubility (8).

CONCLUSION

With the daily increasing volume of the UMT stacks in the U.S. and the world, mitigation of the impact of these mining activities on the natural environment needs to be implemented by a formulation of treatment and disposal methods. Recent studies on uranium mill tailings, and their primary constituents, radium and thorium, have been conducted *in-situ*, on-site and in laboratories. All the bioremediation technologies, oxidation and reduction both coupled with

leaching, biosorption, and phytoremediation, presented in this paper are viable for varying conditions of the UMT and UMT effluents.

Biotechnologies have been applied to the iron, copper and uranium mining industries. Application of these biotechnologies to remediation UMT have been investigated, but not on a large scale. Reduction of radium sulfate in UMT by SRB followed by leaching and subsequent concentration appears to be a viable option for large-scale UMT remediation. Phytoremediation also holds promise as a means for large-scale UMT remediation. Given plants that do not bioaccumulate radioactivity in the roots, the plants can be harvested and the radioactive material properly disposed of. Bioremediation of UMT to remove radium-226 will solve the problem of radon emissions from UMT for hundreds of years, while removal of thorium from the UMT will curtail the radon emissions for thousands of years. Thus, further research into application of biotechnologies to thorium is needed to determine the feasibility of bioremediation of thorium.

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