Microbial Transformations of Actinides and Fission Products in Radioactive Wastes

A.J. Francis

Environmental Sciences Department, Brookhaven National Laboratory, Upton, NY 11973, USA; Division of Advanced Nuclear Engineering, Pohang University Science and Technology, Pohang, Korea

francis1@bnl.gov

Microorganisms are ubiquitous in the subsurface environments and play major role in the biogeochemical cycling of various elements in nature. Of particular concern is the impact of on radioactive wastes to be disposed of in deep geological formations. Microorganisms have been detected in TRU wastes, Pu-contaminated soils, low-level radioactive wastes, backfill materials, natural analogue sites, and waste-repository sites slated for high-level wastes. Under appropriate conditions, various microbial processes can be active, and will potentially affect the mobility of radionuclides and the integrity of the engineered barrier system of the repository.

Depending on the availability of nutrients, electron donors and acceptors, and the environmental factors, various microbial processes with diverse metabolic activity can be stimulated. The environmental factors that can affect microbial growth and activity include moisture, temperature, pH, Eh, availability of organic and inorganic nutrients, and radiation. The microbial activity in a specific repository is influenced by the ambient environment of the repository, and the materials to be emplaced. For example, a repository in unsaturated igneous rock formations such as volcanic tuff rocks at Yucca Mountain is generally expected to be oxidizing; a repository in a hydrologically saturated zone, especially in sedimentary rocks, could be reducing. Sedimentary rocks contain a certain amount of organic matter, which may stimulate microbial activities and, thus maintain the repository and its surrounding areas at reducing conditions.

Although the impacts of microbial activity on high-level nuclear waste and the long–term performance of the repository have not fully investigated, little microbial activity is expected in the near–field because of the radiation, lack of nutrients and the harsh conditions. However in the far–field microbial effects could be significant.

Much of our understanding of the microbial effects on radionuclides stems from studies conducted with selected transuranic elements and fission products and limited studies with low-level radioactive wastes. Significant aerobic- and anaerobic-microbial activity is expected to occur in the waste because of the presence of electron donors and acceptors. The actinides initially may be present as soluble- or insoluble-forms but, after disposal, may be converted from one to the other by microorganisms. The direct enzymatic or indirect non-enzymatic actions of microbes could alter the speciation, solubility, and sorption properties of the actinides, thereby increasing or decreasing their concentrations in solution. Dissolution of radionuclides reflects changes in the Eh and pH of the local environment caused by the microorganisms, by their production of CO_2, or of extra cellular metabolic products, such as organic acids, and sequestering agents such as siderophores. Immobilization or precipitation of radionuclides is due to changes in the Eh of the environment, enzymatic reductive precipitation (reduction from a higher to lower oxidation state), biosorption, bioaccumulation, biotransformation of radionuclide-organic and -inorganic complexes, and bioprecipitation. Free-living bacteria suspended in the groundwater fall within the colloidal size range and may have a strong radionuclide sorbing capacity, giving them the potential to transport radionuclides in the subsurface. Microbial corrosion of the waste canisters can compromise waste integrity. The actinides exist in various oxidation states and may be present as oxide, coprecipitates, inorganic, and organic complexes. Microorganisms by direct enzymatic or indirect non-enzymatic actions could affect the chemical nature of the actinides by altering the speciation, solubility, and sorption properties.

A summary of the various microbial processes and the mechanisms of biotransformation of selected radionuclides are listed in Table 1. Among the actinides biotransformation of various uranium compounds have been extensively investigated. A variety of bacteria are known to catalyze the reduction of soluble U(VI) to insoluble U(IV) which precipitated from solution. We have limited information on other actinides.

Process	**Th**	**U**	**Np**	**Pu**	**Am**	**Tc**	**I**	**Se**	**Sr**	**Cs**
Oxidation[1]		++	ND	ND	NA	+	++	++++	NA	NA
Reduction[2]		++++	++	++	NA	++++	++	++++	NA	NA
Dissolution[3]	+	+++	?	+	?	+++	++		+	+
Precipitation		+++	+	++	?	+++	++		++	?
Biosorption	+	++++	+	++	+	?	++		++	++
Biomethylation		NA	NA	NA	NA	NA	+++	+++	NA	NA
Biocolloid[4]		++	+	++	+	?	?		?	?

Table 1. Summary of key microbial processes and transformations of actinides and other radionuclides.
NA- not applicable; ND- not determined; [1]Dissolution due to oxidation from lower to higher valence state; [2]Reductive precipitation due to enzymatic reduction from higher to lower valence state; [3]Dissolution due to oxidation from lower to higher valence state, changes in pH, production of organic acids and sequestering agents; [4]Radionuclides associated (extracellularand/or intracellular) with suspended bacteria can be transported as biocolloids.

A slight increase in microbial activity (respiration) can alter the oxidation state of Pu(VI) to Pu(IV) because of the very small differences in the reduction potential between Pu(VI), Pu(V), and Pu(IV). Under anaerobic conditions, reductive dissolution of Pu(IV) to Pu(III) by *Clostridium* sp was observed. The presence of Pu(III) was confirmed by XANES. Studies with Pu contaminated soils from Nevada Test Site (NTS) showed solubilization of Pu and other radionuclides due to enhanced microbial activity. The type of carbon source affected the rate and extent of Pu dissolution and Pu in the solution phase. Theses studies show that microbial transformations of actinides and fission products in nuclear waste disposed of in subterranean environment could be significant under appropriate conditions. However, additional studies are clearly needed to evaluate the potential impacts of microbial activity on long-term repository performance.

ACKNOWLEDGMENTS

This research was supported by the Office of Science, Office of Biological and Environmental Research, U.S. Department of Energy under contract No. DE-AC02-98CH10886 and in part by the World Class University (WCU) program through the National Research Foundation of Korea, funded by the Ministry of Education, Science and Technology (R31 – 30005).

REFERENCES

Francis, A.J. 1990. Microbial dissolution and stabilization of toxic metals and radionuclides in mixed wastes. Experientia 46:840-851.

Francis, A.J. 1998. Biotransformation of uranium and other actinides in radioactive wastes. J. Alloys and Compounds 271-273: 78-84.

Wang, Y. and A.J. Francis. 2005. Evaluation of microbial activity for long-term performance assessments of deep geologic nuclear waste repositories. J. Nucl. Radiochem. Sci., 6: 43-50.