Predicting actinide solubilities in various solutions up to concentrated brines: The Fracture-Matrix Transport (FMT) Code

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The Fracture-Matrix Transport (FMT) code developed at Sandia National Laboratories (Babb and Novak, 1997 and addenda) solves chemical equilibrium problems by employing Pitzer activity coefficient equations. The code can predict solubilities of actinides at 25 ^{o}C in high ionic strength brines. The code uses oxidation state analogies, i.e., Am(III) is used to predict solubilities of actinides in the +III oxidation state; Th(IV) is employed to predict solubilities of actinides in the +IV state, and Np(V) is utilized to predict solubilities of actinide in the +V state. This code has been qualified for predicting solubilities of actinides for the Waste Isolation Pilot Plant (WIPP).

In this presentation, solubilities of actinides predicted by FMT are compared with 538 experimental solubility data taken from the open literature. The solutions used in these solubility studies included simple solutions (NaCl, $NaHCO_3$, Na_2CO_3, $NaClO_4$, KCl, K_2CO_3, etc.), binary solutions (NaCl + $NaHCO_3$, NaCl + Na_2CO_3, KCl + K_2CO_3, etc.), ternary solutions (NaCl + Na_2CO_3 + KCl, $NaHCO_3$ + Na_2CO_3 + $NaClO_4$. etc.), and multi-component synthetic WIPP brines. The predicted values are in excellent agreement with experimental values.

Reference

Babb S.C., and Novak C.F., (1997), and addenda. User's Manual for FMT Version 2.3: A Computer Code Employing the Pitzer Activity Coefficient Formalism for Calculating Thermodynamic Equilibrium in Geochemical Systems to High Electrolyte Concentrations. Albuquerque, NM: Sandia National Laboratories. ERMS 243037.

Acknowledgements

This research is funded by WIPP programs administered by the U.S. Department of Energy. Sandia is a multiprogram laboratory operated by Sandia Corporation, a Lockheed Martin Company, for the United States Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.

Subsurface microbial biofilms and nuclear waste disposal – Geochemical friends or foes?

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Background

The Swedish nuclear fuel management company (SKB) has instigated final site investigations for a deep subsurface high level nuclear waste (HLW) repository in granitic rock. This facility will be situated 500 meters below ground level and will isolate the HLW from the environment using an engineered multi-barrier system with a supposed design life of 100 000 years. In hard rock repository environments, any radionuclide migration from the waste will be facilitated by groundwater flowing though fractures. The water in these fractures is geochemically reduced and the fracture surfaces are covered with anaerobic bacterial biofilms.

Simulated *in situ* conditions and biofilms grown 450 meters underground were used to investigate the adsorption capacity of bare rock surfaces versus surfaces covered in monolayer biofilms. The radionuclides used were Co, Pm, Np, Am, Th, U and Mo. Scintillation, NaI γ-detection, ICP-MS and autoradiography was used to investigate the adsorption characteristics of the biofilms.

Results and Discussion

The results suggest that ionic strength, pH, oxidation state and valence state are all extremely important factors in the efficiency of adsorption by rock and biofilms in this environment. Anaerobic biofilms in 1.5 % salt and near neutral pH tend to favour the adsorption of the 3+ valence state (Am, Pm) over other radionuclides such as Co, Th and U. When compared to the rock surface, the biofilms tend to have slightly less capacity than the rock itself except for 3+ valence metals where the sorption capacity is comparable or greater. Results from experiments with in situ biofilms grown on rock surfaces demonstrate that the bacteria isolate the rock from the water reducing the adsorption potential of the rock.

Conclusions

Subsurface biofilms can decrease the adsorption capacity of the host rock for some radionuclides. Biological retardation of radionuclides may not take place until the ground water is more oxidised and closer to the surface.