Sandia National Laboratories

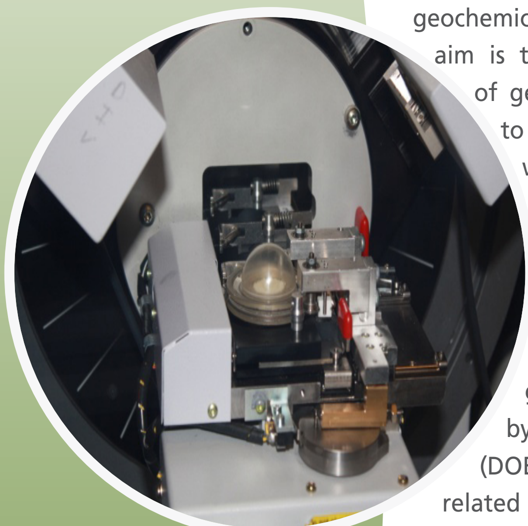
Energy, Climate, & Infrastructure Security

Geochemistry

Researchers in Sandia National Laboratories' Defense Waste Management Programs use geochemical investigations to study near-field conditions associated with the safe deep-geologic disposal of radioactive waste.

Applying Geochemistry to Solve Issues of National Importance

Geochemistry is the study of the interactions between rocks, minerals, natural waters and gases. The Defense Waste Management Programs has an active geochemical research program whose aim is to broaden our understanding of geochemical interactions related to the disposal of radioactive waste. In particular, the work is focused on supporting the safe disposal of radioactive waste in the Waste Isolation Pilot Plant (WIPP) located near Carlsbad, NM. The WIPP is a deep geologic repository developed by the U.S. Department of Energy (DOE) for the disposal of defense-related transuranic (TRU) radioactive waste within the bedded salts of the Permian Salado Formation, which consists of interbedded halite and anhydrite.



XRD analysis of corrosion products using anoxic sample holder

The nature of the environment within the WIPP following closure will, to a large extent, control the speciation of the radionuclides within the waste, and in turn, their mobility in the natural environment. Thus a critical aspect of the Defense Waste Management Programs' geochemistry program is to understand the chemical conditions arising from the interaction of the waste materials with the natural environment (including concentrated NaCl brines; minerals such as halite and anhydrite; and the engineered barrier material, MgO and its reaction products). The data gained from these studies is used to demonstrate compliance with containment requirements set by the Code of Federal regulations through a performance assessment calculation.

Brine Research

Salt formation brines such as those that may be present in the WIPP tend to have high concentrations of sodium, calcium, and chloride with some brines also having high magnesium concentrations. Lesser amounts of borate, sulfate and carbonate are also important components in WIPP brines. The use of geochemical models to describe accurately such high-ionic-strength brines requires that the models be able to evaluate non-ideal solution behavior using the Pitzer activity coefficient model for aqueous species. However, much of the thermodynamic data required for modeling brine-material interactions has not been available due to difficulties in conducting experiments in concentrated brines. In order to meet the needs of the WIPP program, the geochemistry group has invested significant time and resources into developing an extensive research program aimed at determining thermodynamic properties of solid and aqueous phases of interest in a salt repository system.

Carbon Sequestration Studies

WIPP contains significant quantities of cellulosic, plastic and rubber (CPR) materials. With time, microbial activity may consume some portion of the CPR materials, resulting in generation of significant quantities of carbon dioxide. The microbially-produced CO2 has the potential to significantly affect the mobility of actinides by acidifying any brine present in the repository, thus increasing actinide solubilities. An engineered barrier, magnesium oxide (MgO), is emplaced in the WIPP alongside the waste to buffer the concentration of CO2 and brine pH within ranges that favor lower actinide

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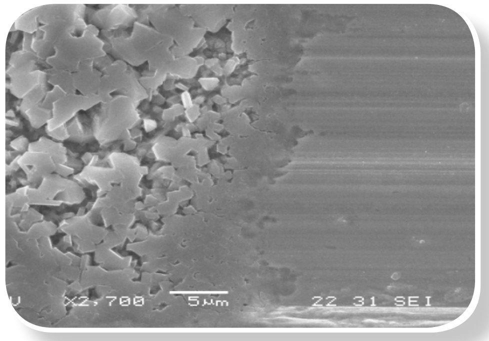
solubilities. Because it is important to understand the near-field geochemical conditions when the engineered barrier MgO interacts with Na–Mg–Cl-dominated brines the Defense Waste Management Programs, has performed CO_2 sequestration experiments in high ionic-strength solutions for over 13 years. These experiments are intended to show that CO_2 generated by decomposition of organic matter in the waste will not adversely affect repository performance. We have all of the experience, equipment, and ongoing experiments needed to address carbon sequestration not only in the WIPP near-field environment but in other environments as well.

Anoxic Corrosion Research

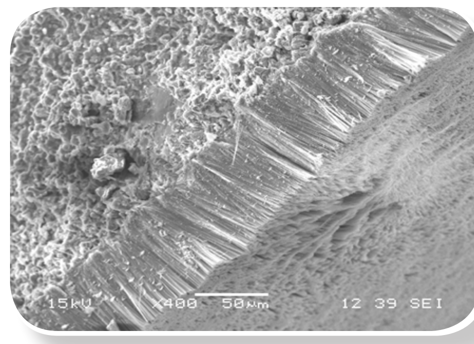
The nature of the environment within the WIPP following closure will, to a large extent, control the speciation of the radionuclides within the waste. More specifically, there are components contained within the waste that can impact the oxidative or reductive nature of the environment, such as metals undergoing active corrosion. If metals undergo active corrosion within the WIPP, the corrosion process will serve to maintain electrochemically reducing conditions. The predominant metals within the WIPP will be iron (Fe) in the form of low-carbon steel and lead (Pb). Anoxic corrosion of these metals will also result in the formation of H_2 gas, which impacts room closure and chemistry. The Defense Waste Management Programs has developed an extensive research program aimed at understanding the corrosion behavior of iron and lead in brines under anoxic conditions. This includes the design and operation of a one-of-kind large flow through reactor, the Mixed Flow Gas Control System (MFGCS), which allows maintenance of a controlled environment for the Fe/Pb corrosion and CO_2 sequestration experiments.



SEM image of anoxic corrosion front on steel immersed in NaCl brine

Laboratory Capabilities for Advancing Repository Science

The Defense Waste Management Programs' extensive research program supporting the WIPP mission is carried out in the geochemistry facility in Carlsbad, NM. This facility is well-equipped to address the many geochemical issues related to the current WIPP mission, as well as future research into other repository options. Multiple VAC anoxic gloveboxes allow work to be done on ferrous iron samples. Solid-phase reaction products can be analyzed by X-Ray diffractometry, carbon coulometry, and Scanning Electron Microscopy (couple with Energy Dispersive Spectroscopy and Electron Back-Scatter Diffraction). Cation and trace metal concentrations can be analyzed by Ultraviolet-Visible Spectroscopy, Inductively Coupled Plasma-Atomic Emission Spectroscopy (ICP-AES) and Inductively Coupled Plasma-Mass Spectrometry (ICP-MS) at the part per billion (ppb) levels. Analyses of anions and organic components are available via the Ion Chromatograph. Gas analysis is done using a Gas Chromatograph/ Mass Spectrometer (GC/MS) coupled with Thermal gravimetric, differential scanning calorimetry (TGA/DSC). Other equipment available includes: an Ultra Centrifuge for use in ultra-filtration in mineral colloid experiments; Accelerated Surface Area and Porosimetry analyzer (ASAP 2020) for determining surface area, pore volume and diameter; Malvern Particle Analyzer for particle sizing; and other standard laboratory instrumentation. In addition, the lab has recently acquired a high temperature/pressure reaction vessel system that will allow brine/material interaction experiment to be conducted to temperatures of 340°C and 345 bars.



SEM image shows a reaction front from an MgO hydration study

Publication

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