Solubility of Neodymium in Simulated WIPP (Waste Isolation Pilot Plant) Brines

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**ABSTRACT**
Neodymium is a redox-invariant analog for americium (III) and plutonium (III), which are key contaminants in the WIPP. The measurements of neodymium solubility in simulated WIPP brine is expected to verify current assumptions in WIPP Performance Assessment that were based on simplified systems.

In the present work the neodymium solubility was measured in GWB (Generic Weep Brine), ERDA-6 (Energy Research and Development Administration Well 6) Brine and 3 M NaCl as a function of carbonate concentration and pH using an over-saturation approach. Samples were equilibrated for three months and the neodymium concentrations were measured in unfilled and filtered aliquots using ICP-MS. Obtained results were compared with the literature data. Precipitation led to a three months and the neodymium concentrations saturated approach. Samples were equilibrated for Well 6) Brine and 3 M NaCl as a function of simplified systems.

**EXPERIMENTAL APPROACH**

**Key Experimental Parameters**
- pH between 6 and 12
- Carbonate concentrations up to 0.01 M
- Temperature of 25°C ± 3°C
- Speciation (complexation)
- Interactions with waste components (Fe, MgO, AI, Ni, Pb and Organics)

**Solubility Experiments**
Solubility is approached from both under-saturation and over-saturation under controlled environmental conditions and in the presence/absence of key waste components. In over-saturation experiments, neodymium as an actinide +3 analog is added sequentially to simulated WIPP brine until a steady-state concentration is achieved. In under-saturation experiments, the expected/predicted solid phases are prepared and contacted with simulated brine under controlled conditions until a steady state concentration is achieved. The results of both approaches, in light of analogous modeling studies and calculations, are combined to establish the likely solubility under the expected subsurface conditions.

**Analytical Techniques**
Total solution concentrations are determined by ICP-MS as a function of solution filtration (when applicable). Solids will be characterized by XRD, SEM and TEM. XANES/EXAFS analysis will be performed on selected samples to establish the near-neighbor structure of the environmentally relevant precipitates obtained by comparison to well-characterized references.

**CALCULATED An(III) SOLUBILITY**

**pH DEPENDENCE**
The experimentally available pH ranges in GWB and ERDA-6 brines are limited at higher pH by precipitation of magnesium hydroxide and at lower pH by the steep slope when small amounts of acid are added (green area in the graph above). Ne(II) concentrations were measured at three equilibration times as a function of pH. All plots are consistent with data reported in the literature for americium.

**EQUILIBRATION TIME**
The kinetics of equilibration in the over-saturation approach was studied. The change in Nd concentration as a function of time for GWB and ERDA-6 brines at various pH were measured. The observed trends in the Nd concentrations with time were similar for both brines, GWB and ERDA-6. The major decrease in Nd concentration occurred during the first 20 days of the solubility experiments.

**pH MEASUREMENTS IN BRINE SYSTEMS**
The extension of commonly used analytical techniques and approaches to brine systems is often problematic and not straightforward. Significant effort is required to confirm and develop experimental protocols that work for brine systems. For example the measurement of hydrogen ion concentration (pH), which is a critical parameter for actinide solubility in geochemical systems, is made difficult by the high ionic strength and buffer capacity of the brines used. In concentrated brines, variations in activity coefficients, the formation of species such as HSO₃⁻ and H₂SO₄ that can consume protons during electrode standardization procedures, and potentially large junction potentials all add to the difficulty in the pH measurement.

**EFFECT OF CARBONATE**
Neodymium solubility in GWB and ERDA-6 brine at a constant pH but with varied carbonate concentrations and various equilibration times were investigated. The neodymium concentrations in these samples were measured using ICP-MS after 39 and 67 days of equilibration.

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