Evaluation of bentonite engineered barrier performance under repository conditions: Diffusion of Np(V) through montmorillonite

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Outline

• Introduction
• Goals
• Experimental Setup and Procedure
• Results
• Future Work
• Questions?
Introduction

- Spent fuel pools and above ground storage
- Deep geological repository suggested
- Yucca Mountain
Introduction

• Long lived minor actinides of concern, in particular $^{237}\text{Np}$
  – Potential for mobility
  – Half-life of 2.14 million years
  – Environmental and human health concerns
Introduction

• Predominant oxidation state of $^{237}$Np is Np(V)
• Least likely for adsorption to occur

Increasing Complexation Affinity

<table>
<thead>
<tr>
<th>An$^{4+}$</th>
<th>AnO$_2^{2+}$</th>
<th>An$^{3+}$</th>
<th>AnO$_2^{+}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>4+</td>
<td>3.3+</td>
<td>3+</td>
<td>2.3+</td>
</tr>
</tbody>
</table>

\[\text{An}^{4+} > \text{AnO}_2^{2+} \approx \text{An}^{3+} > \text{AnO}_2^{+}\]
Introduction

• Heat and water corrode the steel causing ruptures
• Compacted layer of clay as form of secondary containment

V.R. Ouhadi et al. 2010
Introduction

- Properties of clay
  - Low porosity
  - Slow diffusive transport
  - High adsorption of cations
  - Plasticity/swelling

- Will this clay barrier be enough to contain $^{237}$Np, specifically at elevated temperatures?
Goals

1. Study the diffusion of $^3$H and Np(V) through montmorillonite clay
2. Four bulk densities: 1.15, 1.3, 1.45, 1.6 g/cm$^3$
3. Elevated temperatures: 25, 50, and 80ºC
Experimental Setup
• 1,000 ppb Np(V)
• 10,000 ppb $^3$H
• 100 mL DDI H$_2$O
• 100 mM NaCl
• pH 6

Van loon & Soler, 2004

• 20 mL DDI H$_2$O
• 100 mM NaCl
• pH 6
Sample Collection

• Two mL samples collected daily from low reservoirs

• Low reservoir solution exchanged ~5 samples
  – Ensures concentrations do not exceed ~1% of high reservoirs

• High reservoirs sampled weekly

• Analyzed on Liquid Scintillation Counting (LSC) and ICP-MS

• Sampling occurred until breakthrough was seen for cells one and two
LSC Analysis of $^3$H Data at 25°C

1.3 g/cm³ Dry Bulk Density

$D_e$ vs Dry Bulk Density
Np(V) Analysis - ICPMS at 25°C

Cumulative Np(V) Activity

<table>
<thead>
<tr>
<th>Bulk Density (g/cm³)</th>
<th>Dₑ (m²s⁻¹)</th>
<th>α</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.15</td>
<td>1.47x10⁻¹²</td>
<td>3.81x10⁻¹</td>
</tr>
<tr>
<td>1.30</td>
<td>5.07x10⁻¹³</td>
<td>1.26x10⁻¹</td>
</tr>
</tbody>
</table>
Extrusion of Clay Plug
Future Work
Future Work

- Perform autoradiography and microscopy of extruded clay samples
- Diffuse radioisotopes through cells at 50°C 80°C
- Analyze samples for % sorbed
- Image diffusion process with carbon fiber sample chamber and PET scanner
- Develop a better understanding of the diffusion process
Acknowledgments

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References


Questions?