Praseodymium [Pr(III)] as an Analog to Trivalent Plutonium [Pu(III)] and Neodymium [Nd(III)] as analog to Trivalent Americium [Am(III)] and Curium [Cm(III)]: Insights from their Oxalates

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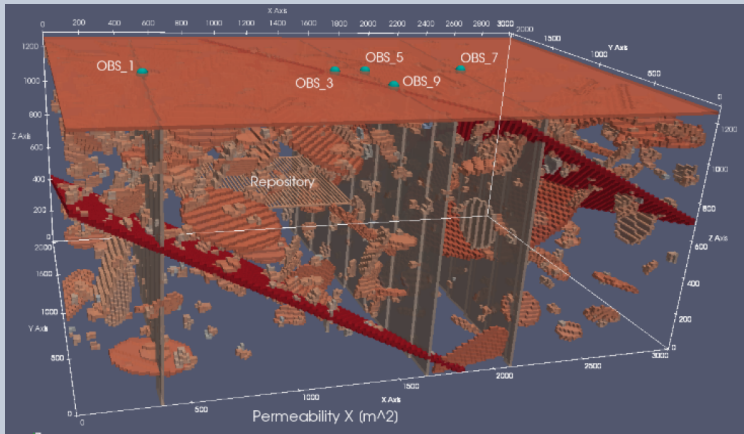


OUTLINE OF PRESENTATION

- Introduction
- Objective of This Work
- Experimental Set-up
- Experimental Results
- Modeling Results
- Applications

INTRODUCTION

- **Actinides and oxalates are both present in nuclear waste streams in geological repositories:**
  - For instance, in the French nuclear waste management programs, the liquid waste containing both actinides and oxalate from the spent fuel reprocessing is conditioned for the disposal in clay formations.
- **As actinide oxalates have very low solubilities, they could become solubility-controlling phases and prevent the mobility of actinides**
- **Rare earth elements (REE) are ideal for this investigation because they are good analogs to actinides and do not have radiotoxicity.**

OBJECTIVE OF THIS STUDY

- **The solubility of Pr- and Nd-oxalates is determined as a function on ionic strength.**
- **Based on the experimental data, the Pitzer model is developed to describe the solubilities of Pr- and Nd-oxalates, which can be applied to actinide oxalates.**

EXPERIMENTAL

- **Synthesis of Pr- and Nd-oxalates**

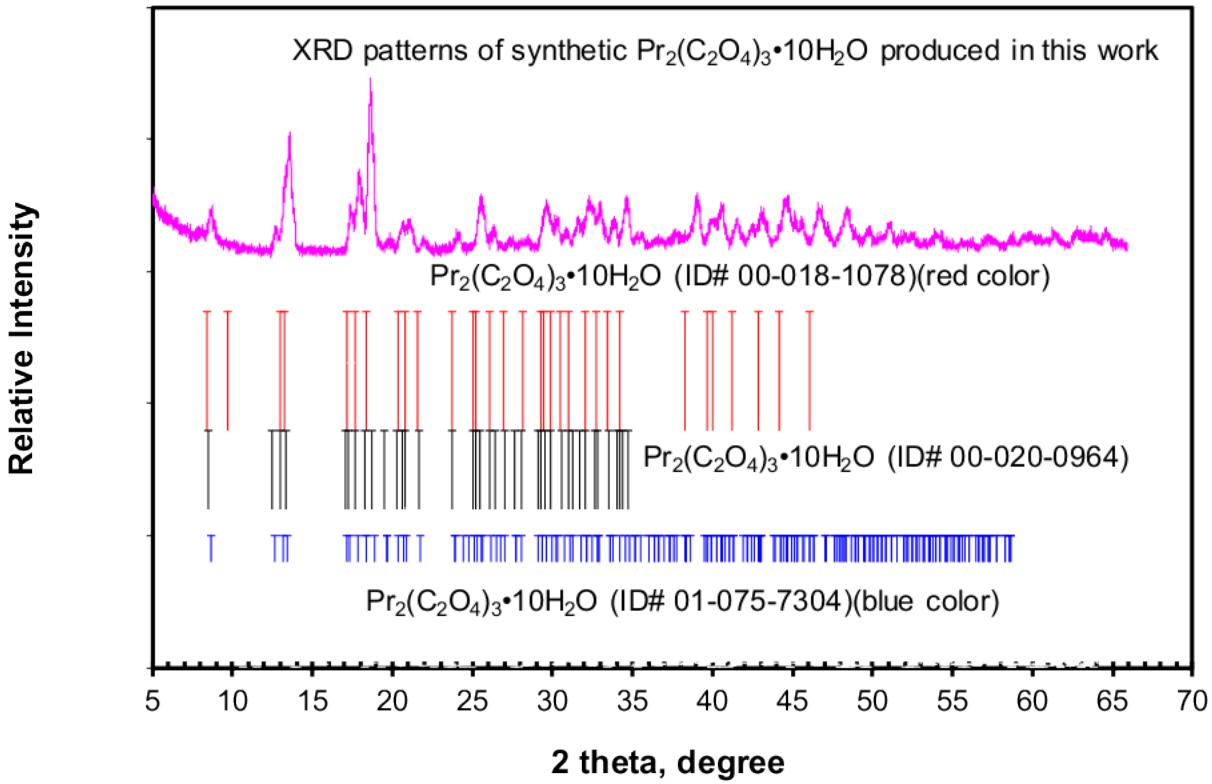
➢ **$Pr_2(C_2O_4)_3 \cdot 10H_2O$ was synthesized**

- Dropwise addition of 0.67 M $PrCl_3$ into 0.18 M $H_2C_2O_4$
- $Pr_2(C_2O_4)_3 \cdot 10H_2O$ has an identical stoichiometry with the plutonium analog, $Pu_2(C_2O_4)_3 \cdot 10H_2O$
- $Pr_2(C_2O_4)_3 \cdot 10H_2O$ has the characteristic light green color
- Pr(III) has effective ionic radii almost identical to those of Pu(III)
  - For instance, for six coordination number, 0.99 Å for Pr(III) versus 1.00 Å for Pu(III)

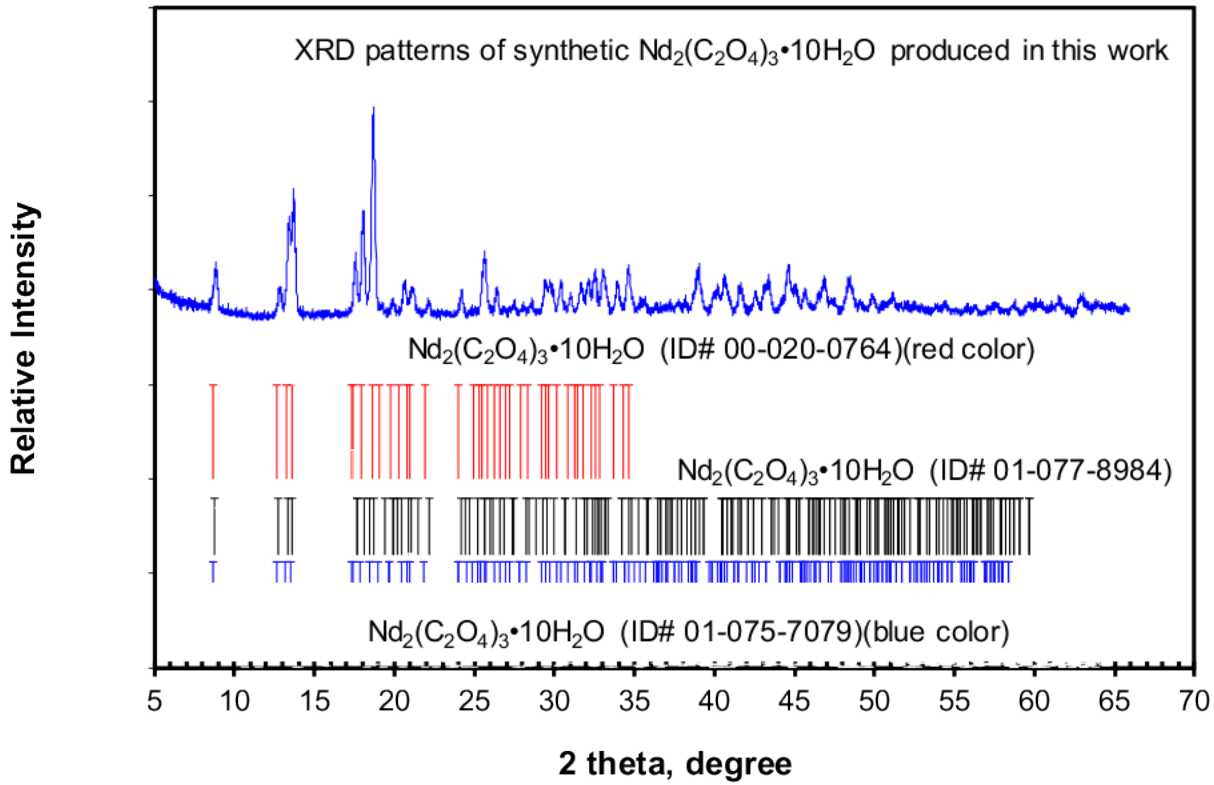
➢ **$Nd_2(C_2O_4)_3 \cdot 10H_2O$ was synthesized**

- Dropwise addition of 0.70 M $NdCl_3$ into 0.18 M $H_2C_2O_4$
- $Nd_2(C_2O_4)_3 \cdot 10H_2O$ has an identical stoichiometry with the americium and curium analogs, $Am_2(C_2O_4)_3 \cdot 10H_2O$ and $Cm_2(C_2O_4)_3 \cdot 10H_2O$
- $Nd_2(C_2O_4)_3 \cdot 10H_2O$ has the characteristic light purple color
- Nd(III) has effective ionic radii close to those of Am(III) and Cm(III)
  - For instance, for six coordination number, 0.983 Å for Nd(III) versus 9.75 Å for Am(III) and 0.97 Å or Cm(III)

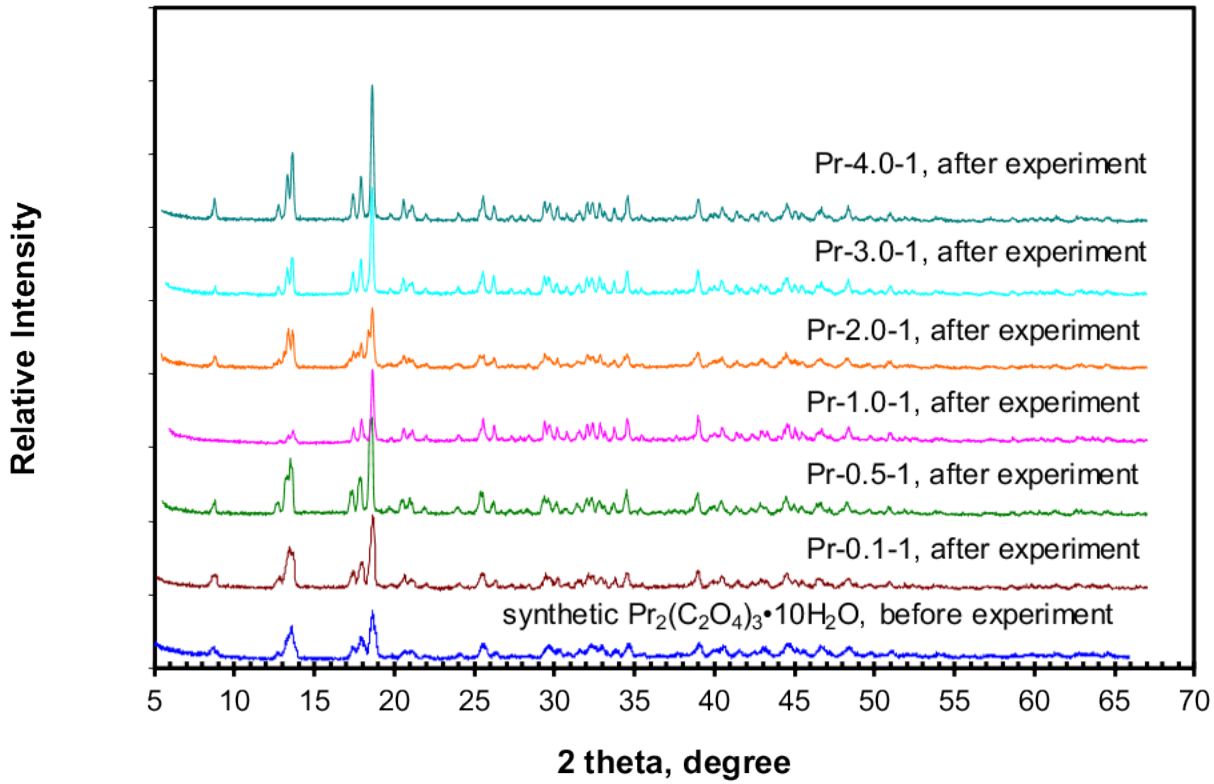
EXPERIMENTAL RESULTS: STARTING MATERIAL—SYNTHETIC PR-OXALATE



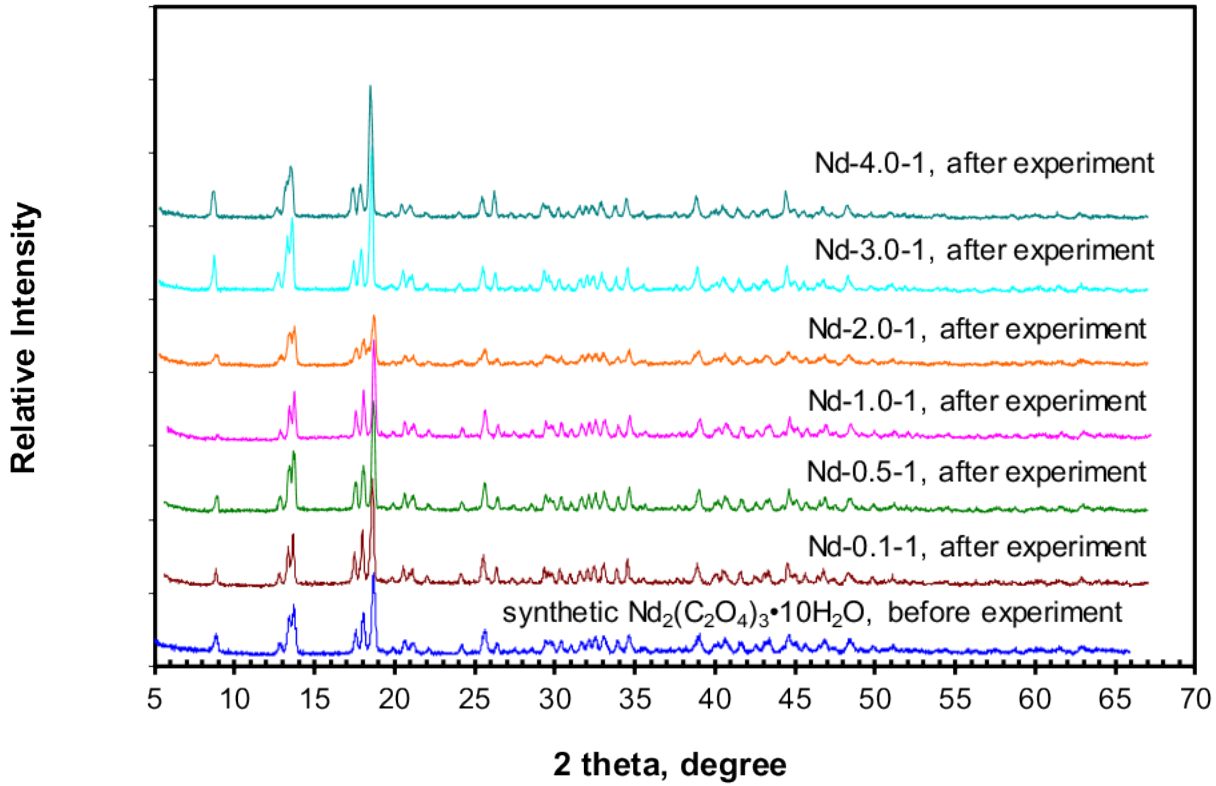
EXPERIMENTAL RESULTS: STARTING MATERIAL—ND OXALATE



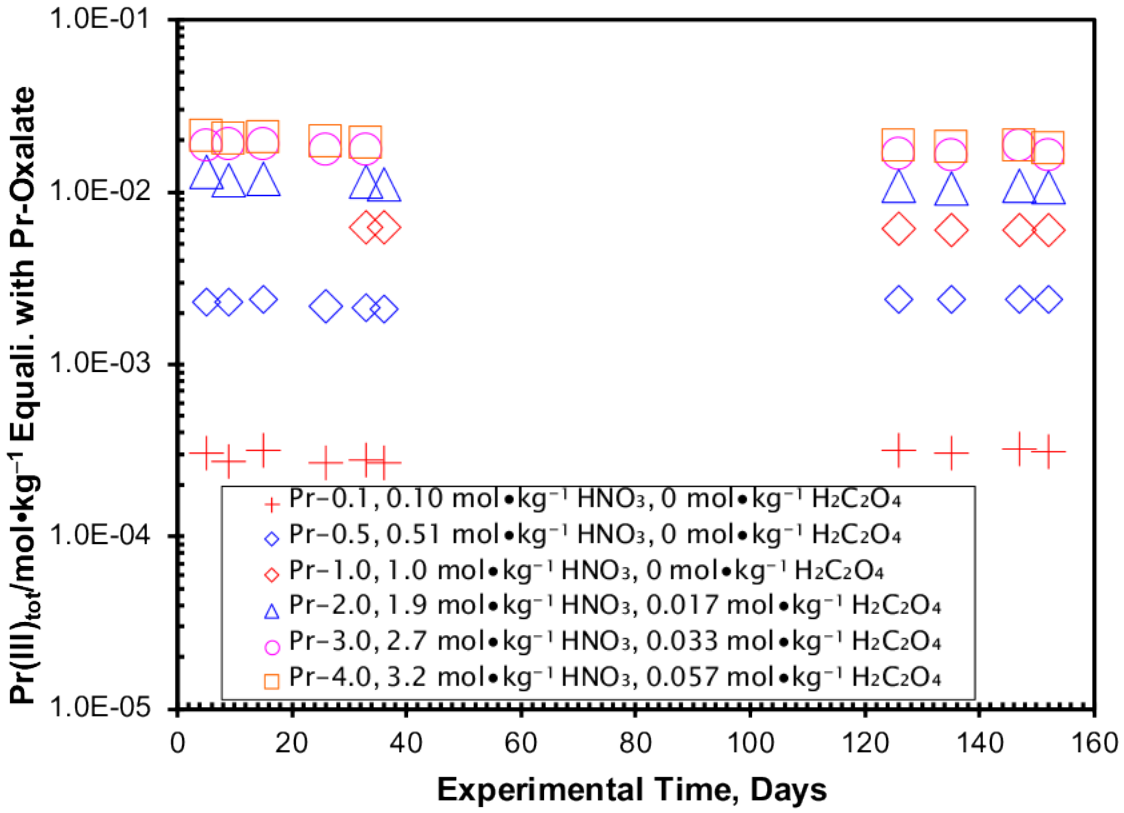
EXPERIMENTAL RESULTS: XRD PATTERNS OF EXPERIMENTS FOR PR-OXALATE



EXPERIMENTAL RESULTS: XRD PATTERNS OF EXPERIMENTS FOR ND-OXALATE



EXPERIMENTAL RESULTS: SOLUTION CHEMISTRY



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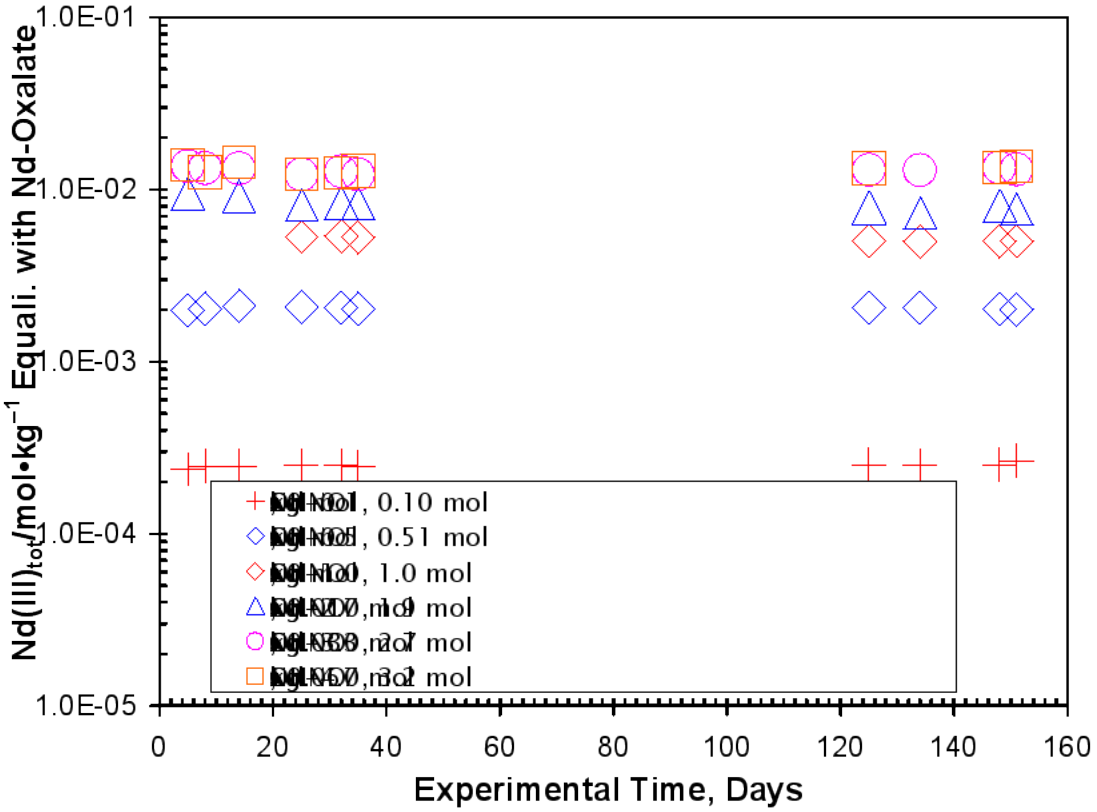
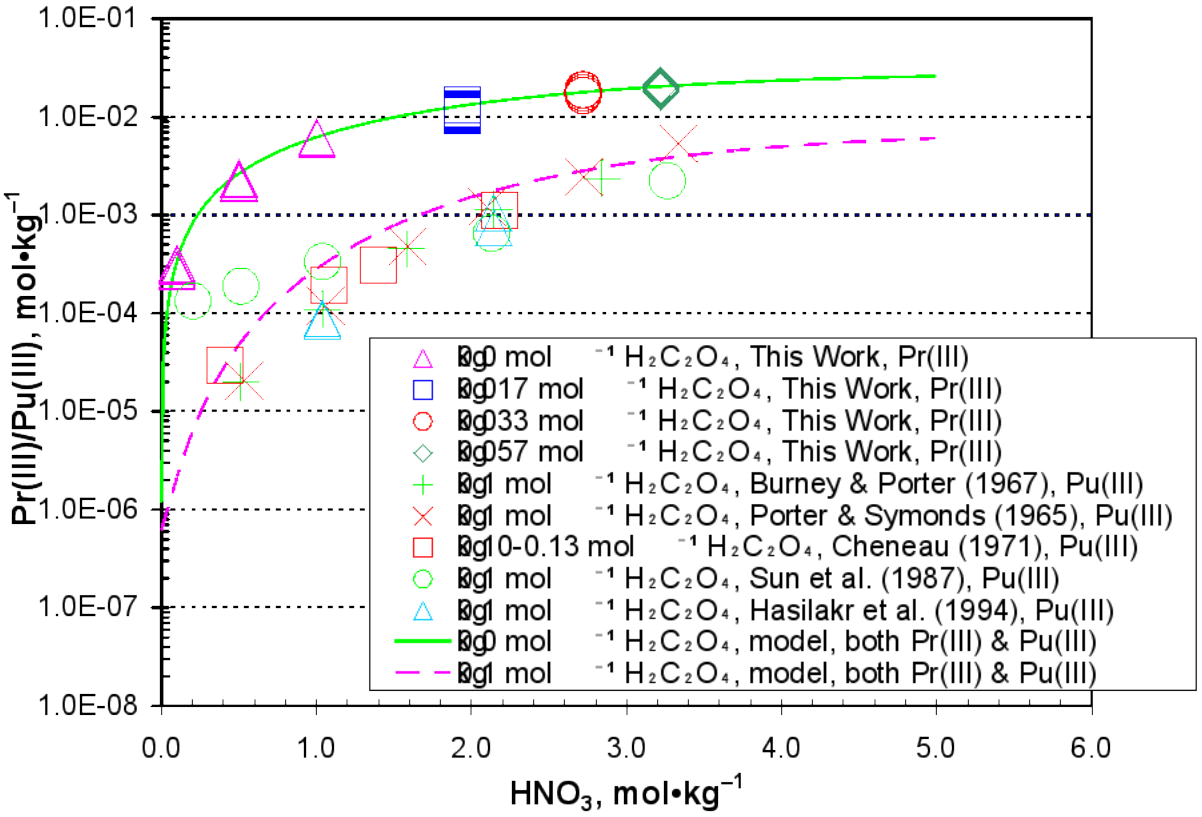
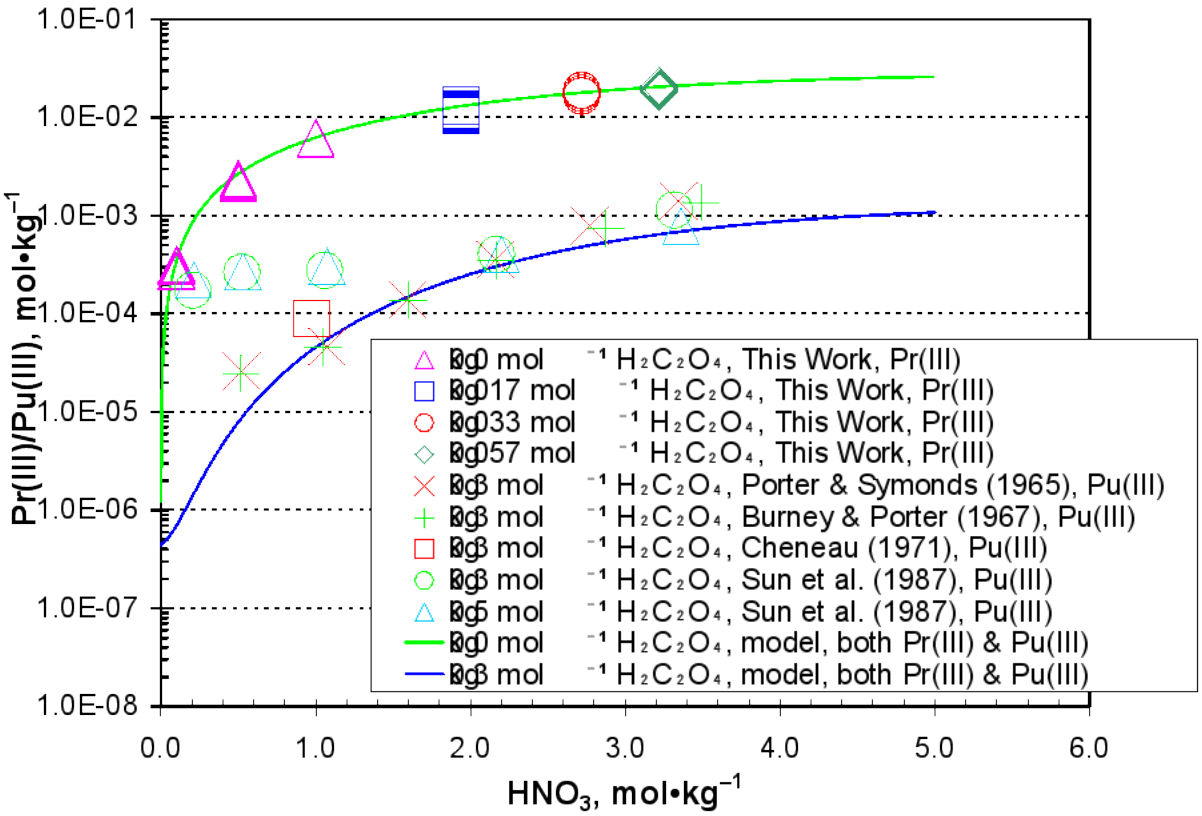


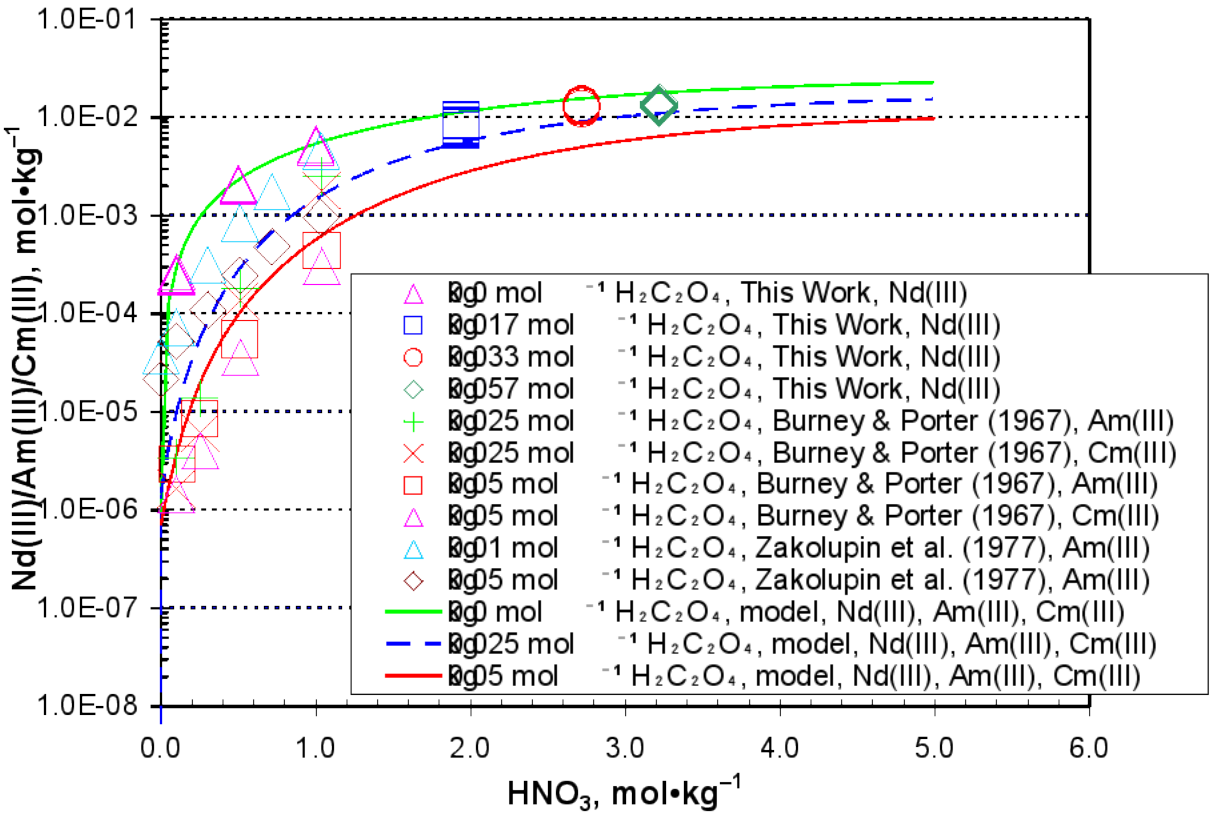
MODELING RESULTS: PR(III)/PU(III)



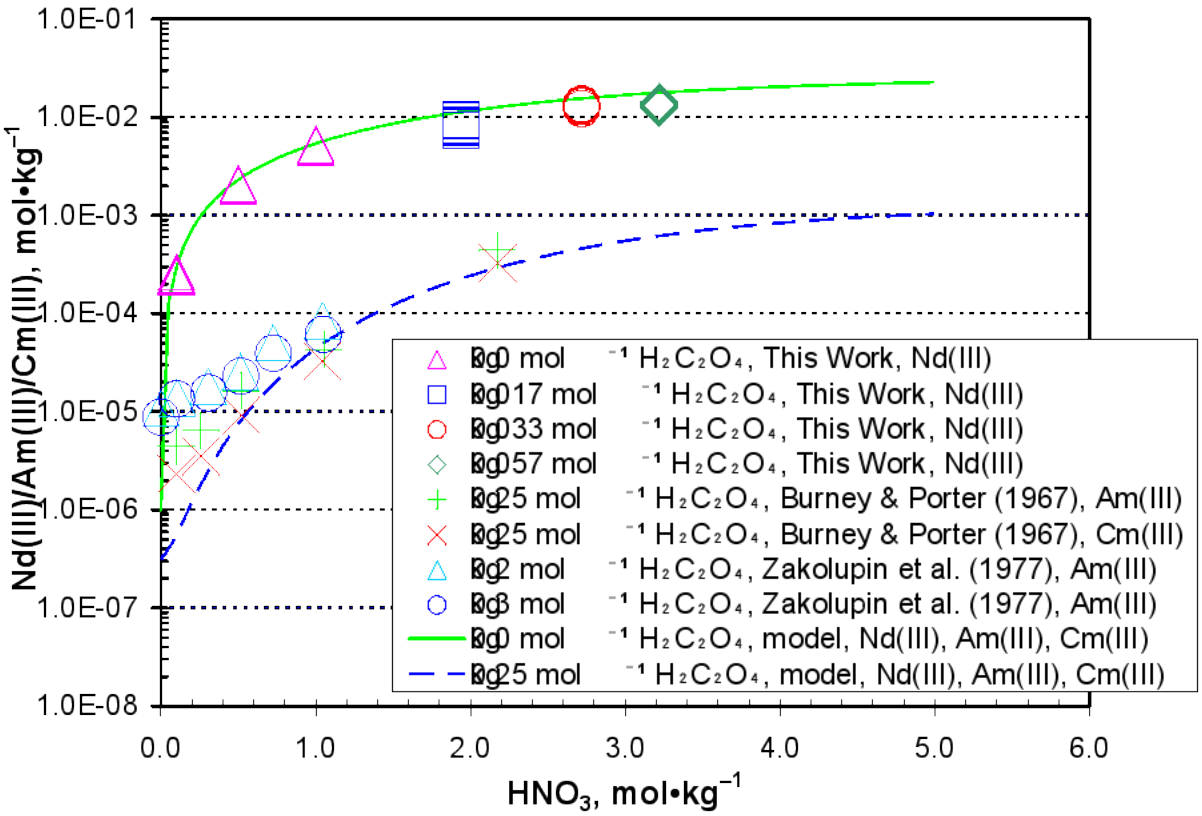
MODELING RESULTS: PR(III)/PU(III)



MODELING RESULTS: ND(III)/AM(III)/CM(III)



MODELING RESULTS: ND(III)/AM(III)/CM(III)



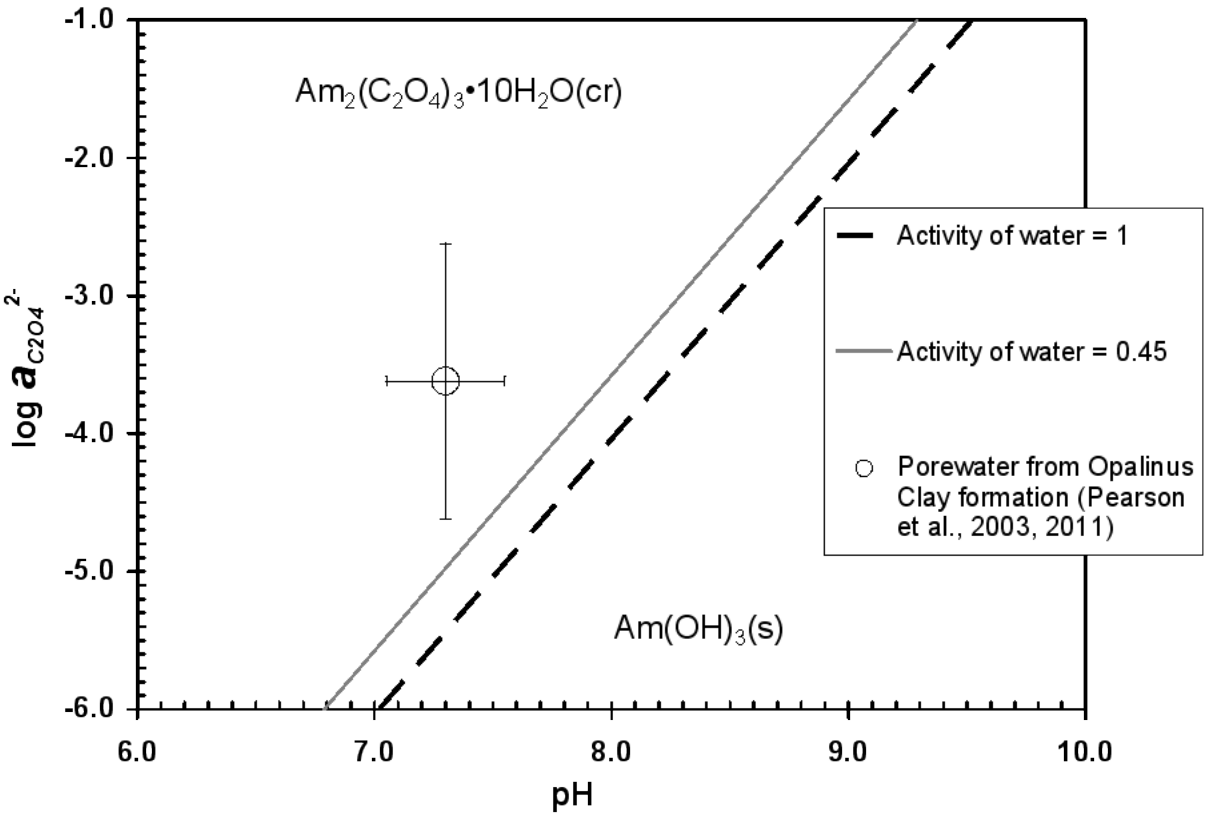
APPLICATIONS

- Nuclear Waste Contaminated Sites
  - Chernobyl
  - Lake Karachay
- Near Field of Geological Repositories
- Far Field of Geological Repositories
  - Leaked actinides from a breached HLW repository interact with oxalate, e.g., whewellite, $CaC_2O_4 \bullet H_2O$, to form actinide oxalates to retard and limit the movement of actinides.

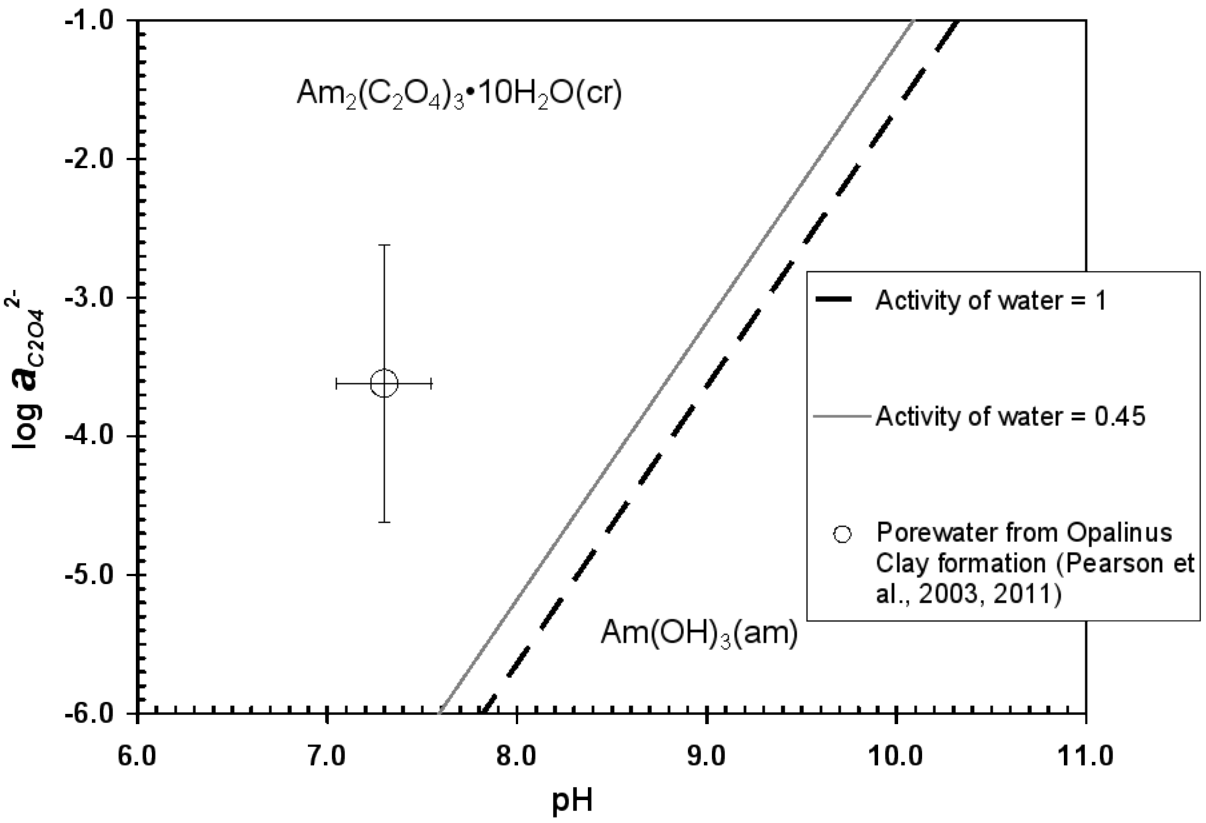
For details, please see:

Xiong, Y. and Wang, Y., 2021. Experimental and modeling studies of PR and ND oxalate solubility to high ionic strengths: Insight into actinide (III) oxalates. *Chemical Geology*, *573*, p.120200.

APPLICATIONS: NEAR FIELD



APPLICATIONS: NEAR FIELD



APPLICATIONS: NEAR FIELD