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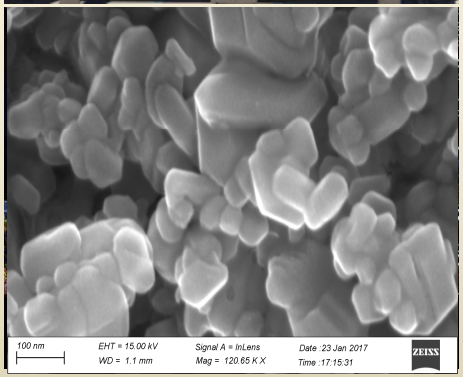

Solubility of $Nd(OH)_3$ at High pH

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Jonathan P. Icenhower*, Charlotte Sisk-Scott, Leslie D. Kirkes, Jandi Knox, Cassandra R. Marrs and Justin Dean

*Repository Performance Group, Sandia National Laboratories, Carlsbad, New Mexico, USA

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$Nd(OH)_3$ Solubility—The Link to Actinide Behavior

- The WIPP repository is disposing actinide-bearing waste, including Pu(III) and Am(III).
- The mobility of the actinides is dependent upon the solubility of $An(OH)_3$.
- Nd(III) has an ionic radius [112 pm] similar to that of Pu(III) and Am(III) [114 and 112 pm, resp.] in 6-fold coordination.
- Solubility of $Nd(OH)_3$ is therefore a good proxy for the solubility of $An(OH)_3$.

Previous Work and Motivation for this Study

- Numerous studies of $Nd(OH)_3$ solubility have been carried out, but with little consensus of results.
- In cement-lined repositories, initial interaction between aqueous solution and cement will result in highly alkaline (pH > 9) conditions.
- Our investigation will focus on the solubility of $Nd(OH)_3$ as a function of pH (10 to 13) and ionic strength (0.2 to 5.0 M NaCl).
- Are data for $Nd(OH)_3$ representative of data for $Am(OH)_3$ and $Pu(OH)_3$?

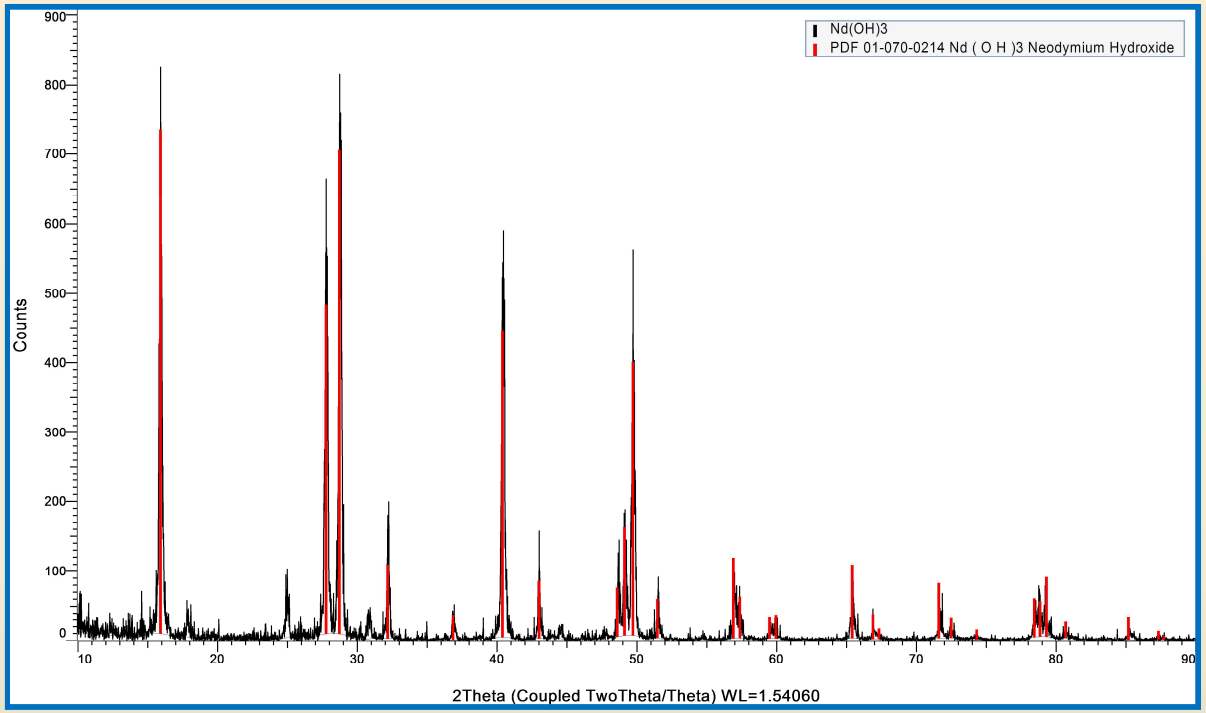
Outline of Presentation

- Overview of experimental design, direction.
- Materials and Methods.
  - Experimental setup
  - Crystal synthesis
- Results.
  - Maintenance of solution pH
  - Filtration
  - Post-run XRD
  - Solution analyses—comparison to Am and Pu work
- Future work.
- Conclusions.

Materials and Methods
Experiments at High pH (≥10)

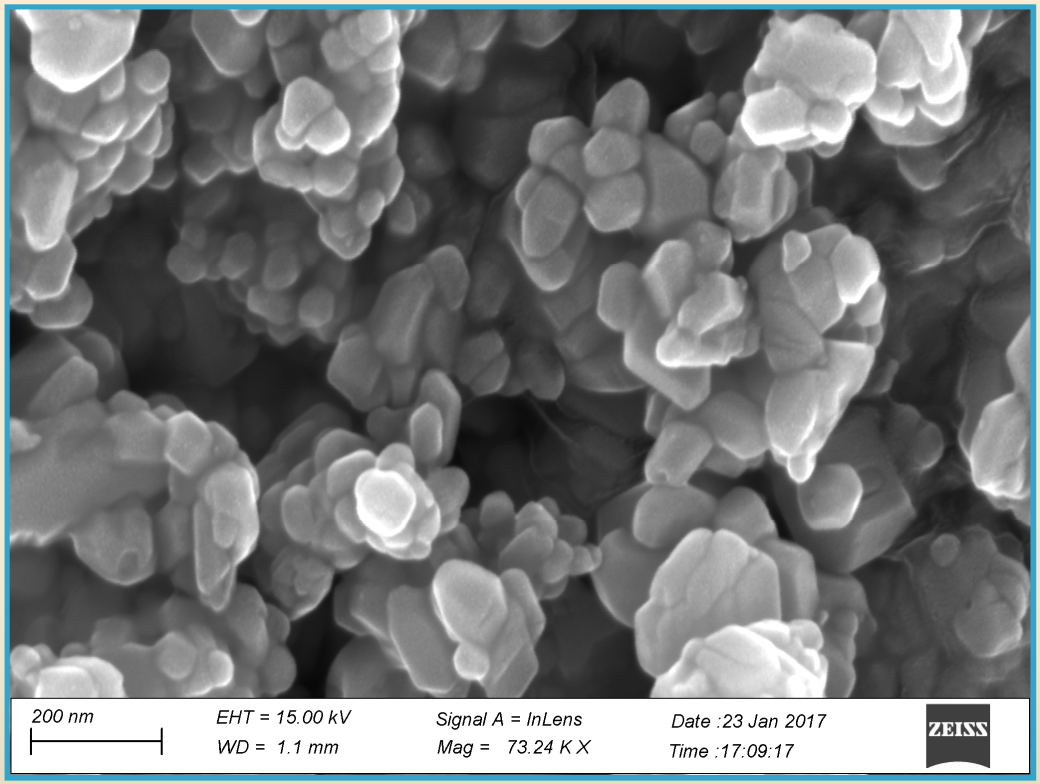
- Experiments conducted in the undersaturated direction in dilute (Na <0.2 *m*).
- Experiments performed in glove box to avoid formation of $Nd_2(CO_3)_3$ and to help maintain pH.
- The pH (10 to 13) was set by addition of NaOH; pH values tracked during experiment.
- $Nd(OH)_3$ synthesized hydrothermally from Nd_2O_3 powder.
- Identity of $Nd(OH)_3$ confirmed by XRD.
- Solutions filtered through 0.2 μm opening.
- Solutions processed in all glass labware; no plastics!

XRD of Synthesized $Nd(OH)_3$ Crystals



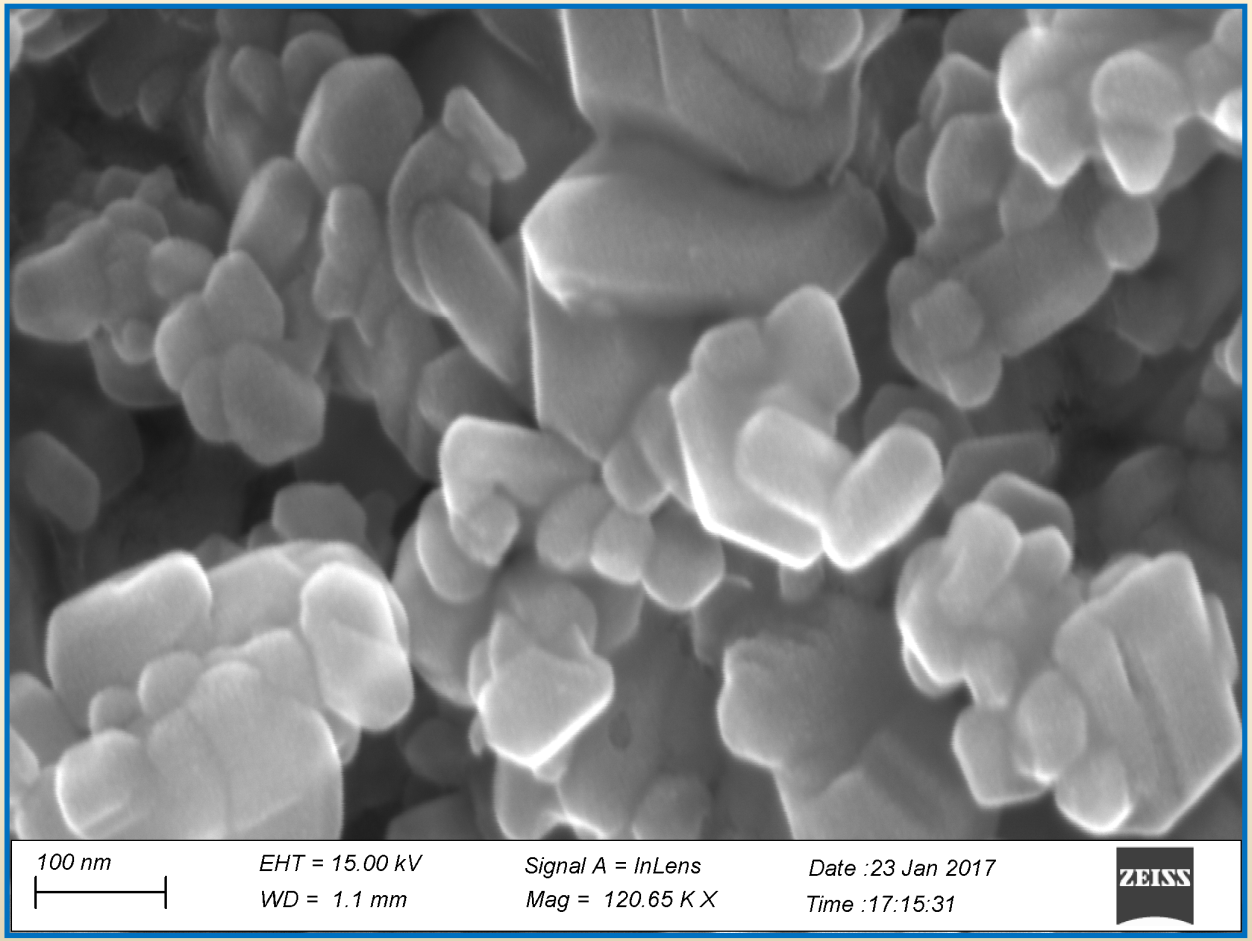
- Synthesized powders analyzed by XRD using controlled atmosphere holder.
- Sharp narrow peaks indicate high crystallinity.

SEM Images of Synthesized $Nd(OH)_3$

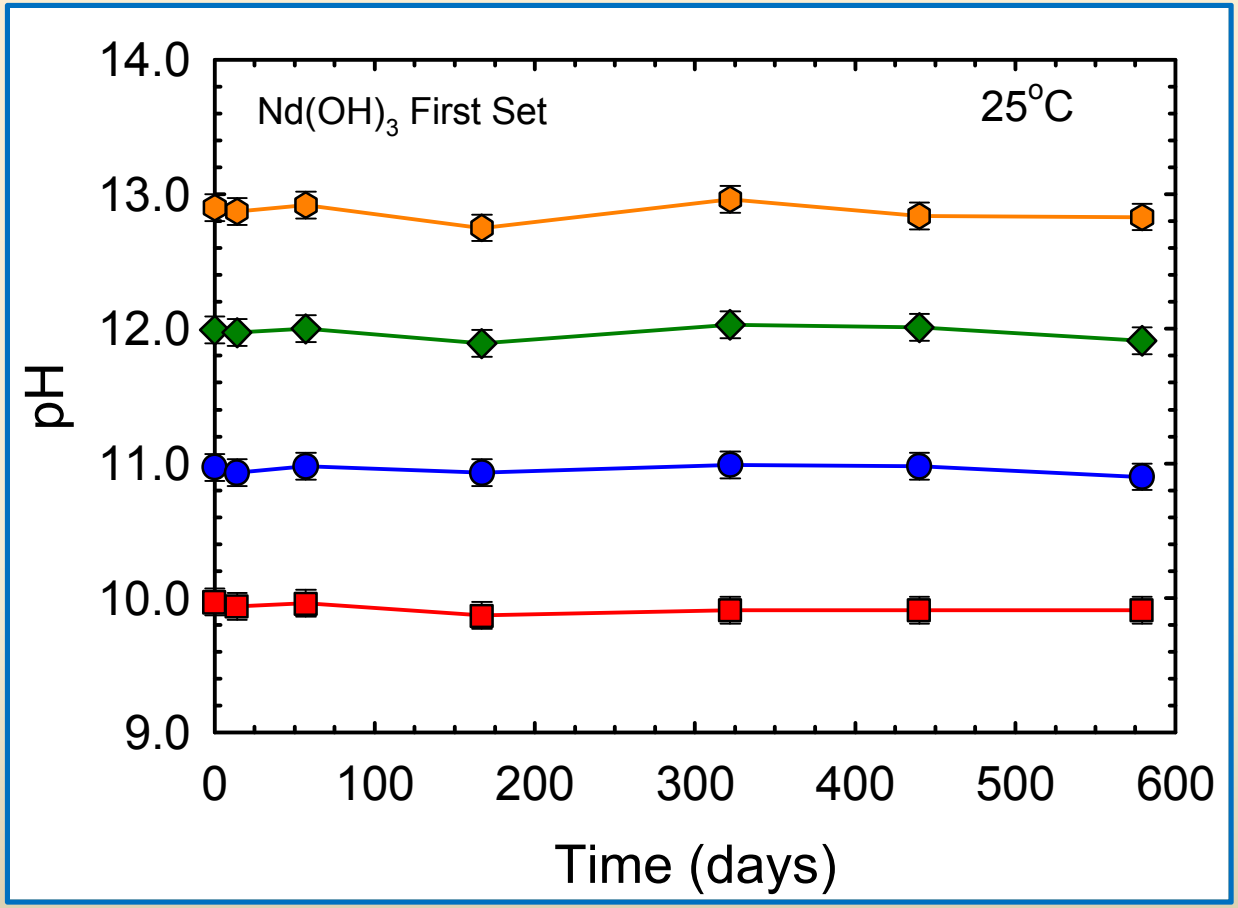


- Images reveal very fine-grained product (<1 μm dimensions).
- Crystals are well-formed, highly crystalline.
- Post-run crystals will also be imaged by SEM.

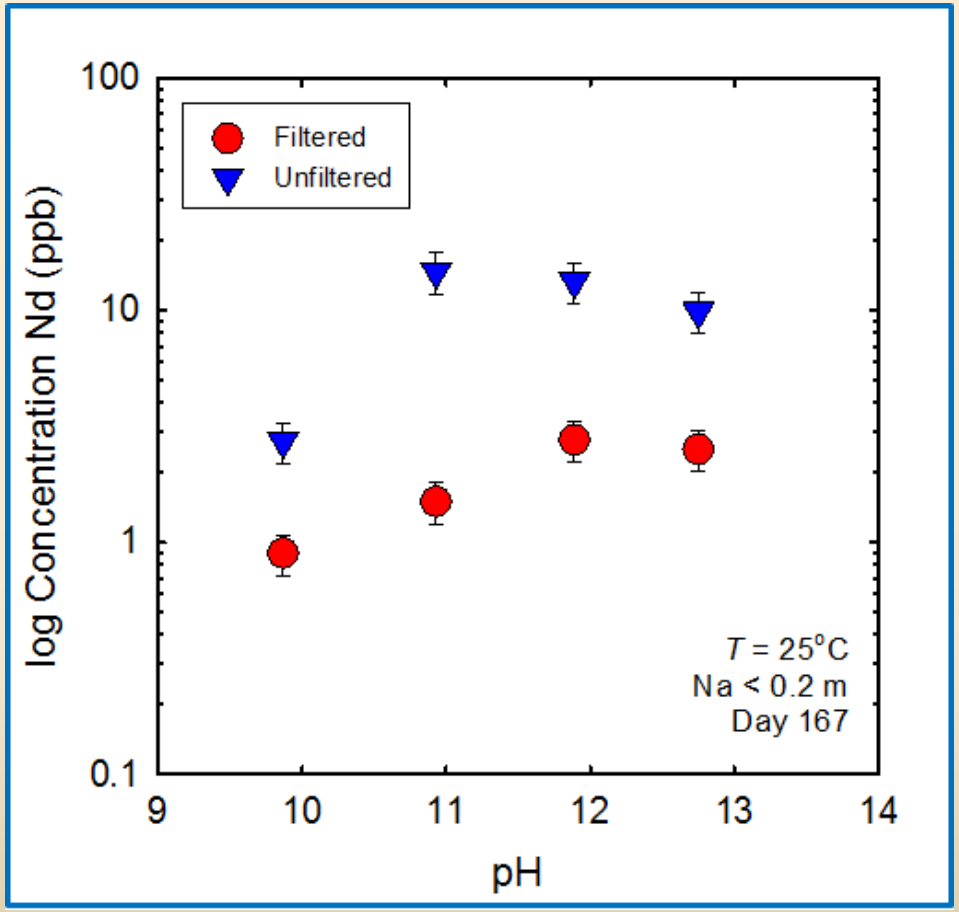
SEM Image of Synthesized $Nd(OH)_3$



Results—Maintenance of pH over Time

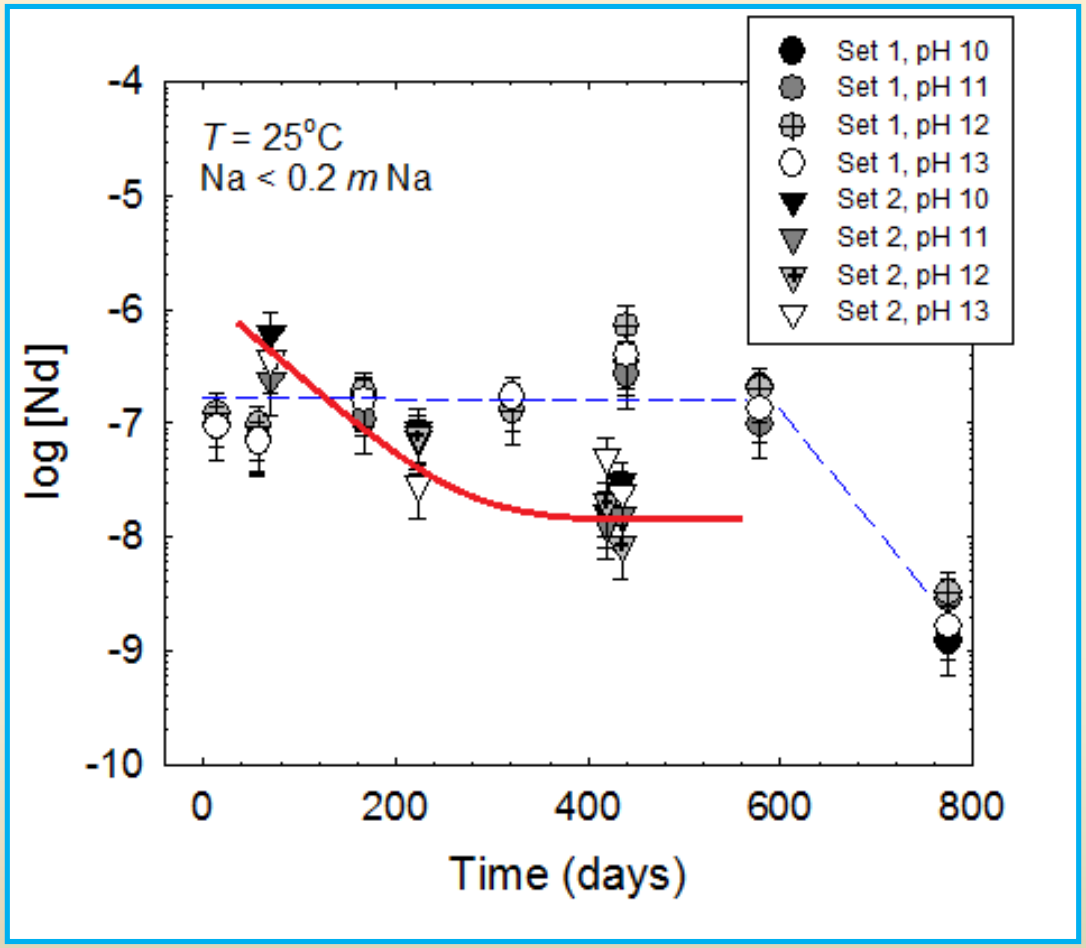


Solution Chemistry—The Importance of Solution Filtration

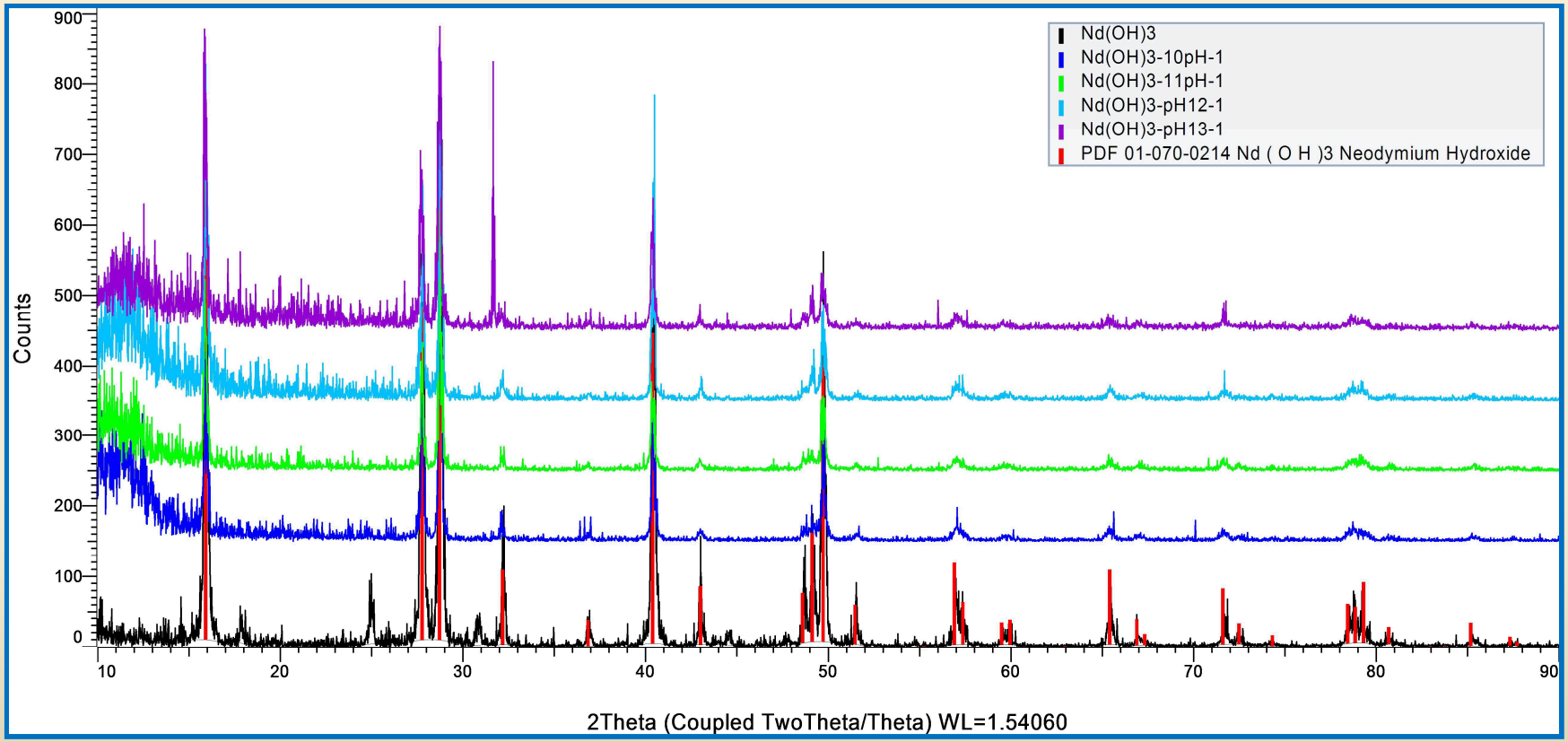


- Samples that were filtered (0.20 μm) yield lower Nd concentrations than those that were not.
- No difference between those filtered at 0.20 μm and those filtered down to 10 kDa.

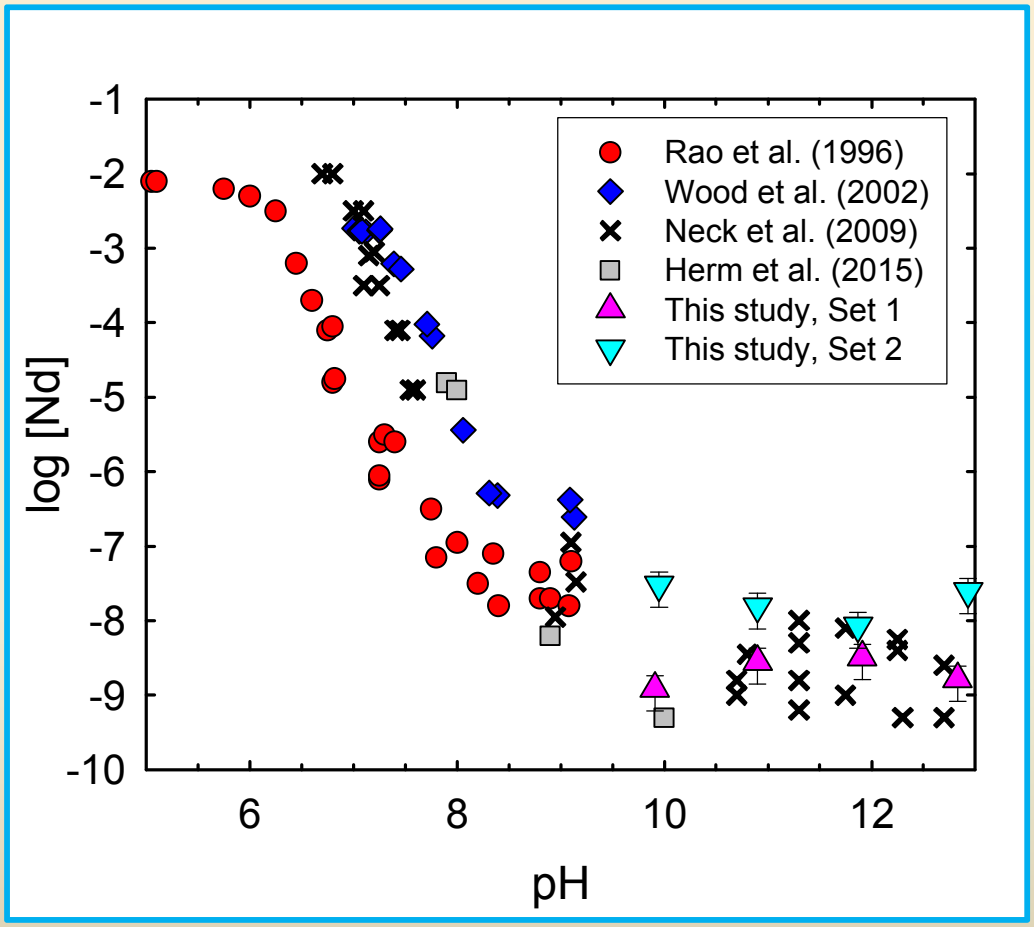
Equilibrium Concentrations of Nd



XRD of Post-Run Powders

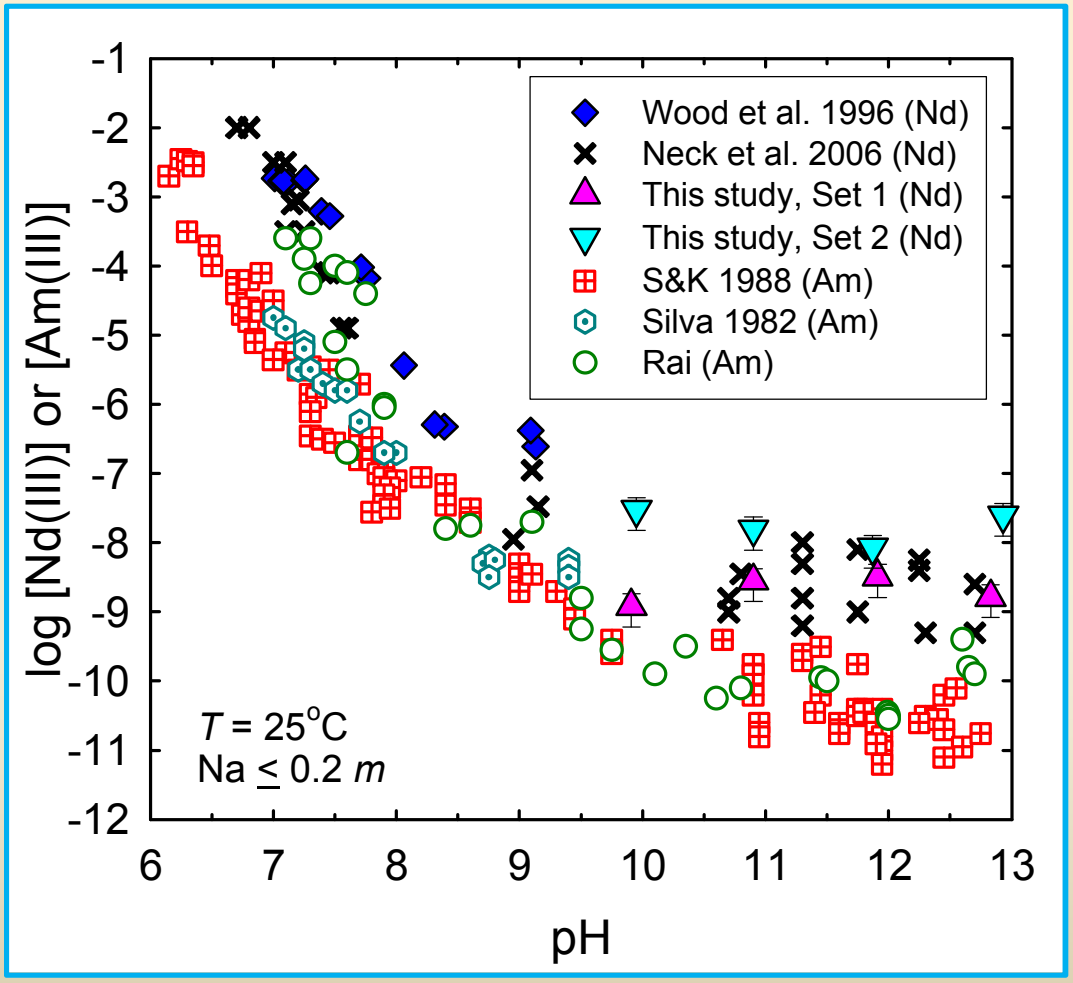


Concentrations of Nd—Comparison to Previous Nd Work



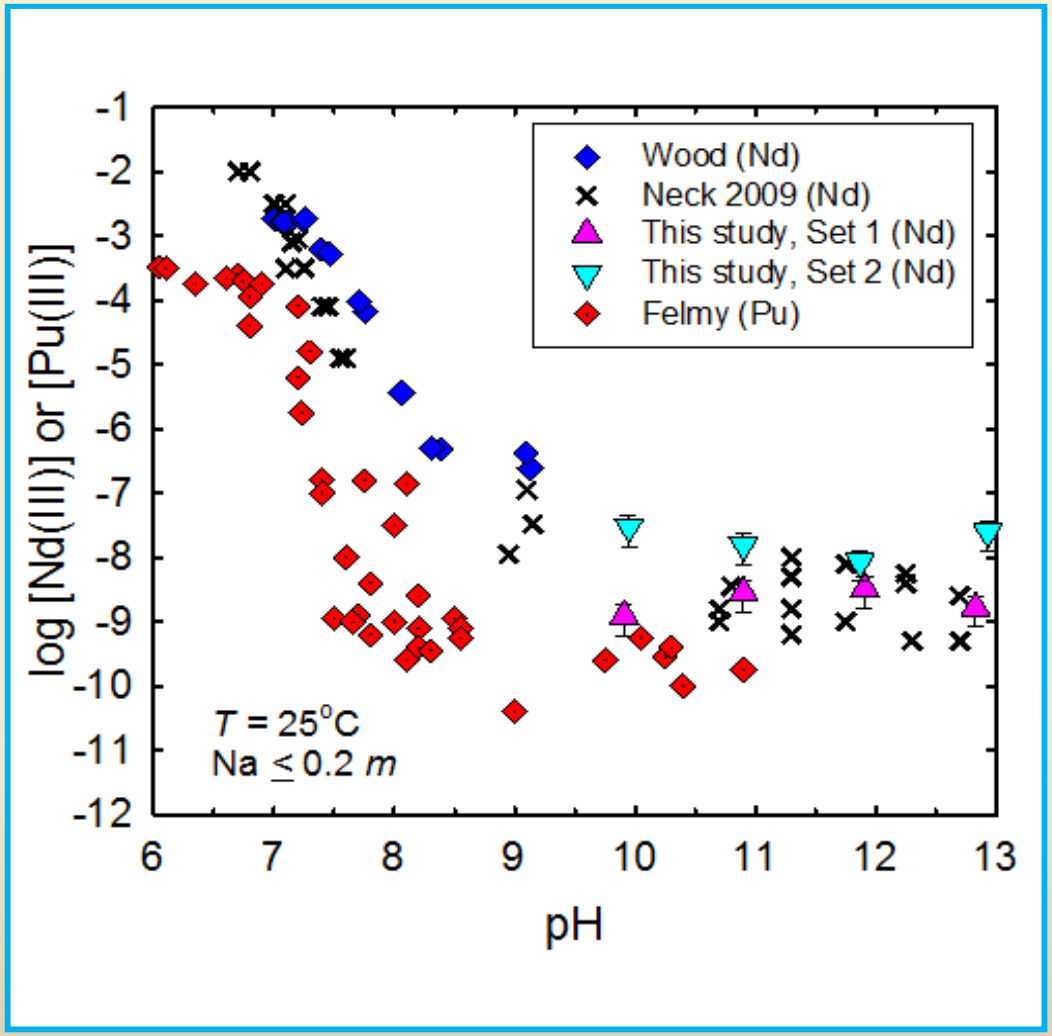
- Our results form a continuum with those from previous investigators.
- Concentrations of Nd are independent of pH.
- No evidence for the existence of $[Nd(OH)_4]^-$ species up to pH = 13.

Concentrations of Nd—Comparison to $Am(OH)_3$ Results



- Results for $Nd(OH)_3$ are generally similar to results reported for $Am(OH)_3$.
- Yet, Nd data plot at higher concentrations compared to Am.
- Applying the results for $Nd(OH)_3$ to $Am(OH)_3$ would thus be conservative.

Concentrations of Nd—Comparison to $Pu(OH)_3$ Results



- Results for $Nd(OH)_3$ are also generally similar to results reported for $Pu(OH)_3$.
- Yet, Nd data plot at higher concentrations compared to Pu.
- Applying the results for $Nd(OH)_3$ to $Pu(OH)_3$ would thus also be conservative.

Conclusions and Future Work

- Preliminary work at 25°C at high pH (10 to 13) indicates that solubility of *crystalline* $Nd(OH)_3$ is very low (Nd $<1.0 \times 10^{-8}$ mol/L).
- Results form a continuum from previous work at higher pH.
- Results of all previous work indicate that Nd concentrations are higher than Am and Pu by 2 - 3 and 1 - 2 O.M., respectively.
- Future work will focus on combination of high-*T* (up to 200°C) and pH (10 to 13) and ionic strength (up to 5 M).
- Explore amorphous vs. crystalline materials, especially in view of self-inflicted radiation damage in $Am(OH)_3$ and $Pu(OH)_3$.