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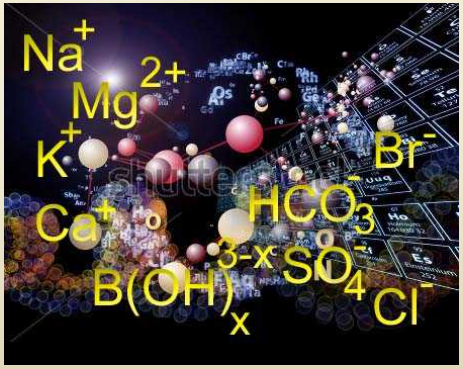

Solubility of $Nd(OH)_3$: Preliminary Results at Room T and an Outline of the High-T Strategy

HiTAC (II) Workshop; April 16, 2015; Heidelberg, Germany

Jonathan P. Icenhower*, Leslie D. Kirkes and Cassandra R. Marrs

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

U.S. DEPARTMENT OF ENERGY

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Importance of $Nd(OH)_3$ Solubility

- The WIPP repository hosts actinide-bearing waste, including Pu(III) and Am(III).
- The solubility 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.
- Several studies at high *T*, but none at high *T* + high pH, or high *T* + ionic strength.
- Our investigation will focus on the solubility of $Nd(OH)_3$ as a function of *T* (25 to 250°C), pH (10 to 13), and ionic strength (0.01 to 5 M NaCl).

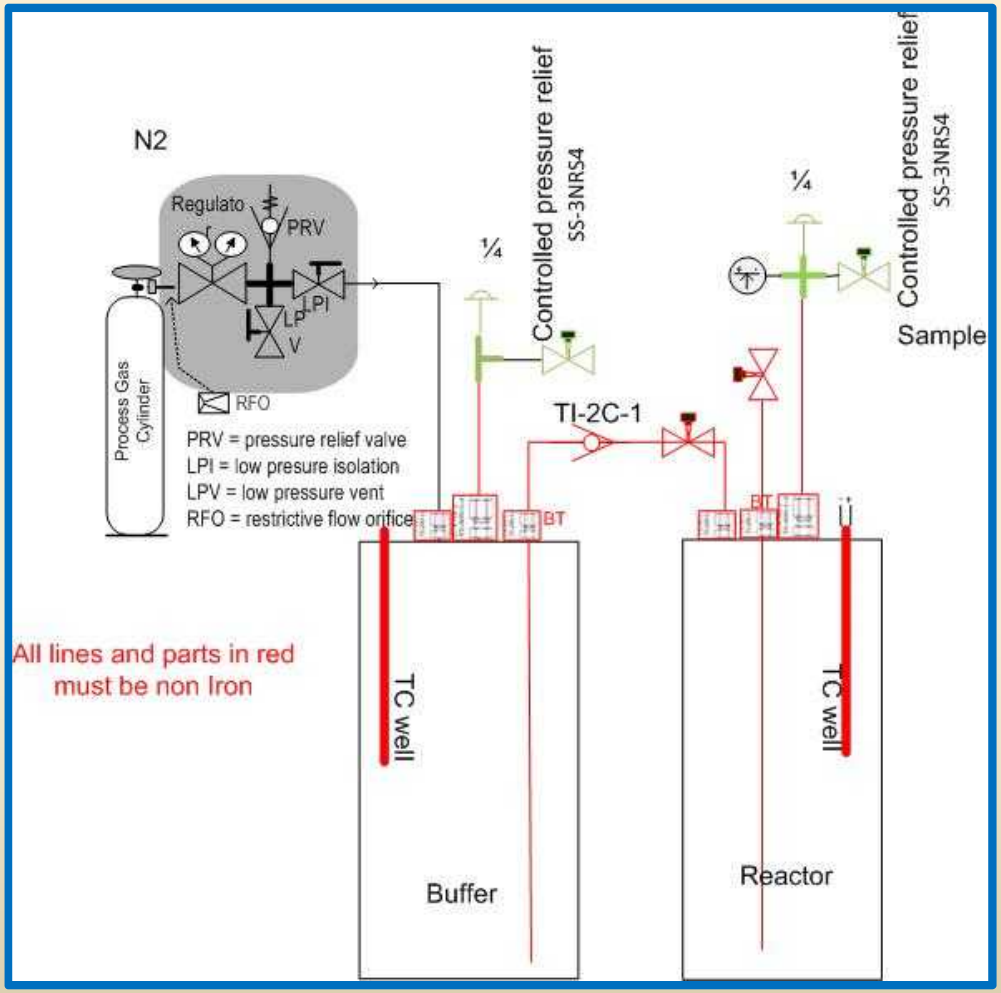
Outline of Presentation

- Overview of high-*T* and –*P* apparatus at Sandia, Carlsbad, and experimental strategy.
- Discussion of preliminary data on $Nd(OH)_3$ solubility at 25°C, pH 10 to 13.
- Future work.

Hi-TAC Apparatus

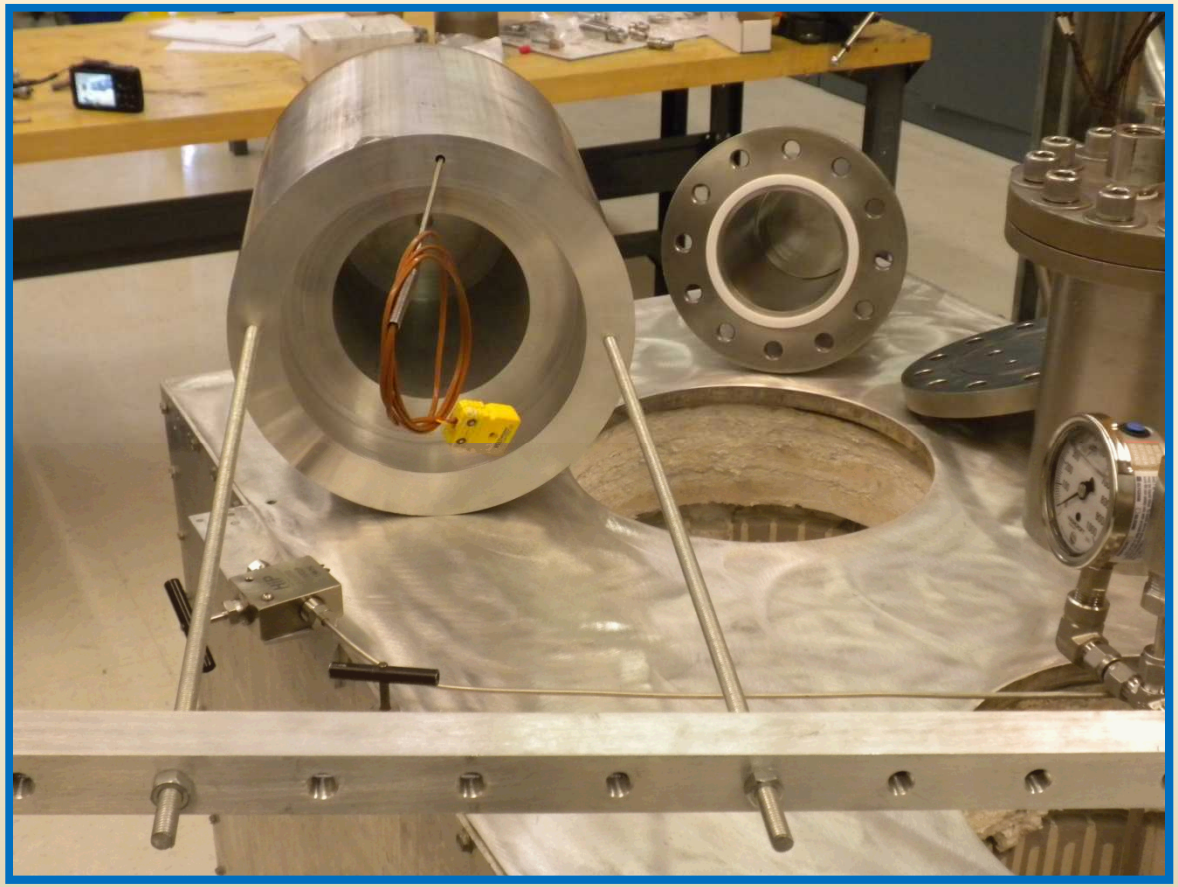
- Made from non-reactive Ti.
- Designed for temperatures up to 250°C and pressures up to 40 bar.
- Designed so that aqueous solution samples can be withdrawn at constant T & P.
- A 1-liter reactor vessel connected to a 1-liter “pressure buffer” vessel.
- “Static” reactor with no circulation of solution.

Schematic of Hi-TAC Apparatus

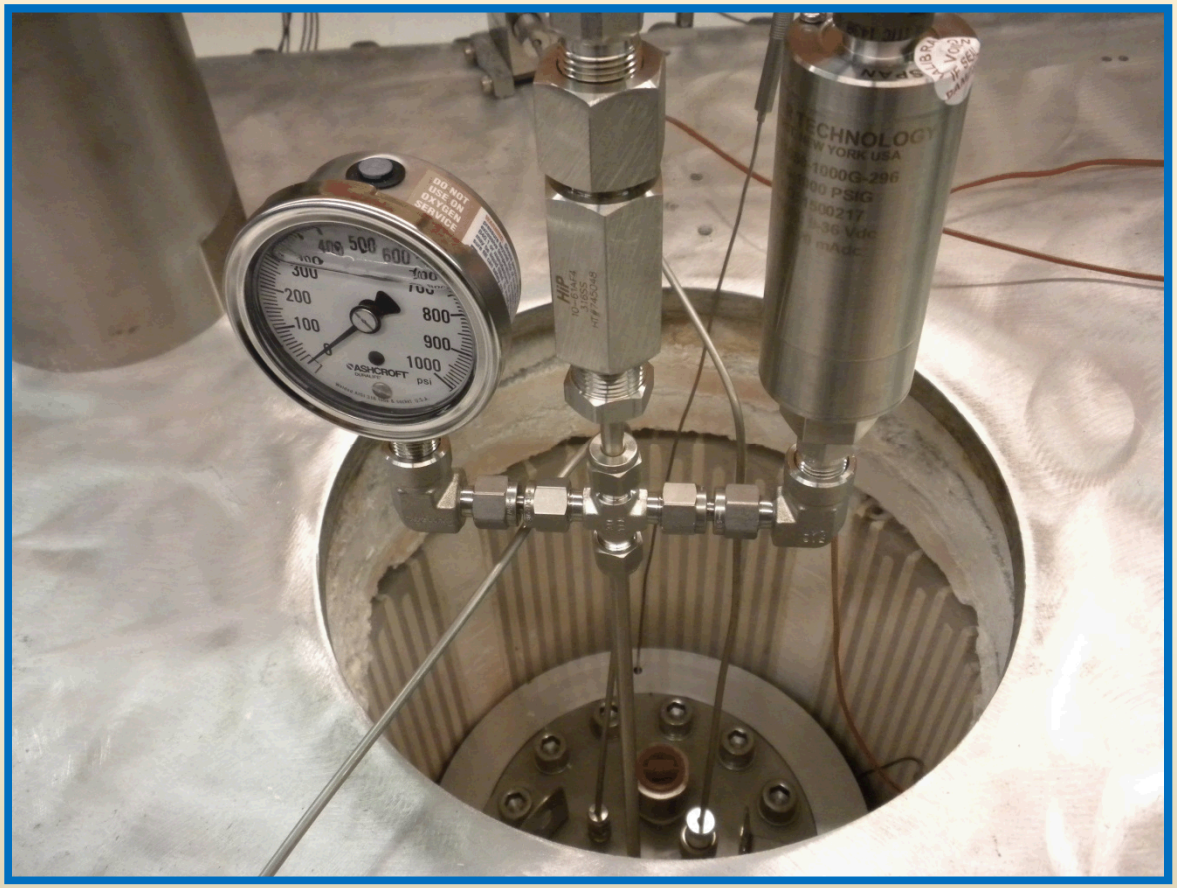


Photos of Hi-TAC Apparatus (1/4)

Photos of Hi-TAC Apparatus (2/4)



Photos of Hi-TAC Apparatus (3/4)

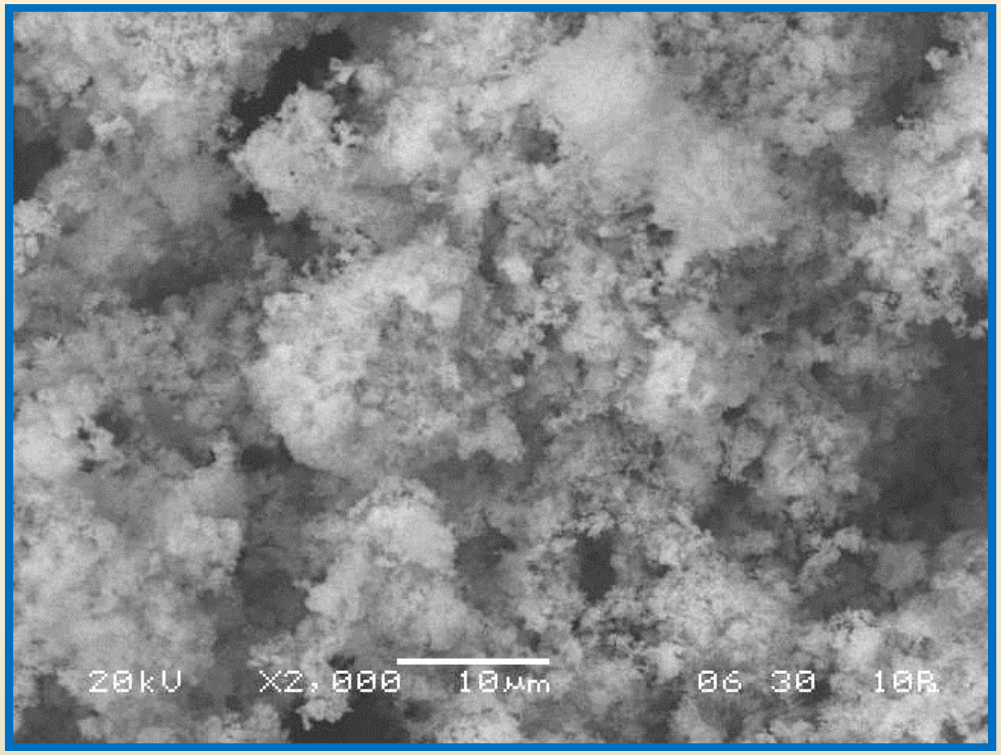


Photos of Hi-TAC Apparatus (4/4)

Experiments at High pH (≥10)

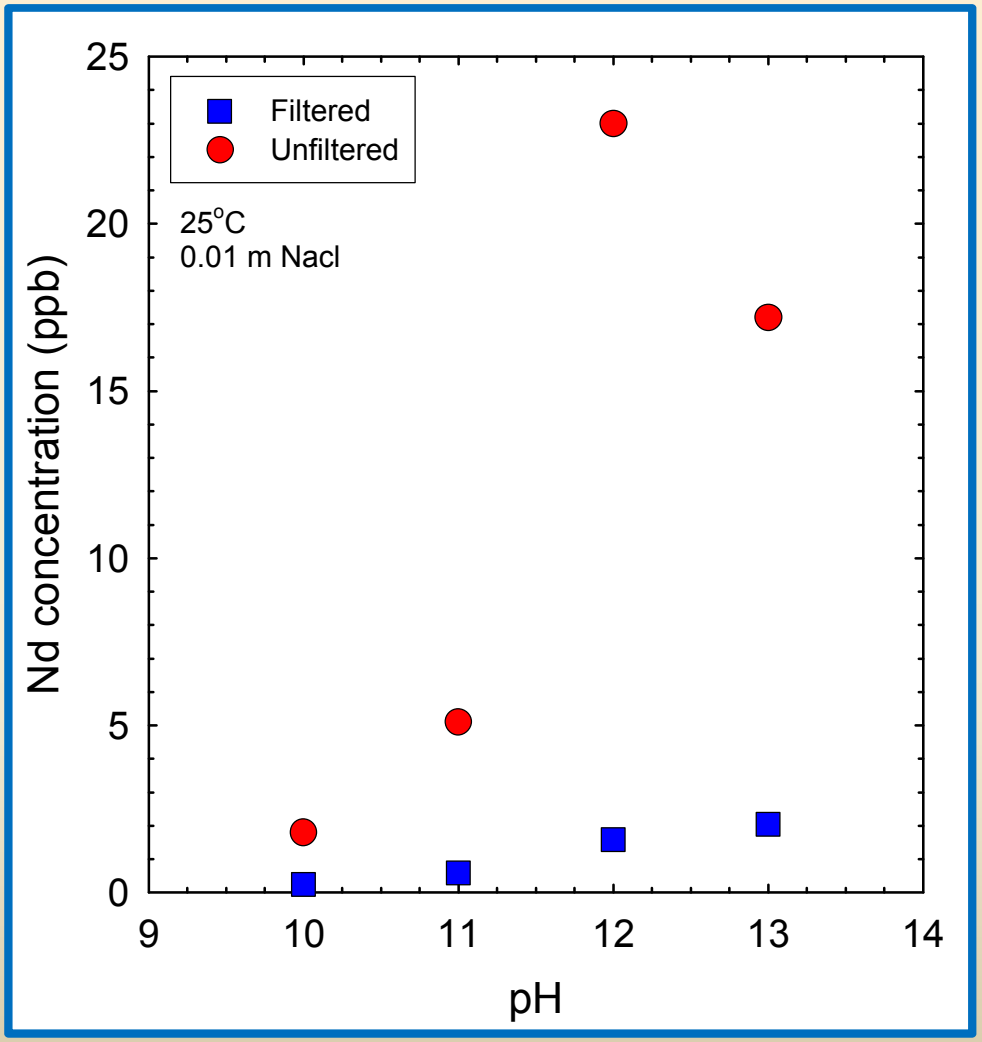
- $Nd(OH)_3$ synthesized hydrothermally from Nd_2O_3 powder.
- Identity of $Nd(OH)_3$ confirmed by XRD.
- Experiments performed in glove box to avoid formation of $Nd_2(CO_3)_3$.
- The pH (10 to 13) was set by addition of NaOH; pH values tracked during experiment.
- Solutions filtered through 0.2 μm opening.
- Filtered and unfiltered solutions analyzed by ICP-MS

SEM Images of Synthesized $Nd(OH)_3$



- Images reveal very fine-grained product (<1 µm dimensions).
- Possibility of fine crystals becoming entrained with fluid withdrawn during sampling.
- Need to synthesize larger crystals.

Preliminary Solution Chemistry



Conclusions and Future Work

- Preliminary work at 25°C at high pH (10 to 13) indicates that solubility of $Nd(OH)_3$ is very low.
- Large difference in Nd concentrations between filtered and unfiltered samples; suggests presence of small crystals and/or colloids.
- Future work will focus on combination of high-*T* (up to 250°C) and pH (10 to 13) and ionic strength (up to 5 M).