SMECTITE ILLITIZATION

Melissa Mills, Yifeng Wang, Clay Payne, Amanda Sanchez, Lydia Boisvert

Sandia National Laboratories

SFWD

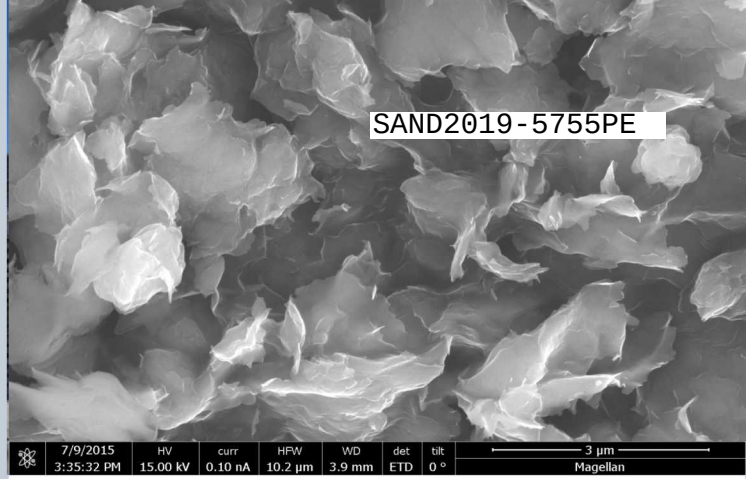
SPENT FUEL & WASTE DISPOSITION

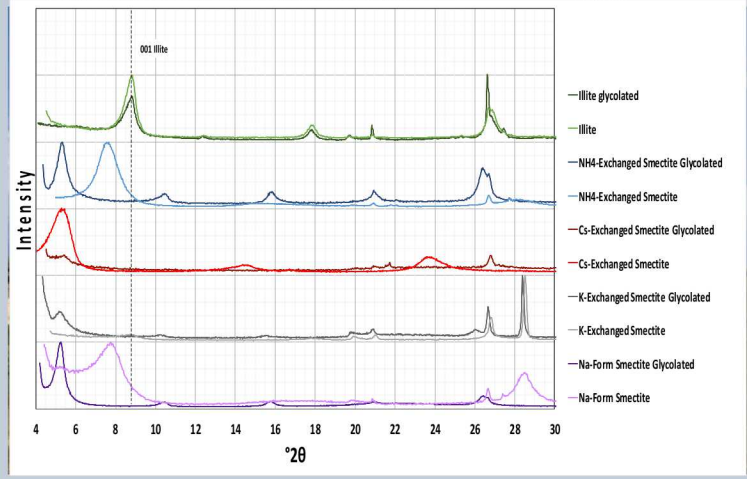
Annual Working Group Meeting

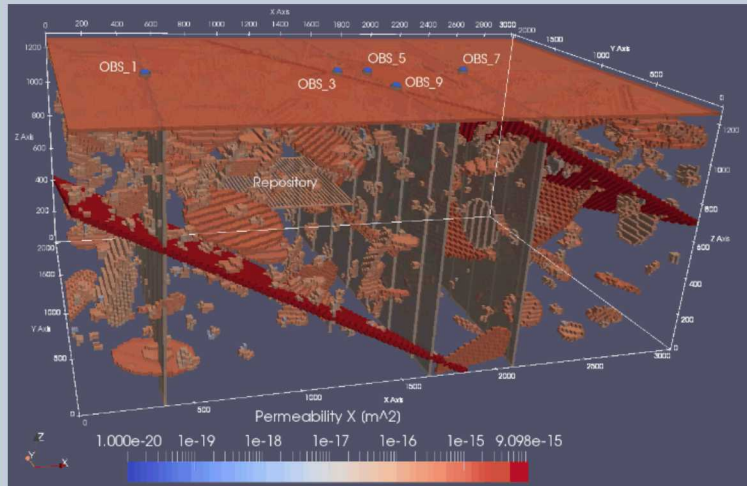
UNLV-SEB – *Las Vegas, Nevada*

May 21-23, 2019

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology & Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.







Overview

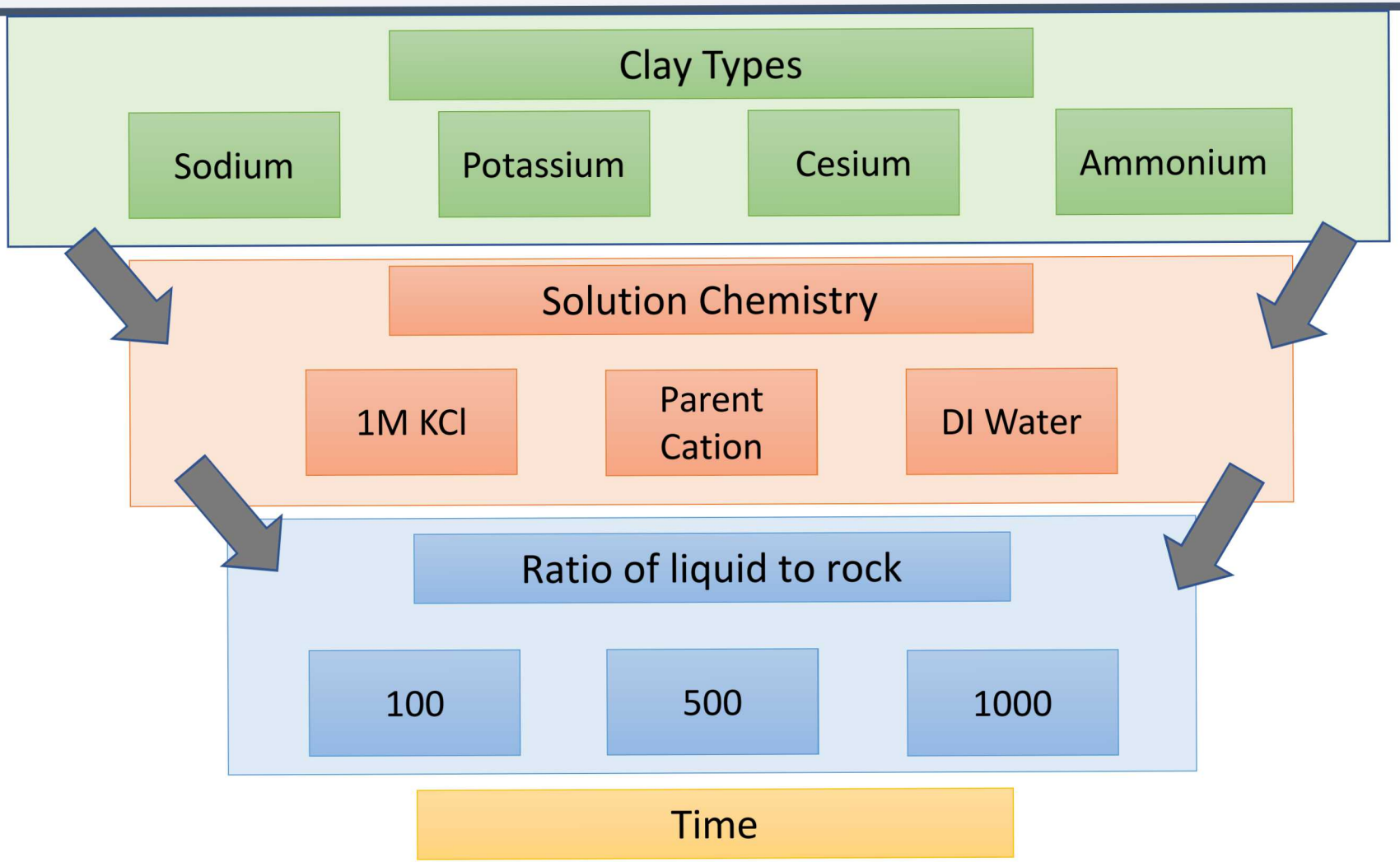
- To predict barrier functions over time within repository relevant temperatures, important to understand thermal alteration effects on montmorillonite (smectite) a main constituent of bentonite barriers
  - Conversion to illite weakens barrier functions
- Path to illitization is complex
- Important to safety case in crystalline since buffer break down is the main source of any release

Experimental Approach

200C
Start at 1 Week
Vary Rock:Liquid
Vary liquid Chemistry

- Effect of rock to liquid ratio
- Effect of interlayer cation
- Effect of solution chemistry
- Effect of time

Matrix (36 samples so far)



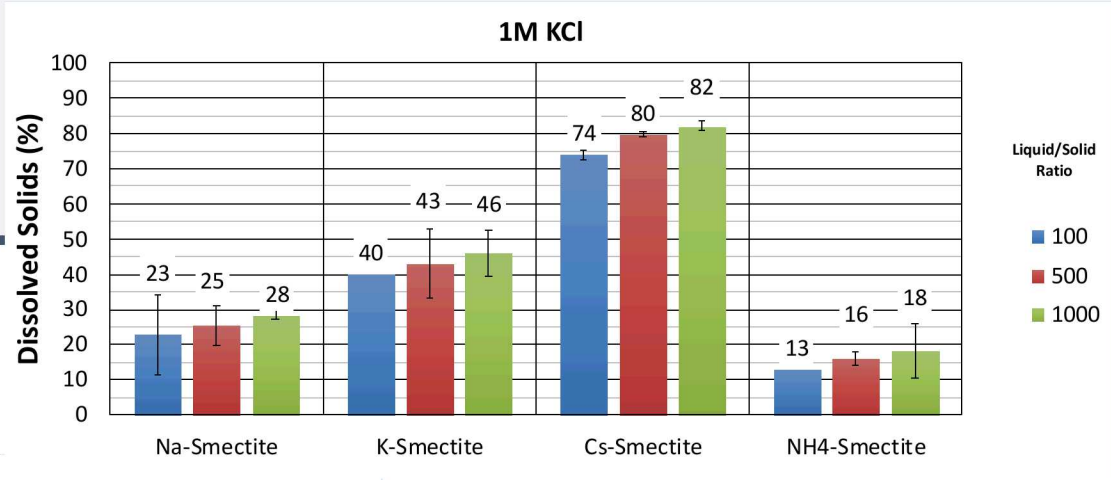
Outline of Experimental Techniques

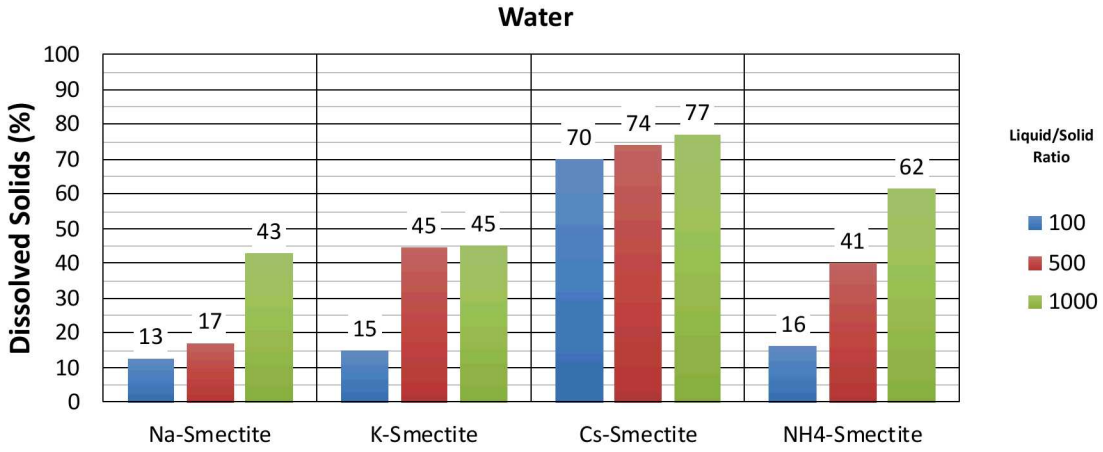
- XRD for basal spacing
- Cation exchange capacity (CEC)
- Recorded dissolved solid data and pH of reacted solutions
- Surface area by BET nitrogen adsorption
- Analysis of solutions after reaction by IC, ICP-MS, ICP-OES to determine dissolved elements
- Particle size by Malvern Zetasizer
- XRF for composition
- SEM-EDS for morphology and compositional mapping

Observations

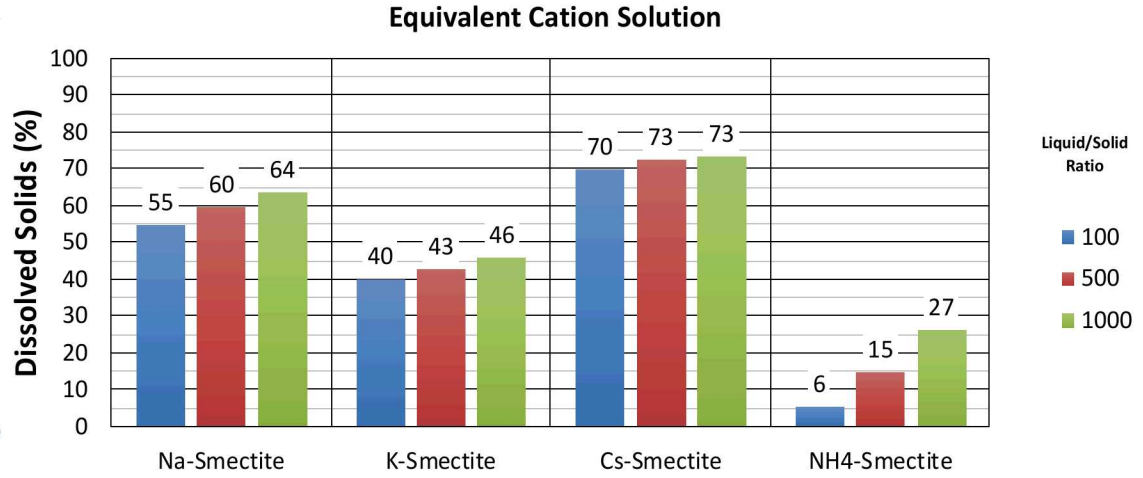
- Variable dissolution amounts
  - Solid in ≠ solid out
  - Dependent on loading and cation
- Basal spacing changes
  - XRD peaks shift
  - Absent peaks after glycolation
- Surface area and particle size increase in certain cases
- No significant change in pH
- Variability among concentration of Silica in solution

DISSOLUTION

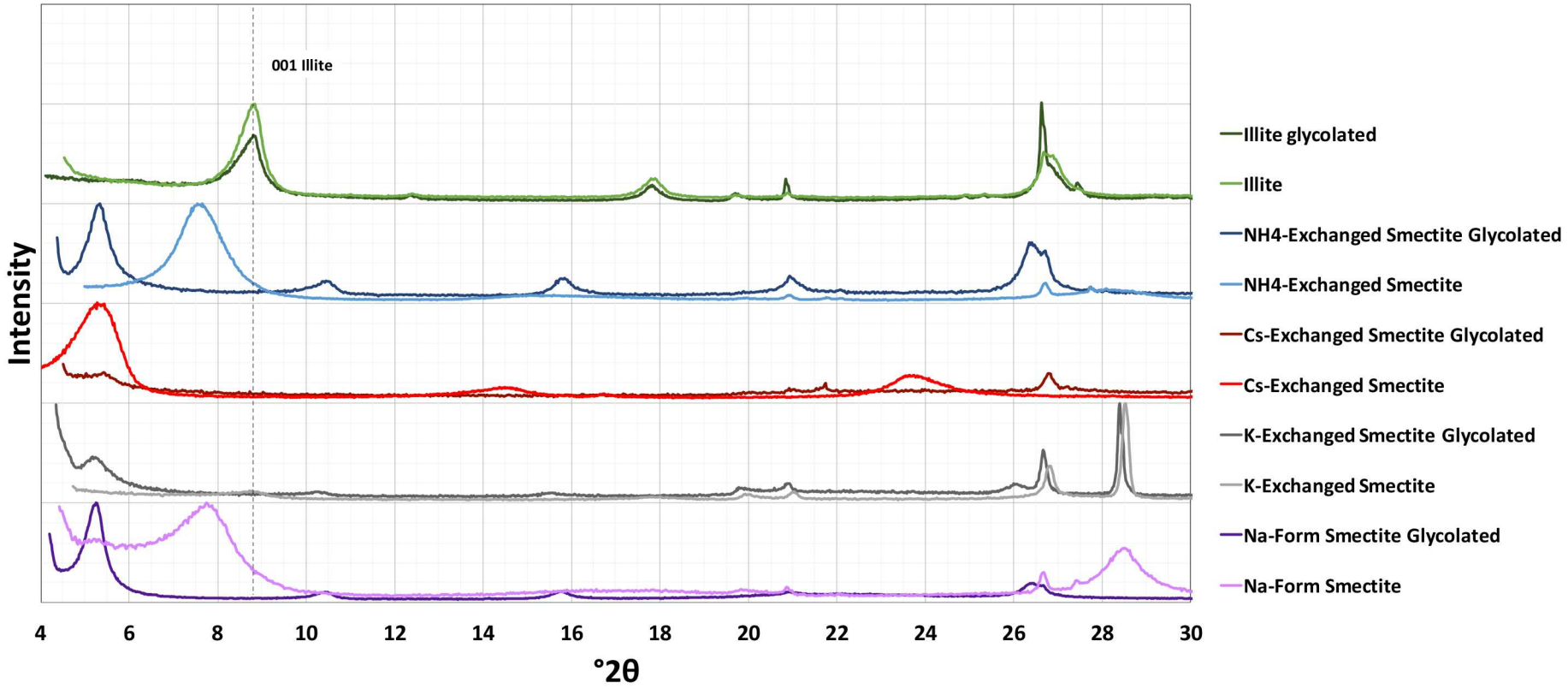




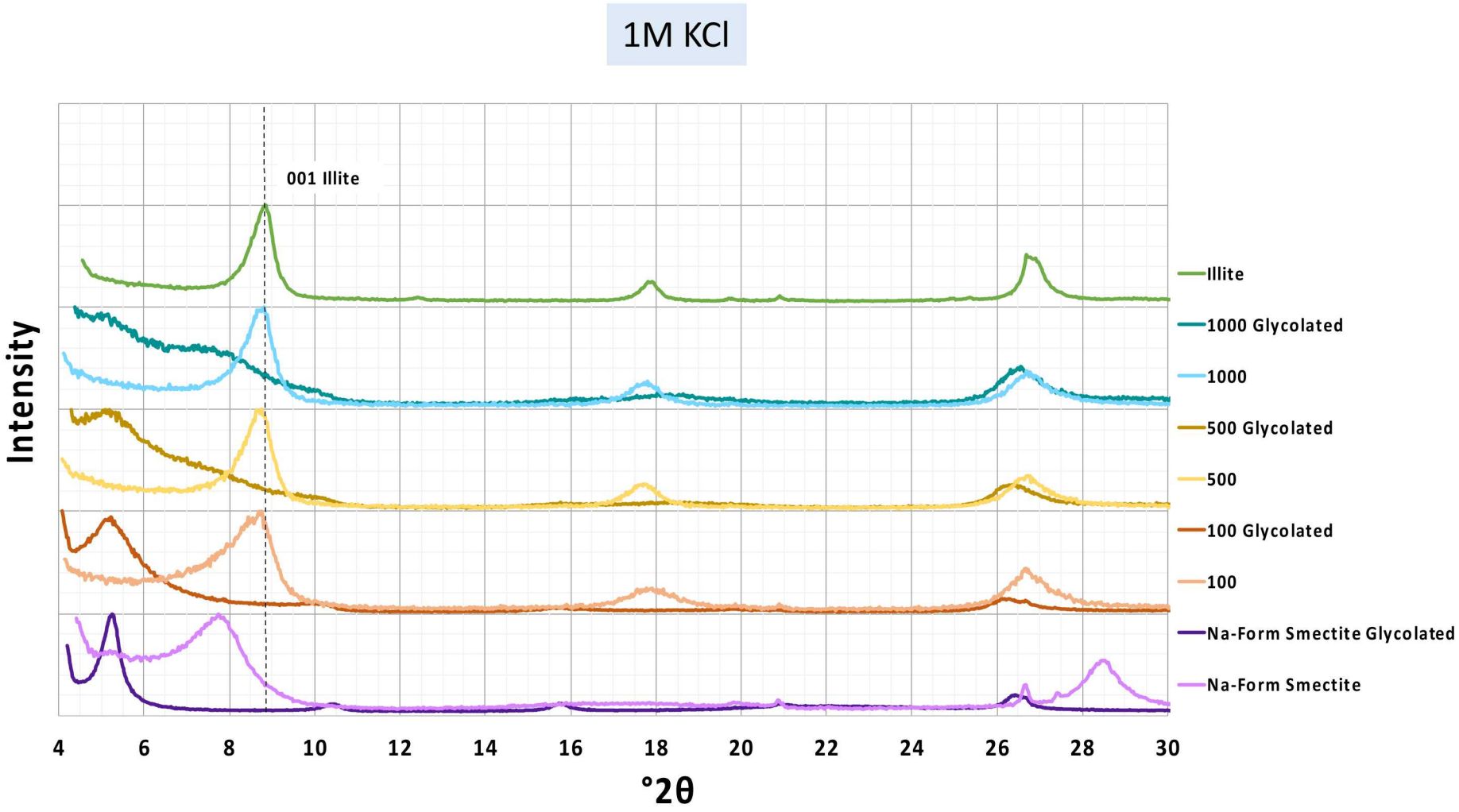
Difficult to get enough sample to perform analyses!



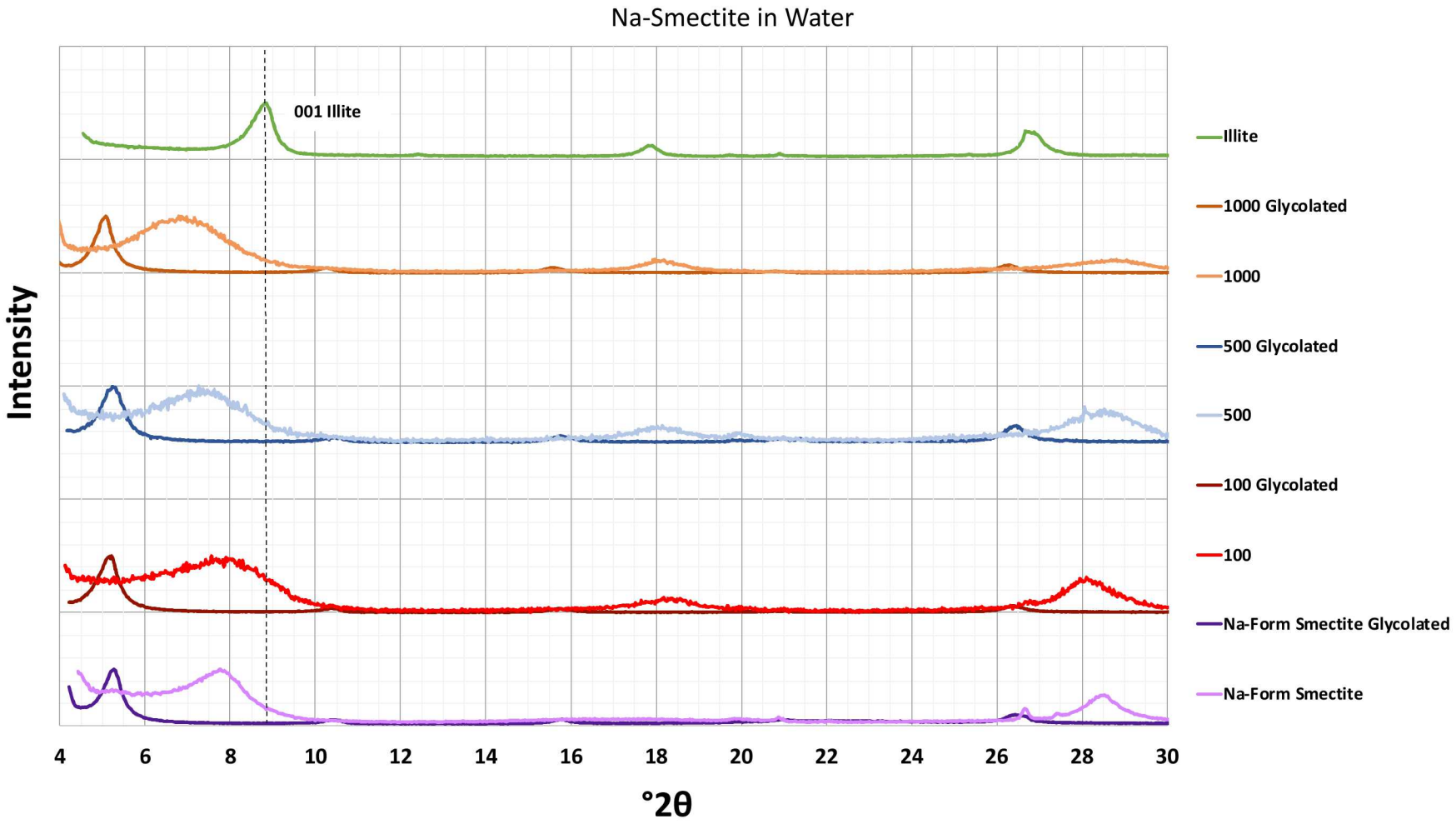
XRD: Starting Material



XRD: Sodium Form Smectite

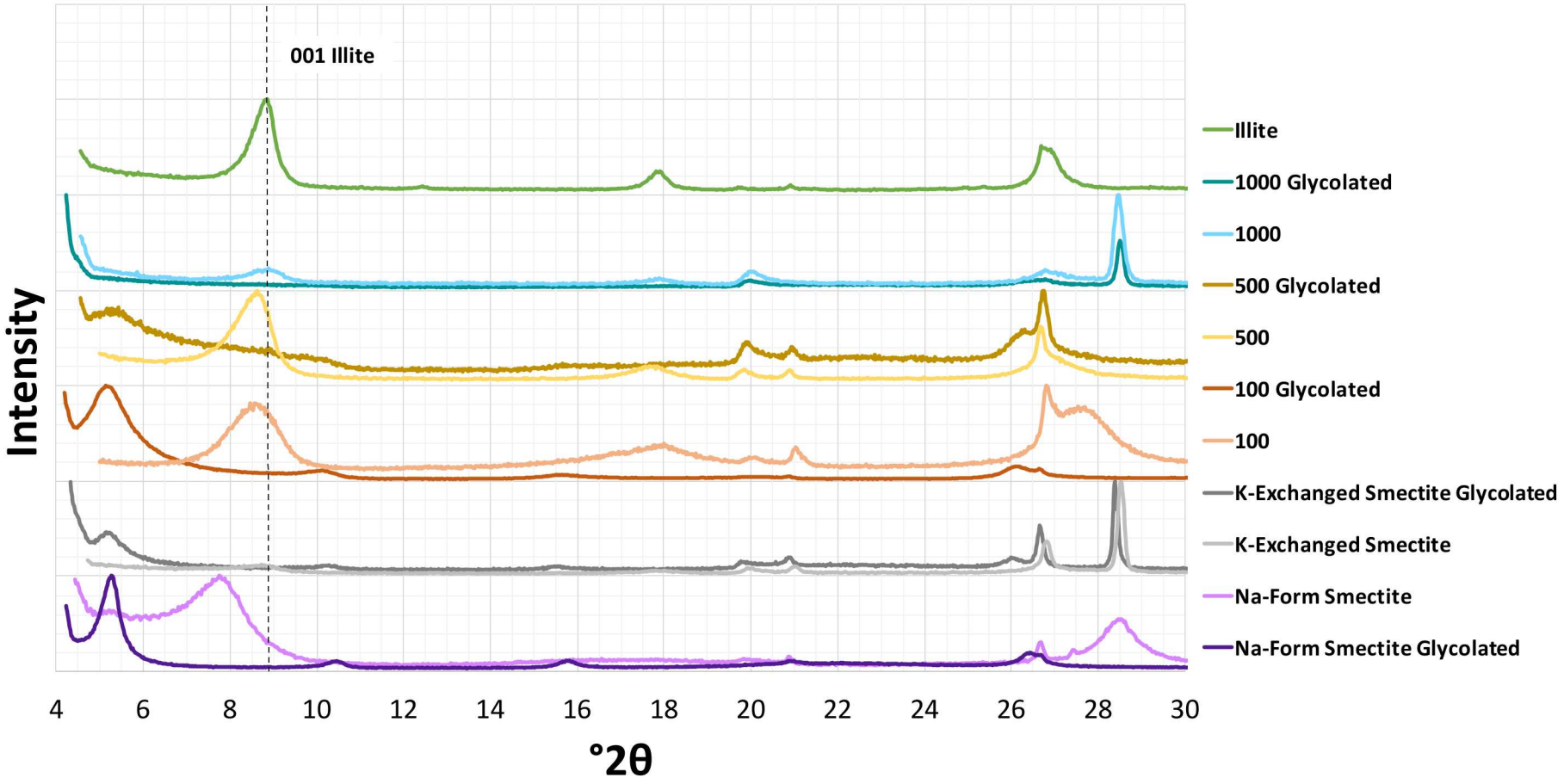


XRD: Sodium form smectite



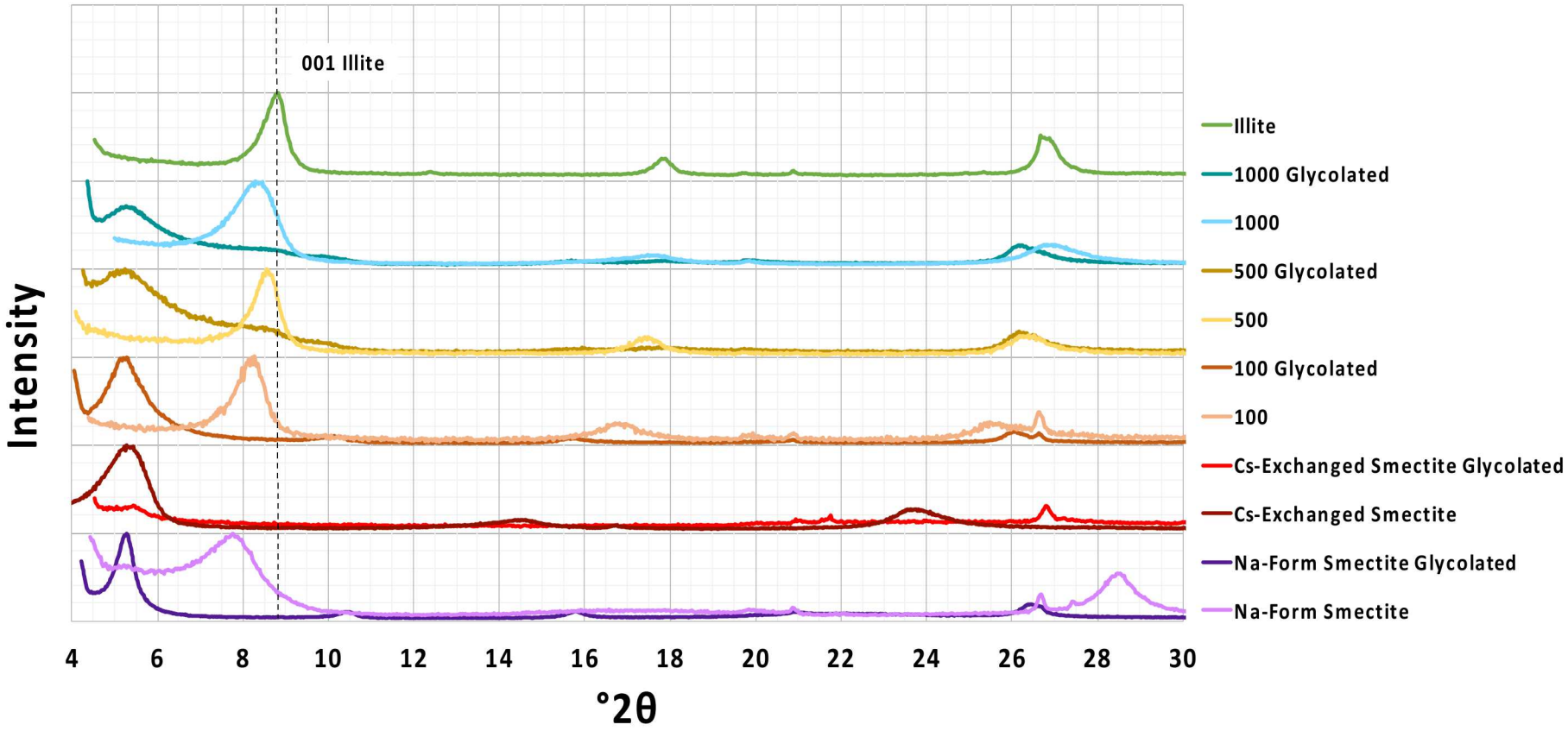
XRD- Potassium Form Smectite

1M KCl

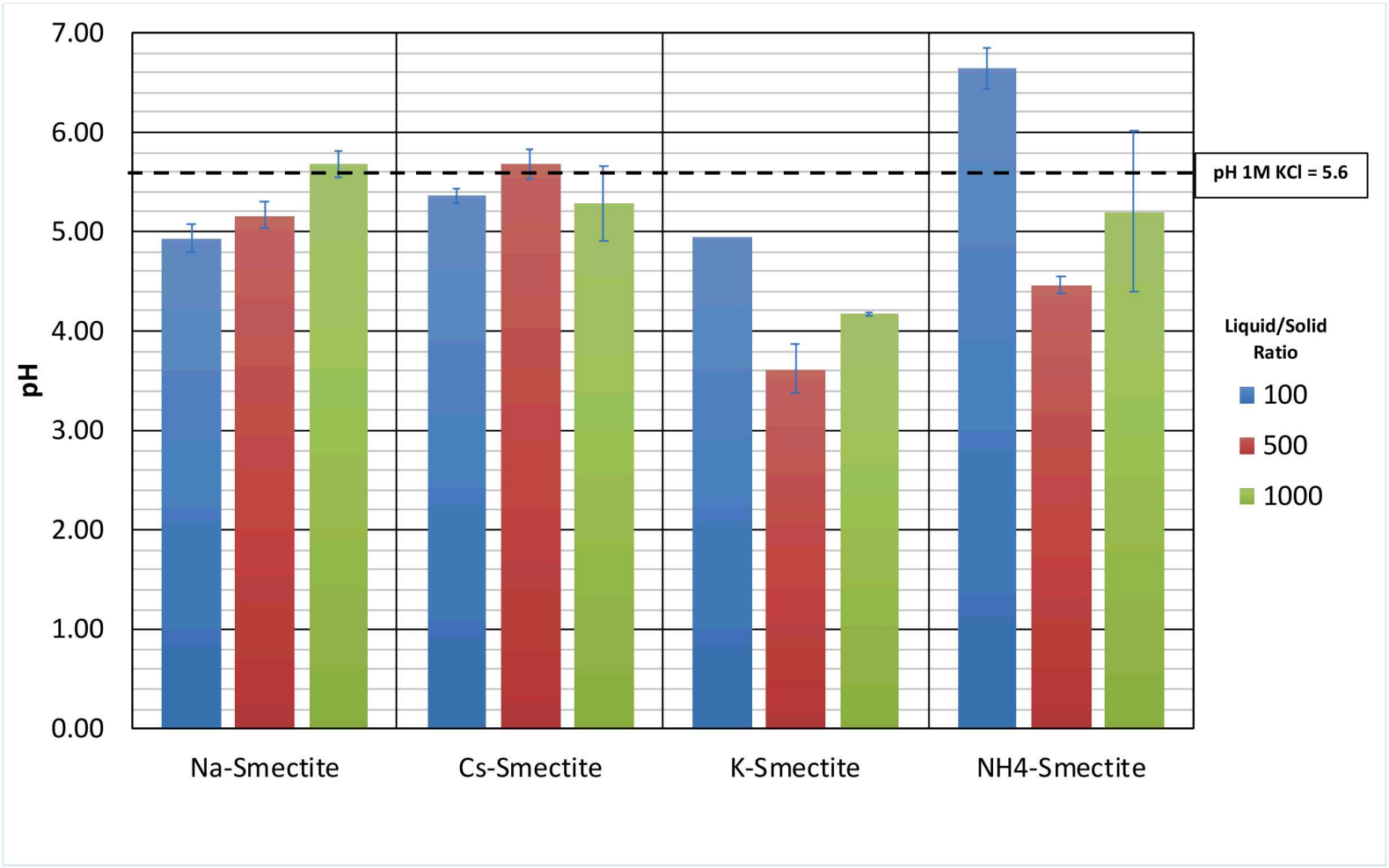


XRD: Cesium Form Smectite

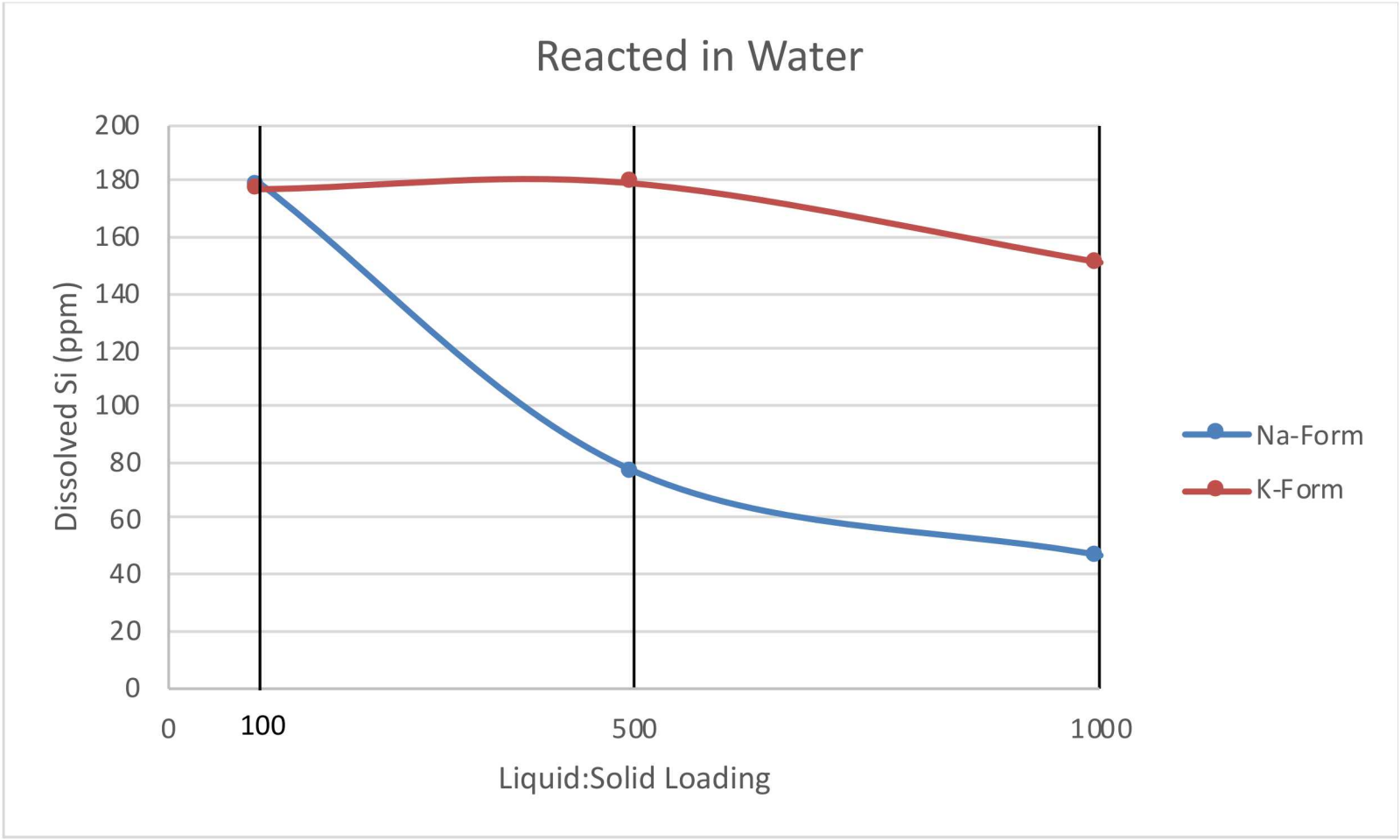
1M KCl



pH Changes: 1M KCL



Dissolved Silica



Remaining Tasks

- Explore longer time scales (2 weeks, 1 month, etc.)
- Continue analysis of solution chemistry
- Continue analysis of composition
- Calculations of %Illite
- Changes in morphology by SEM
- Investigate behavior of iron in reaction
- Addition of mica to possibly escalate conversion
- Addition of quartz to inhibit conversion

-----A great 40 year research project ☺-----

Questions?

SFWD

Spent Fuel & Waste Disposition

Annual Working Group Meeting
May 21-23, 2019

UNLV-SEV – *Las Vegas, Nevada*