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Interactions across interfaces between Ordinary Portland Cement (OPC) paste and oil shale

Goldschmidt, July 2022

SAND#:

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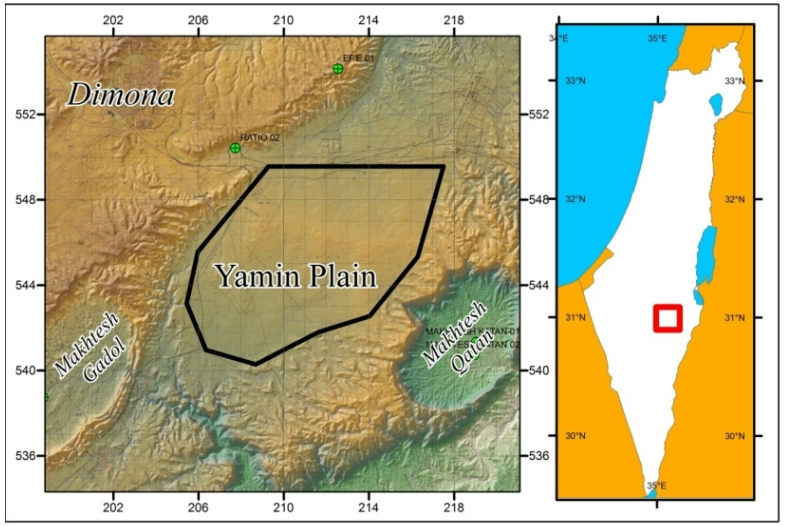
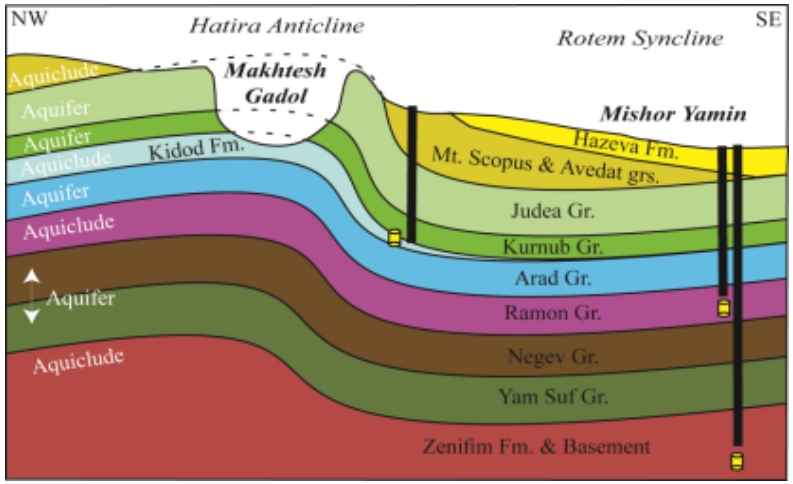
Motivation

- The Israel Atomic Energy Commission is examining the possibility of using Intermediate Depth Borehole (IDB) for radioactive waste disposal storage
- The suggested site for IDB is partially located within oil shale rock formation in the northern Negev, Israel
- Oil shale is not traditionally being used as a host rock for geological waste disposal sites
- A thorough characterization of oil shale – cement interface is needed





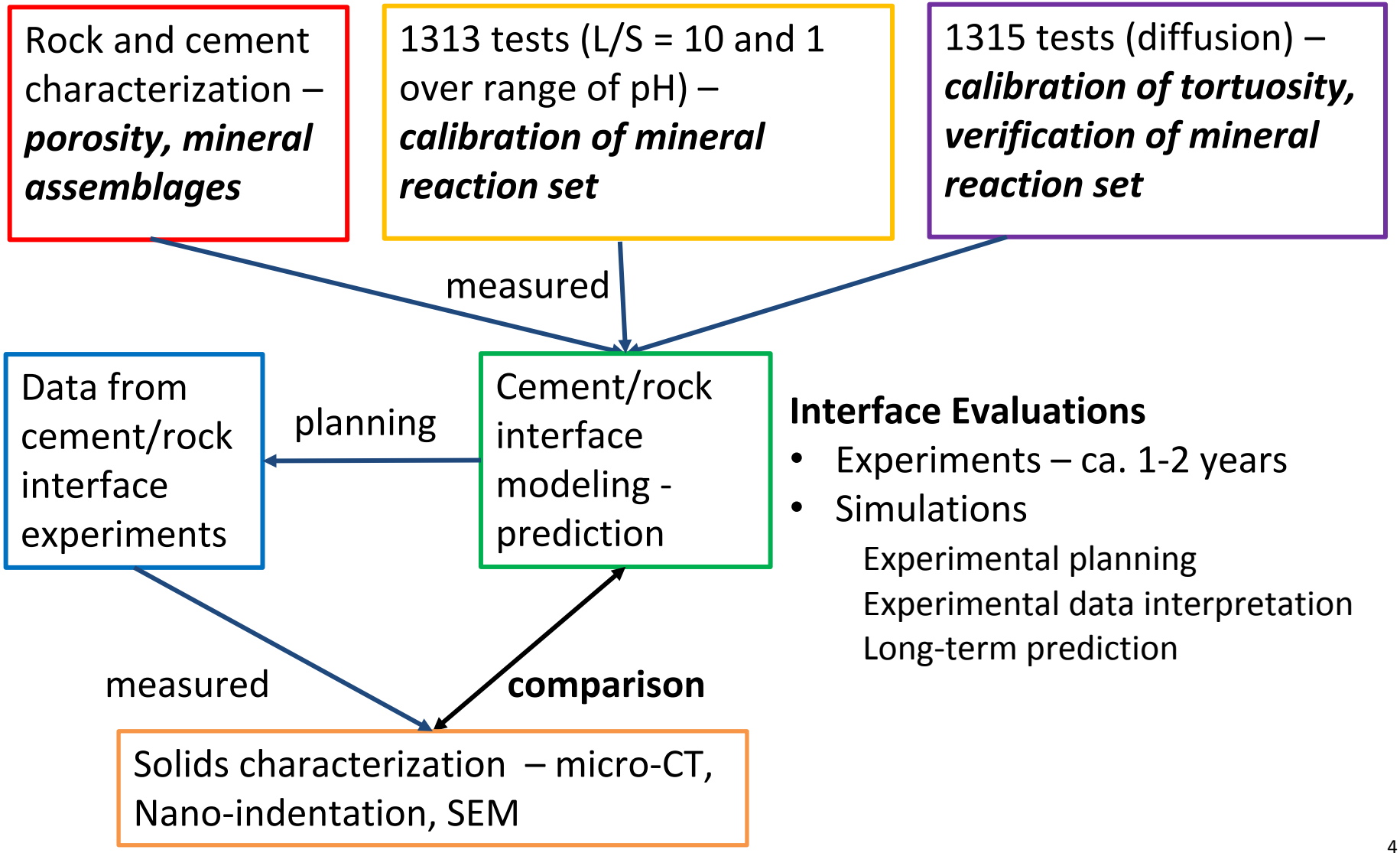
Objectives

Objectives:

i) Use laboratory experiments for chemical and physical characterizations of oil shale and OPC paste (CEM I)

ii) Long-term performance assessment of the interface and validation using laboratory experiments.

Project Approach



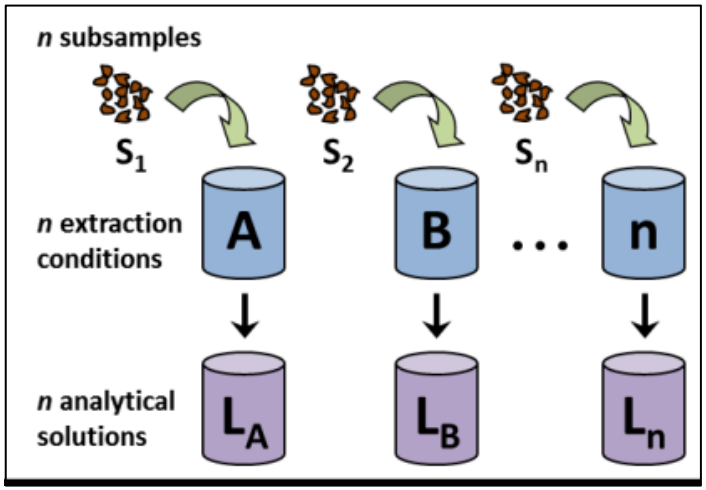
Interface Evaluations

- Experiments – ca. 1-2 years
- Simulations
  - Experimental planning
  - Experimental data interpretation
  - Long-term prediction

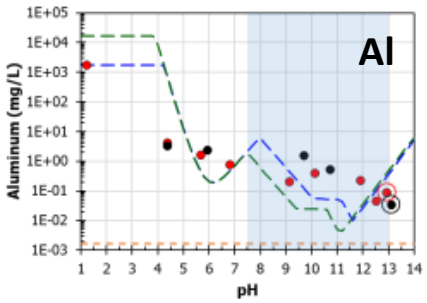
EPA Method 1313 and Mineral Reaction Set Calibration

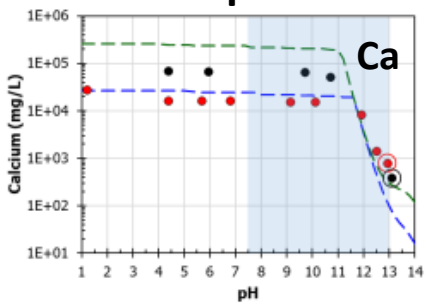
EPA Method 1313

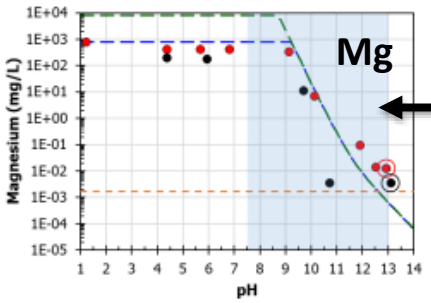
- **pH dependent leaching test**
  - OPC paste and oil shale
  - L/S of 10 and 1
- **Analysis**
  - Extracts analyzed using ICP-OES, ICP-MS, TOC, and IC. Additional measurements include pH, conductivity.



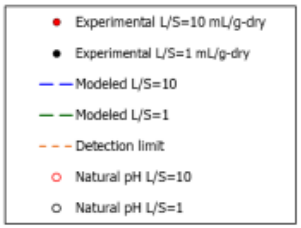
OPC paste

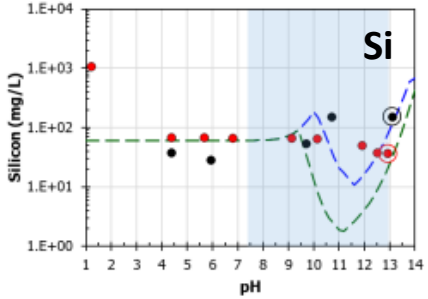


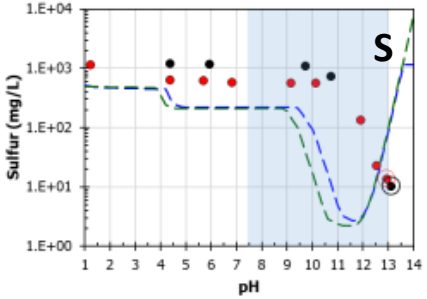




Shaded area represents the expected pH range during interaction between OPC and rocks





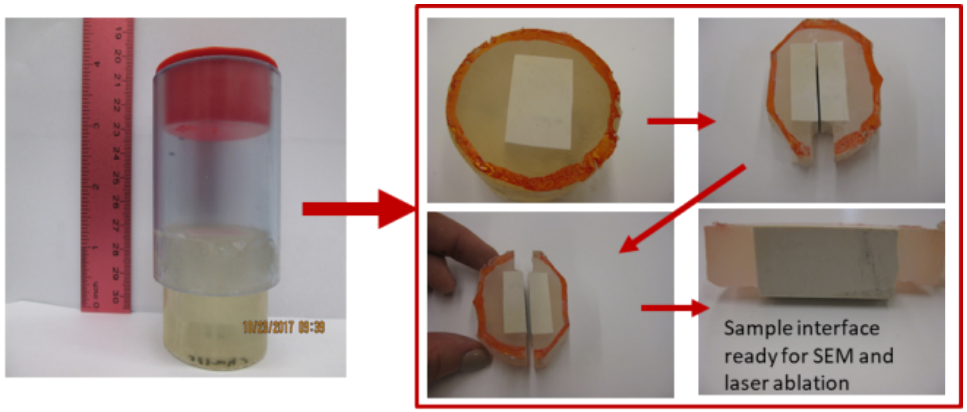


Data from EPA Method 1313

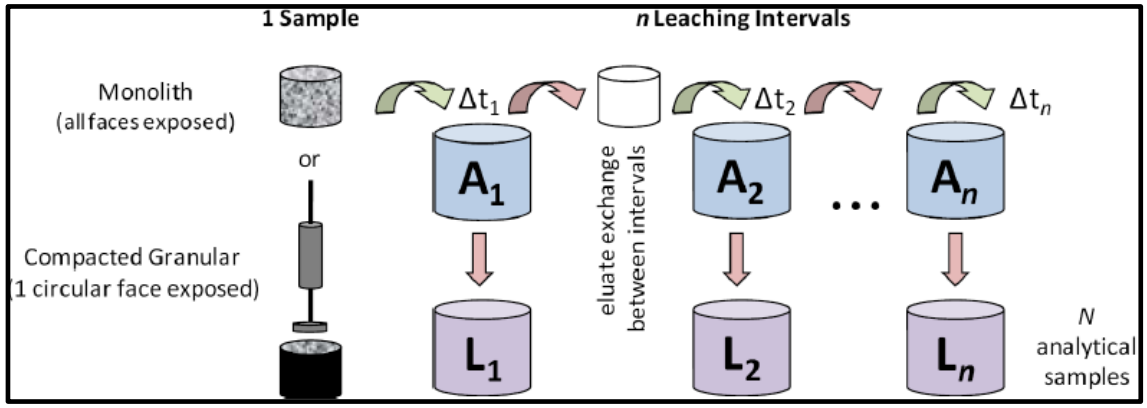
EPA Method 1315 and Conceptual Model of 1315 Test

Method 1315

- Mass transfer rate tank leaching test – design modified for post-test profile characterization



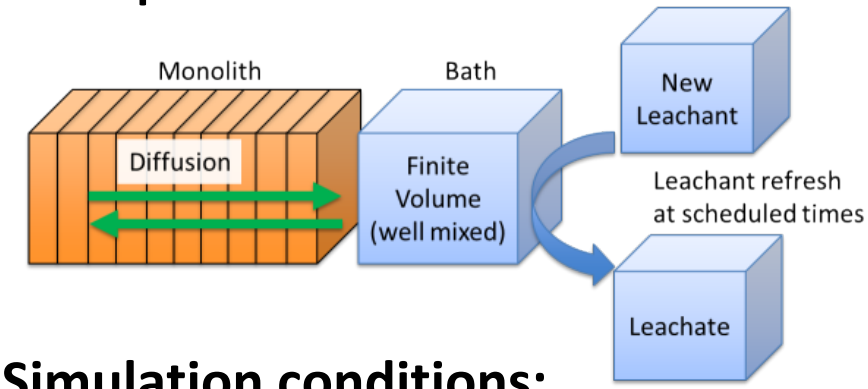
Method 1315 – Experimental set up and sample processing



Tortuosity Calibration

Validation of mineral reaction set calibration
Prediction of 1315 results based on mineral reaction set calibrated by 1313 test

Conceptual model



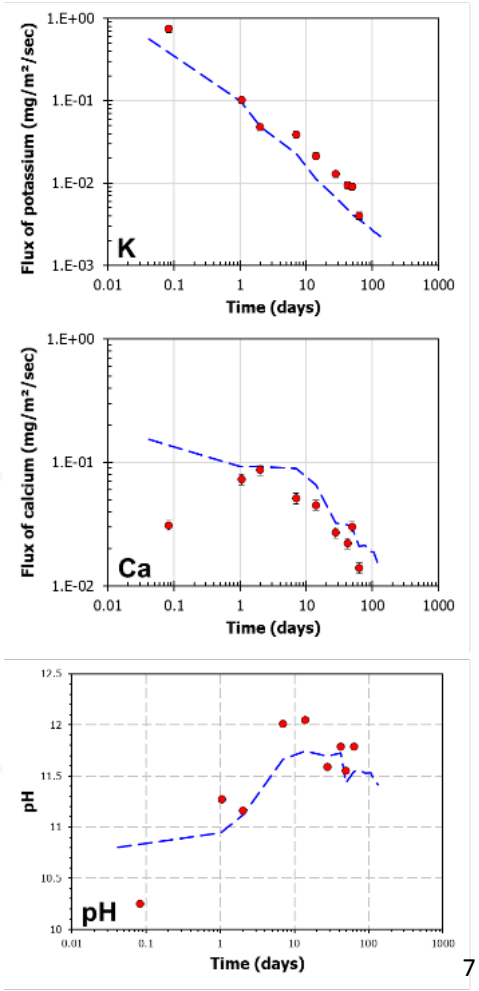
Simulation conditions:

- 133 days simulated to represent 1315 tests duration
- Saturated conditions with measured porosity
- No fluxes at boundaries – diffusion only (multi-ionic diffusion)
- 1-D, 30 cells, each cell is well mixed
- Actual dimensions of monoliths

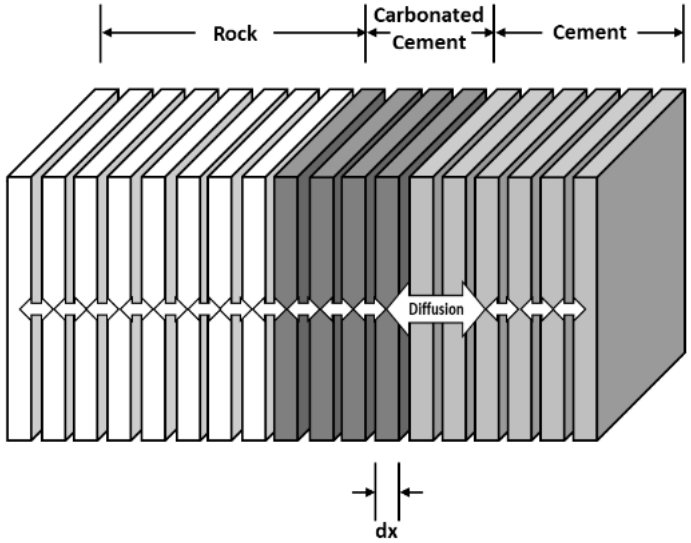
calibration

Validation

OPC paste



Conceptual Model – Rock/Cement Interface



Model assumptions:

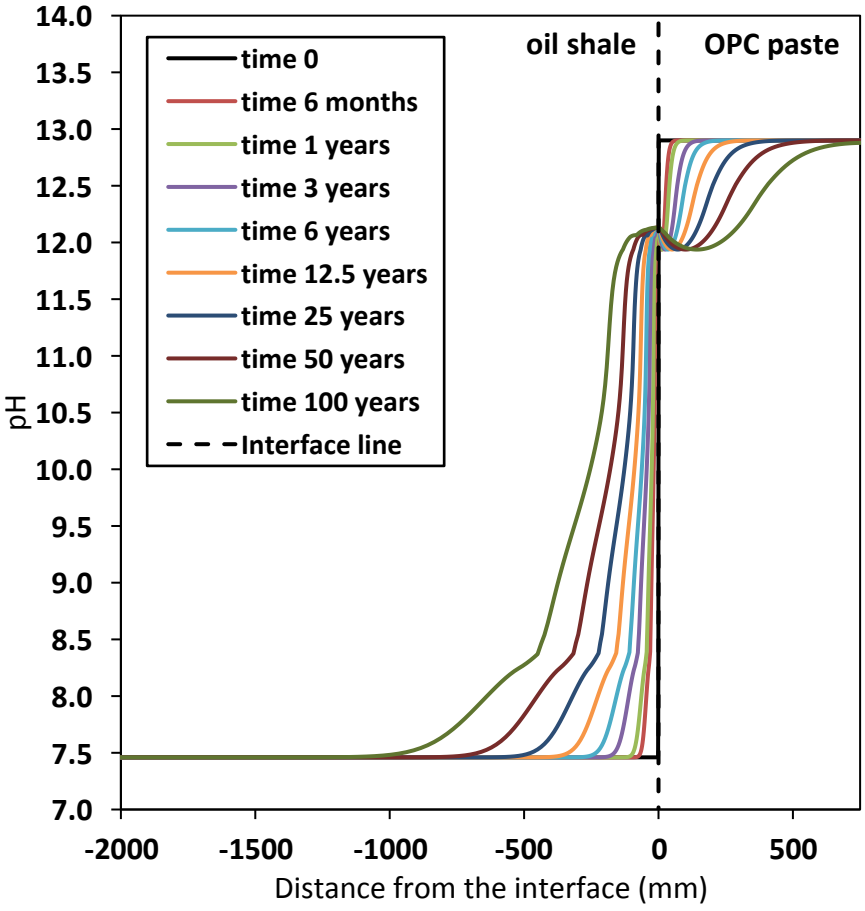
1. Each cell is well mixed
2. Local equilibrium
3. C-(N-)A-S-H solid solutions
4. Multi-ionic diffusion only
5. Materials intact throughout the entire simulation

Model conditions for experimental case:

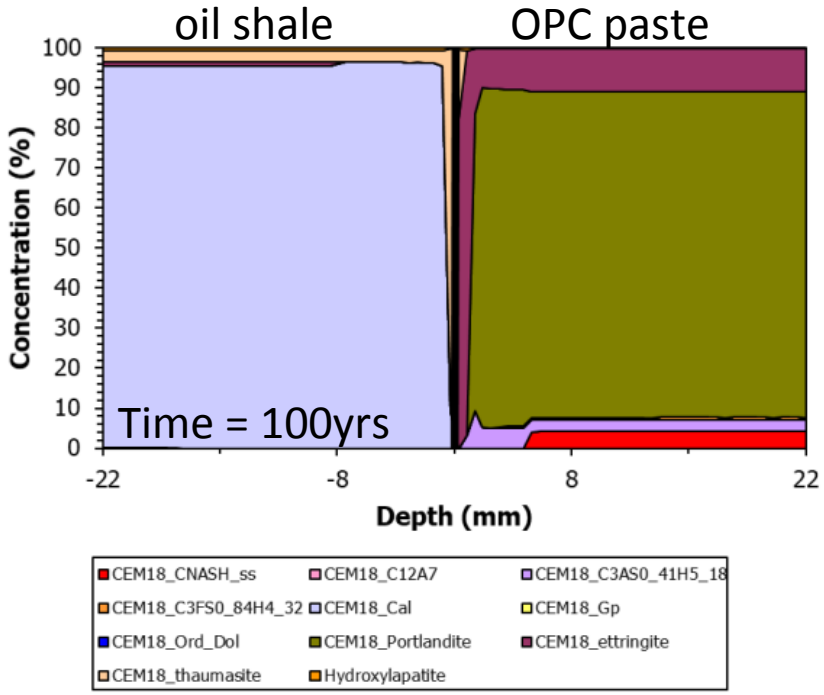
- 100 years simulated, saturated conditions, 30 C
- 1-D, 378 cells, Finite volume
- No fluxes at external boundaries
- Thermodynamic databases – Minteq v4; LLNL, CEMDATA18 (Lothenbach et al. (2018))
- Initial carbonate content – based on 1313 test
- Tortuosity – calibrated values
- Porosity – measured values

Simulation Results

pH profile



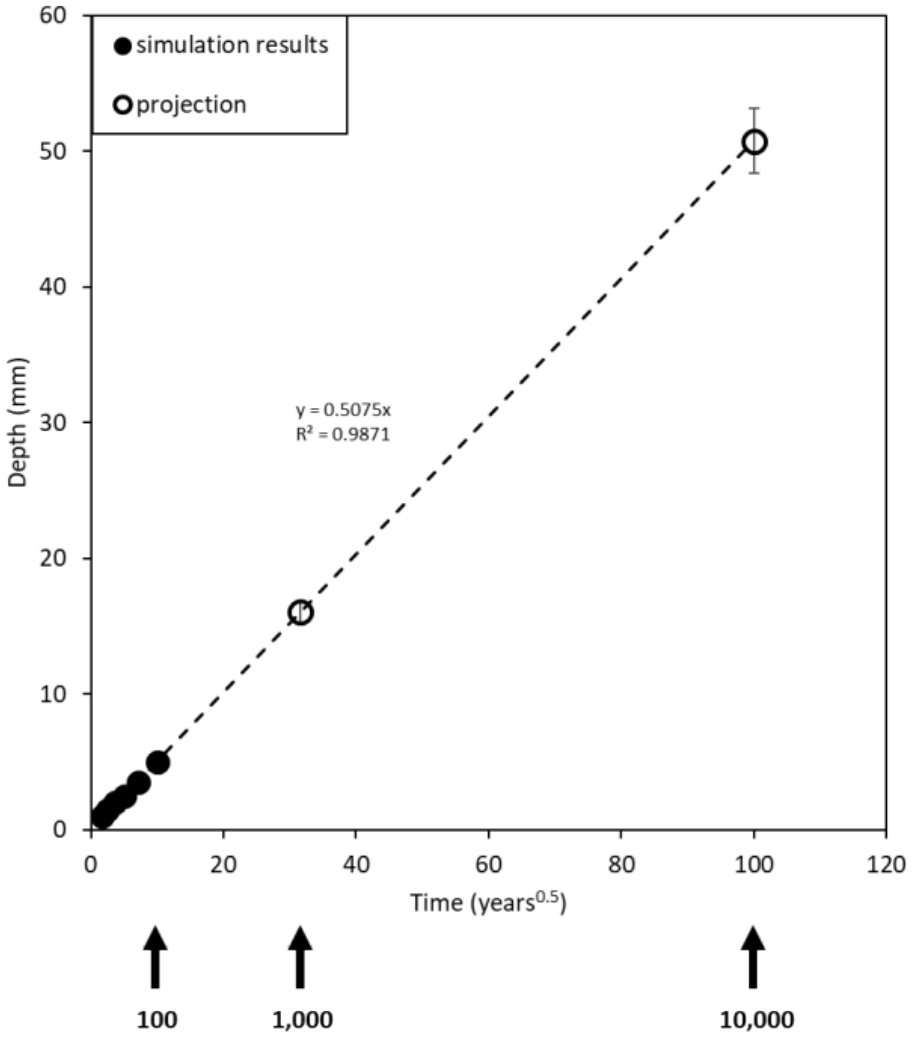
Solid phases distribution profile



Oil shale – OPC paste:

- Complete CSH depletion near interface
- Portlandite depletion
- Ettringite formation
- Thaumsite formation (both sides of interface)

Interface Model Results
Carbonation Front Progress Prediction



The location, X_C , of the moving carbonation front as a function of cement composition and conditions, when the relative humidity is above 50%, is (Papadakis et al., 1989):

$$X_c = A\sqrt{t}$$

X_c - the location, of the moving carbonation front (mm)

A - proportionality constant (mm $yr^{-0.5}$)

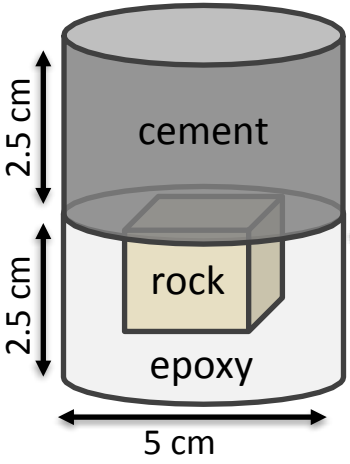
t - time (years)

Long-term scenarios:

- Oil shale-OPC: **16** and **51** mm of OPC are carbonated in 1,000 and 10,000 years

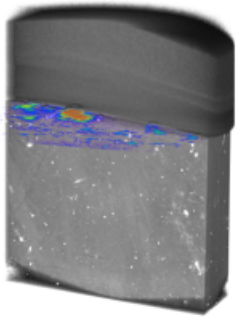
Experimental Project Approach

- Cements were cast on **saturated** rock
- **Curing and aging conditions:** 30 °C and 100% relative humidity

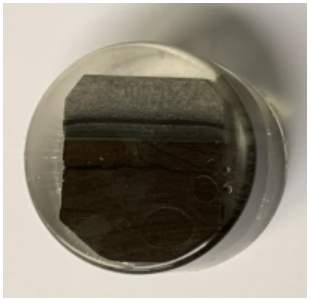


Aging
1-3 yr

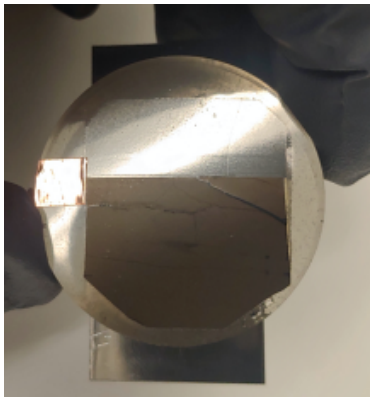
MicroCT



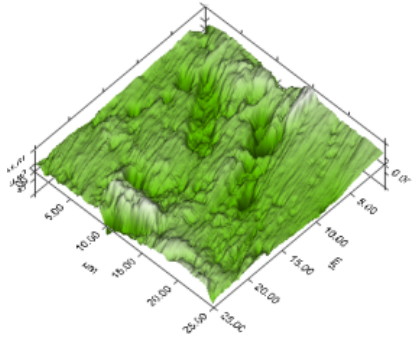
Sectioning



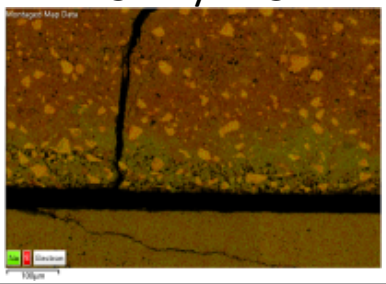
Polishing



Surface Roughness (Triboindenter)

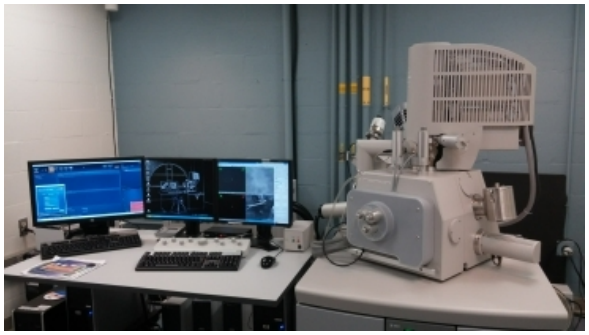


SEM/EDS



Micro/Nanoindentation & Fiducial Placement
micro – rocks + cement
nano – cement

SEM/EDS



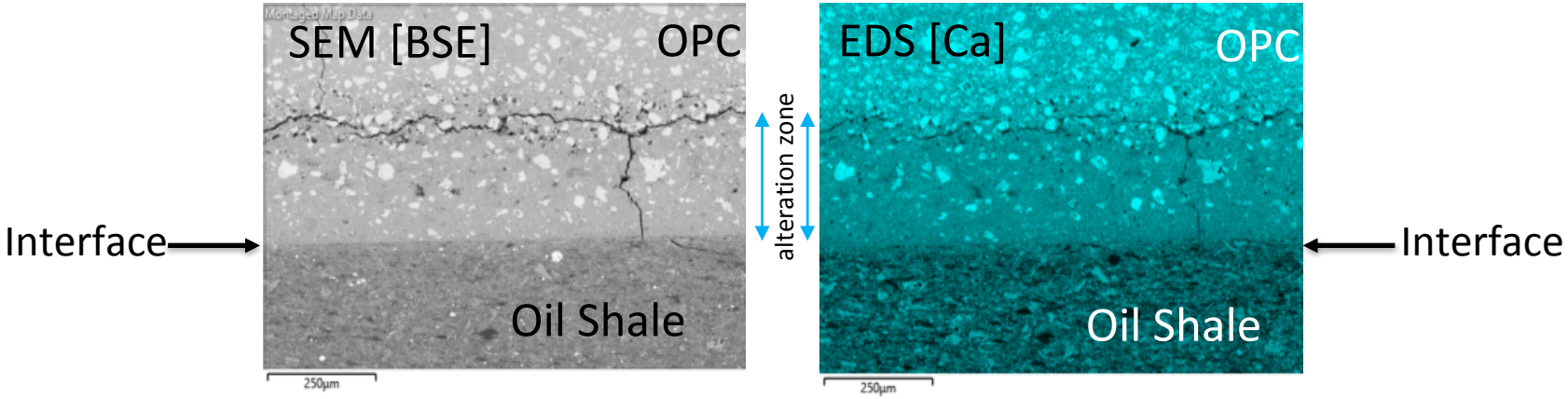
- Large area mapping utilized to investigate microstructure, morphology, and chemistry (major elemental concentrations)

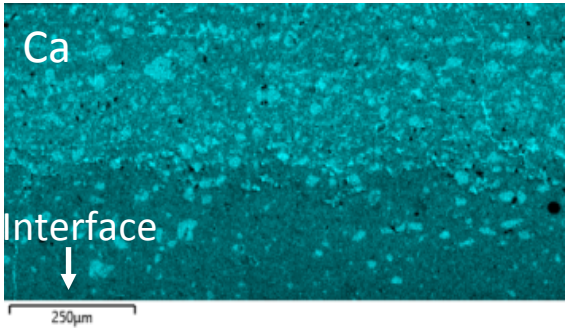
ESEM imaging conditions:

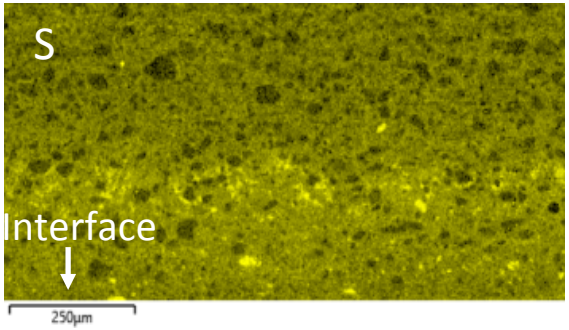
15kV, spot size 3.5, 130 Pa, 600x, pixel dwell time 30µs, resolution 1024x874

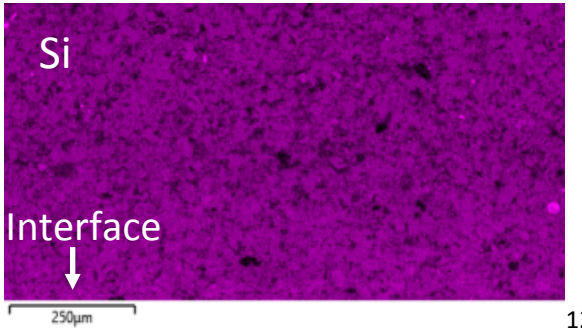
EDS collection conditions:

pixel dwell time 30µs, frame count 20, process time 5, resolution 1024x874

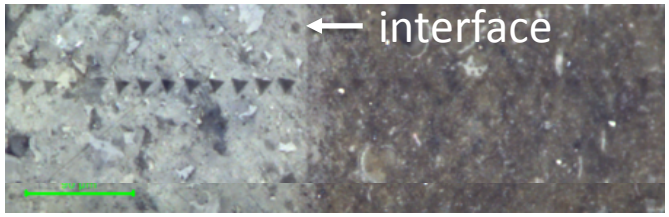








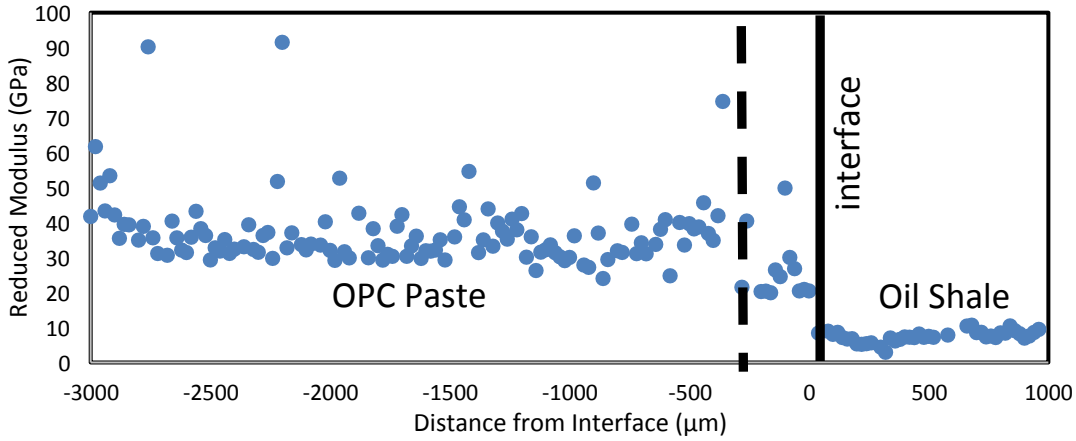
Microindentation

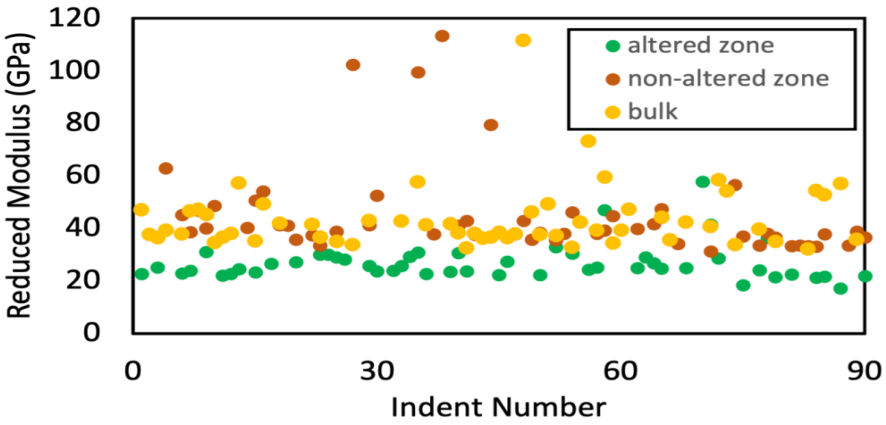


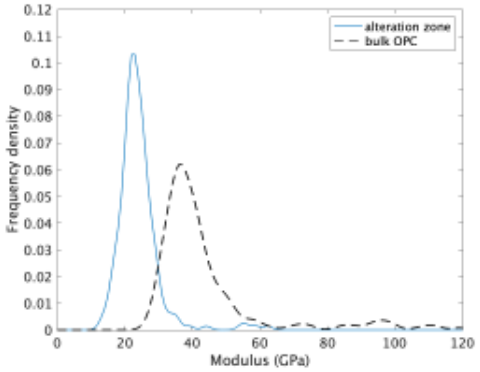
Triboindenter Conditions

Berkovich tip
peak load 75 mN
10-15-10 sec – trapezoidal load function

Reduced Modulus from **Cement to Oil Shale** (constant force)







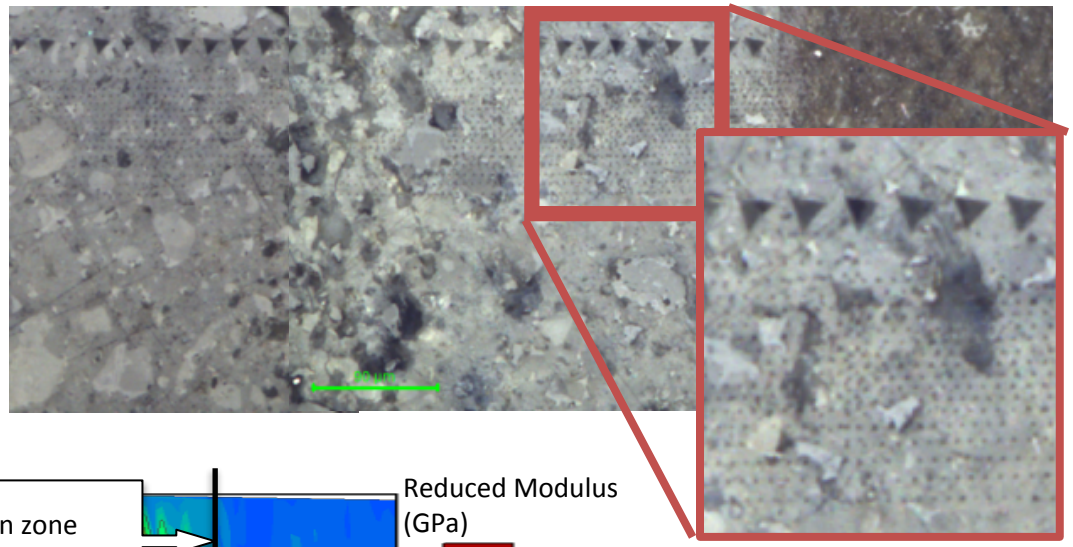
- Within 300 µm of the interface there is a 30% reduction in average moduli of the OPC paste

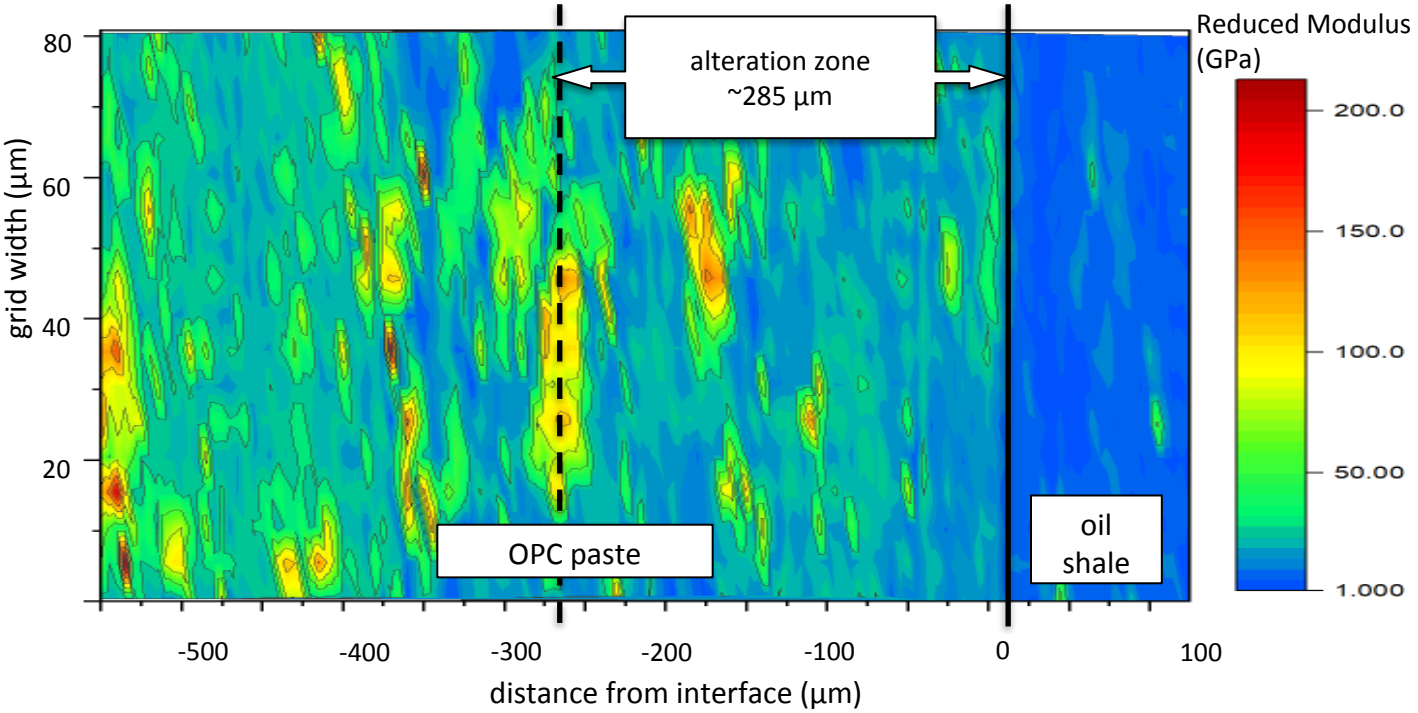
Accelerated Property Mapping (XPM)

- **Series of square grids (85 x 85 µm) crossing over interface region**

Triboindenter Conditions

Berkovich tip
peak load 2 mN
3-5-3 sec – trapezoidal load function





- Nanoscale tip shows gradually reducing modulus values in a nearly identical alteration zone as the micro-indentation data

From Simulations → Experiments

After 1 year of exposure:

- At the interface between OPC paste and oil shale, based on simulation results, an alteration zone of 300 µm is estimated
- In laboratory experiments for the same interface, chemical data shows elemental gradients in Ca, Si, Al, and S, forming an alteration zone of ~300 µm
- Similarly, mechanical data suggests reduced moduli values in OPC paste up to a distance of 300 µm ± 20 µm from the interface
- At the interface between OPC paste and limestone, experiments confirm simulation data showing no chemical or mechanical alteration zone

Oil shale – OPC paste interface		
SEM-EDS (measured chemistry)	Mechanical properties (reduced moduli)	Simulation results
300 µm	300 µm	300 µm

Acknowledgments

- *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.*

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