Colloid-Facilitated Transport of Contaminants through the Vadose Zone

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Introduction

- Hazardous waste stored in the underground tanks has leaked into the vadose zone (more than 1 million curies of radioactivity Cs-137).

- Radionuclides have been detected at much deeper depth than predicted (Cs was found at $\geq$ 40m). This could result from preferential flow, overprediction of Cs sorption, or colloid-enhanced transport.

- The tanks show extreme conditions of high pH, ionic strength and Al concentration. A considerable amount of Si could be released from the sediments beneath the tanks.

Hypothesis

- There is a strong potential for *in situ* formation of colloids as leaking tank waste enters the vadose zone.

Si (sediments) + Al (waste tank) + NaOH (waste tank)

↓

Aluminosilicates: e.g. feldspathoids, zeolites

Objectives

- Determine whether and, if so, what colloids are formed from reaction between simulated tank waste and Hanford sediments.
- Determine how colloid formation changes with temperature and tank waste composition.
- Determine colloid characteristics that govern their mobility through the vadose zone.
- Determine Cs and colloid transport through Hanford sediments.

Materials and Methods

1. Colloid formation and characterization
2. Cesium sorption
3. Colloid, Cesium, and colloid-facilitated Cesium transport

Materials and Methods

Batch experiments:

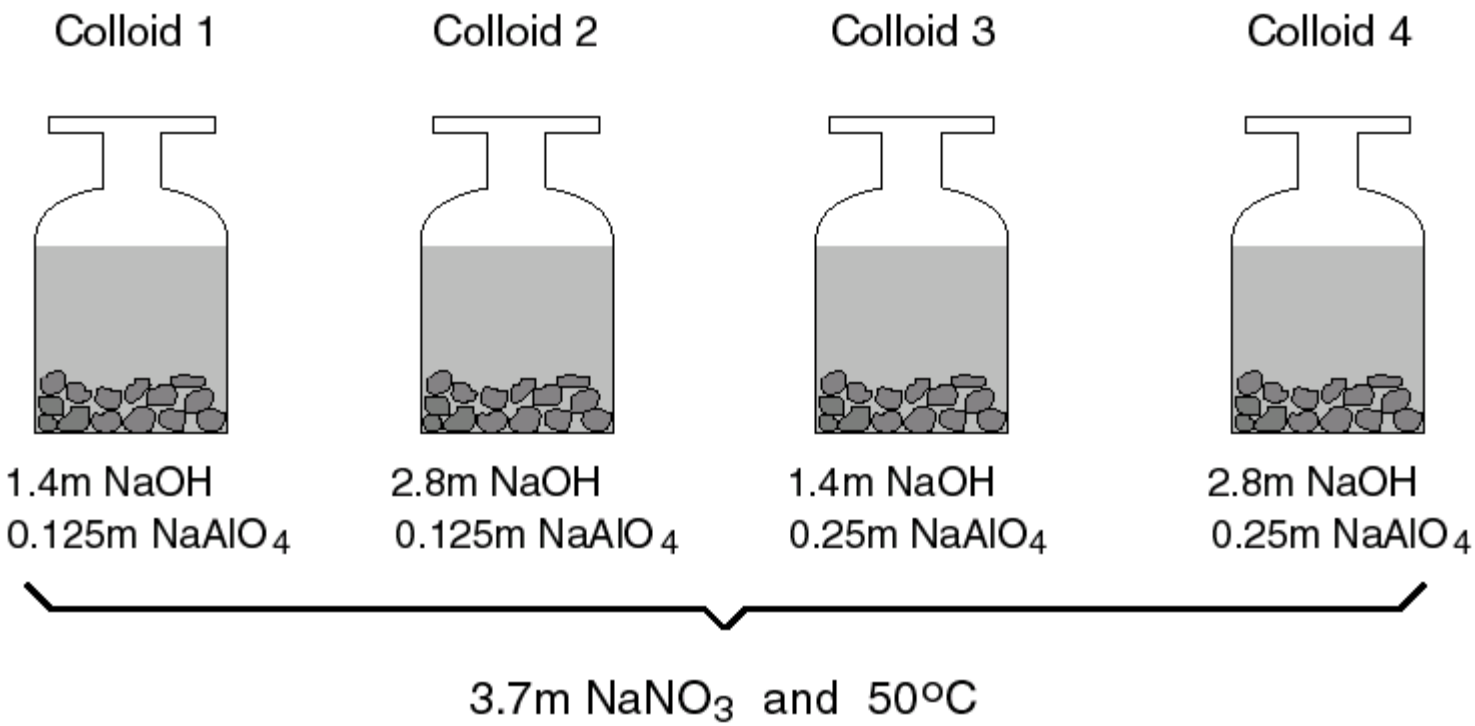
1. Reacted simulated tank waste (STW) with Hanford Formation sediments (1:1) at 50°C for 25 days and 40 days.
2. Reacted simulated tank waste (STW) compositions with 0.036 *M* Si (tetraethyl orthosilicate) at 50°C for 25 days.

Materials and Methods

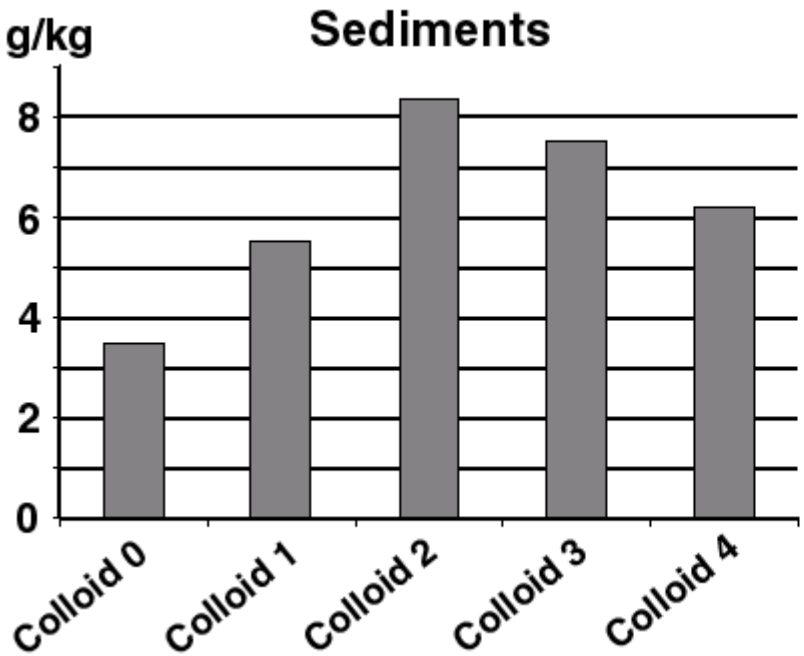
Column experiments:

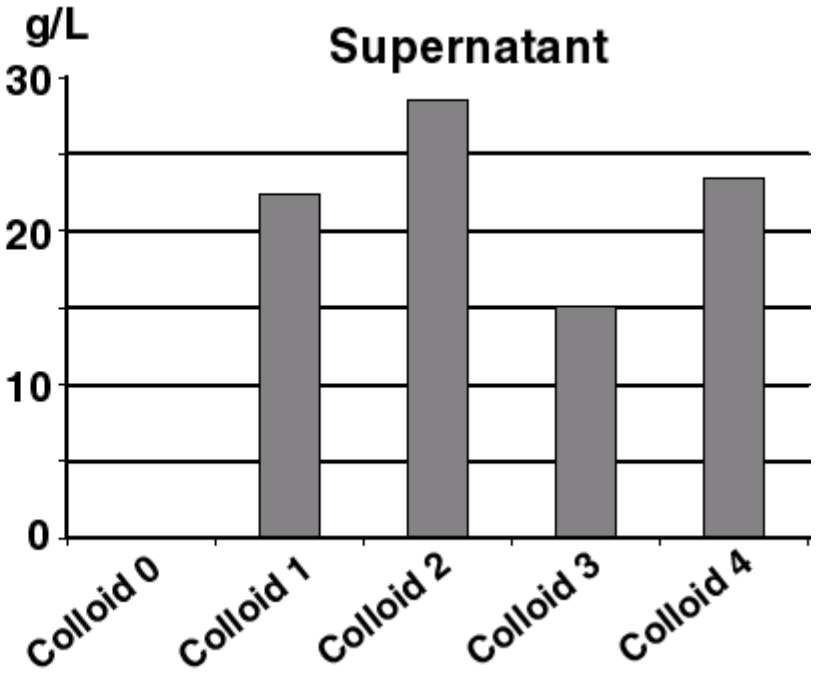
1. In situ colloid mobilization under saturated conditions.
2. Colloid and colloid-facilitated transport as a function of ionic strength and water saturation.

Simulated Tank Waste



Colloid Formation

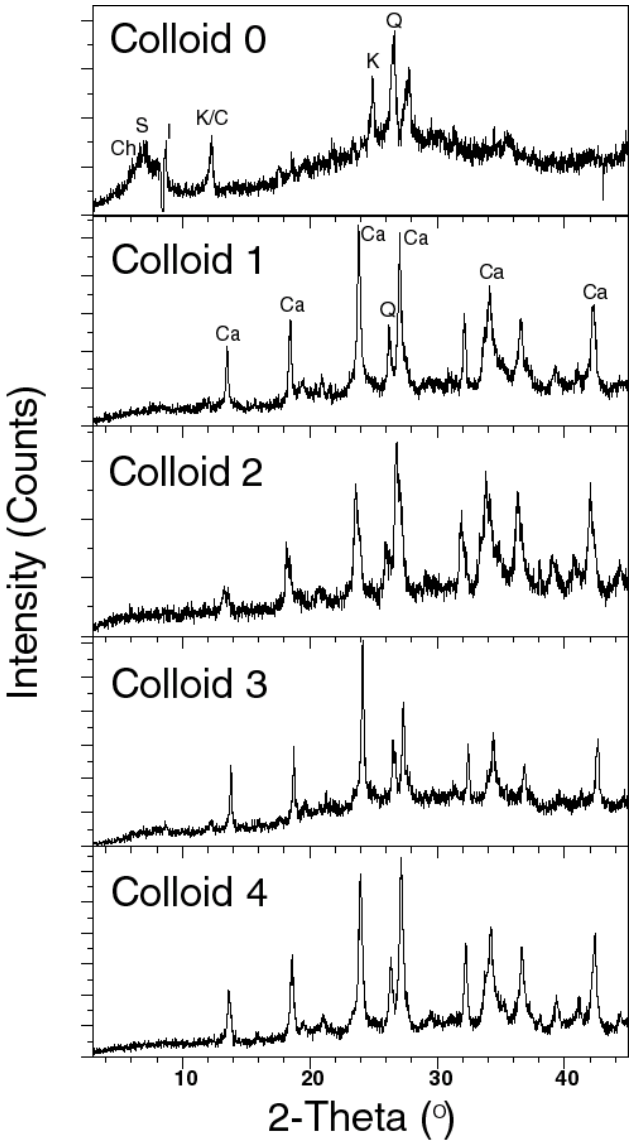




Characterization

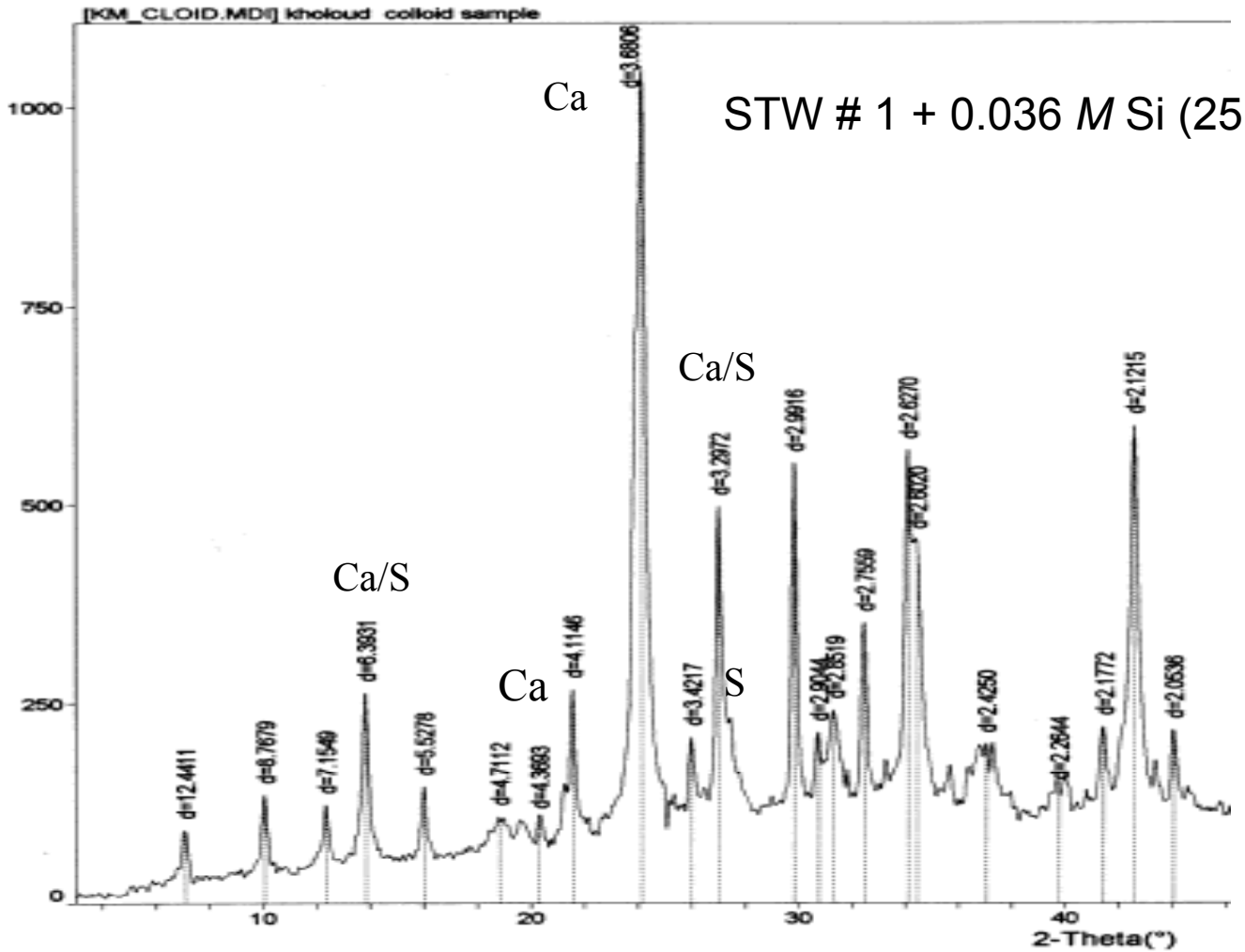
- Electrophoretic mobility
- Morphology (SEM, TEM)
- Mineralogy (XRD, EDAX, FTIR)

XRD Patterns

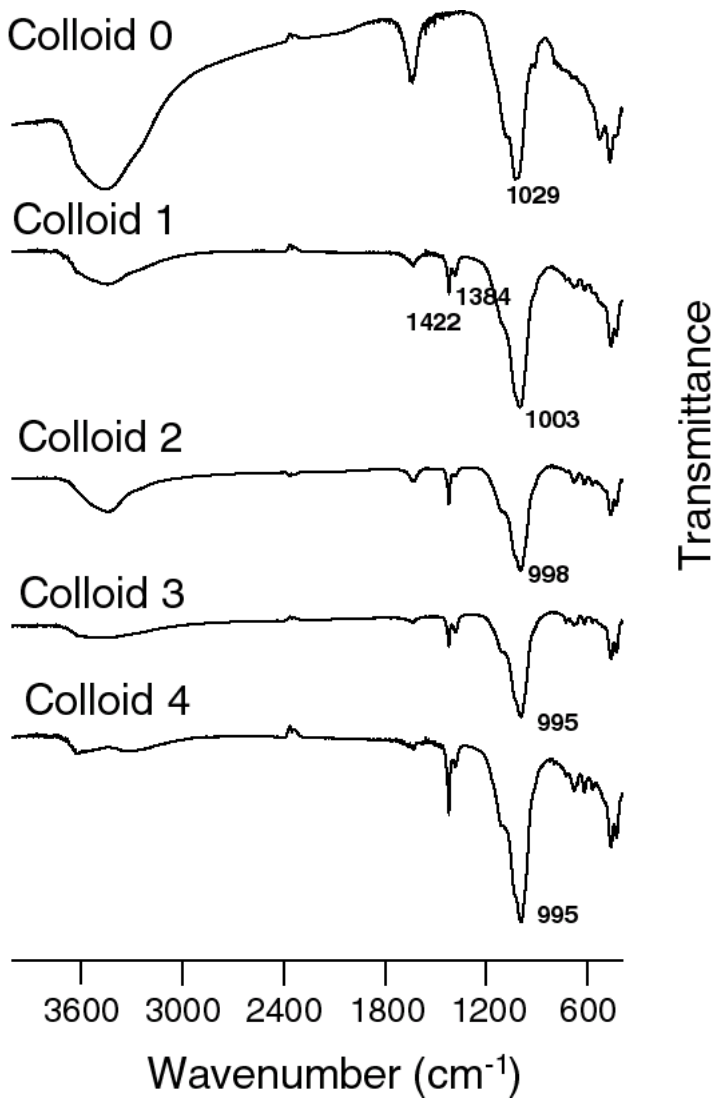


XRD Patterns

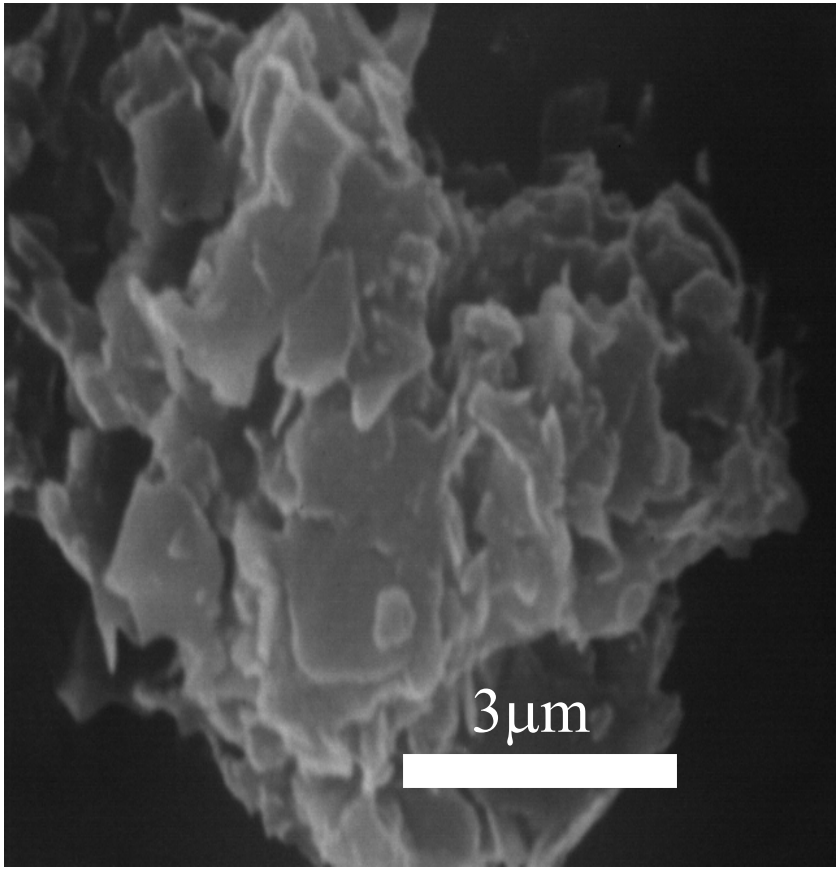
Intensity (Counts)

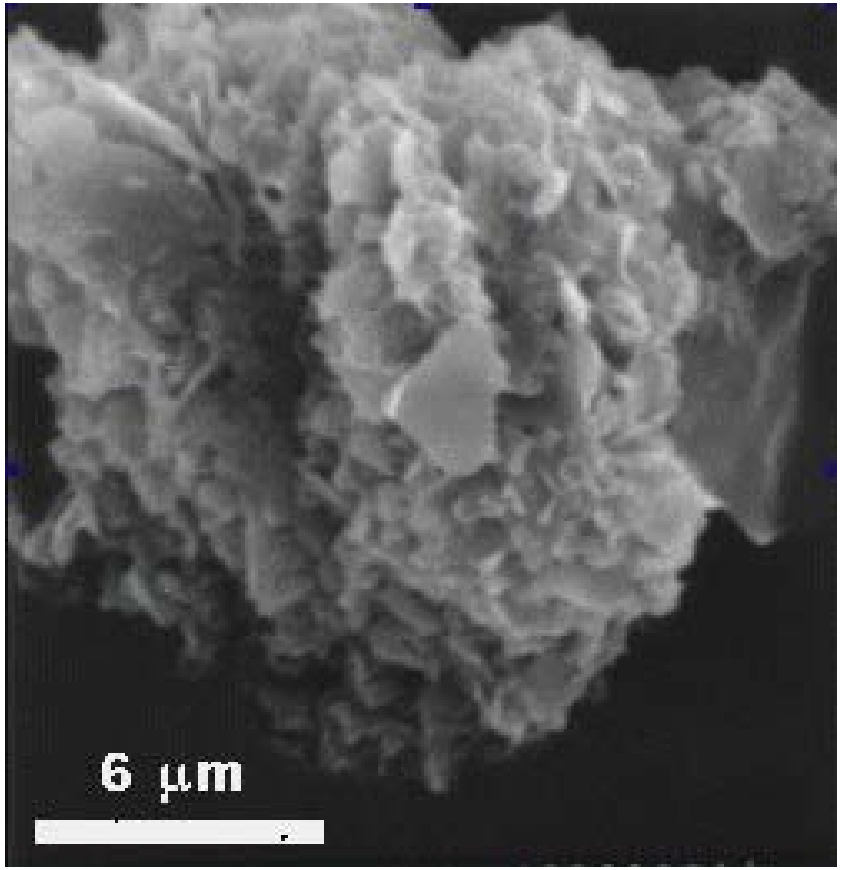


FTIR Spectra

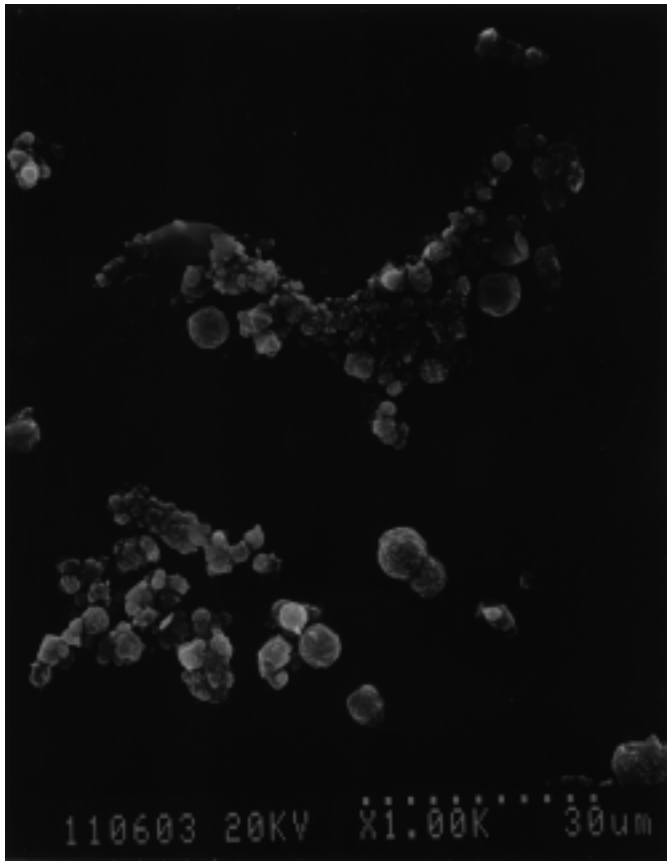


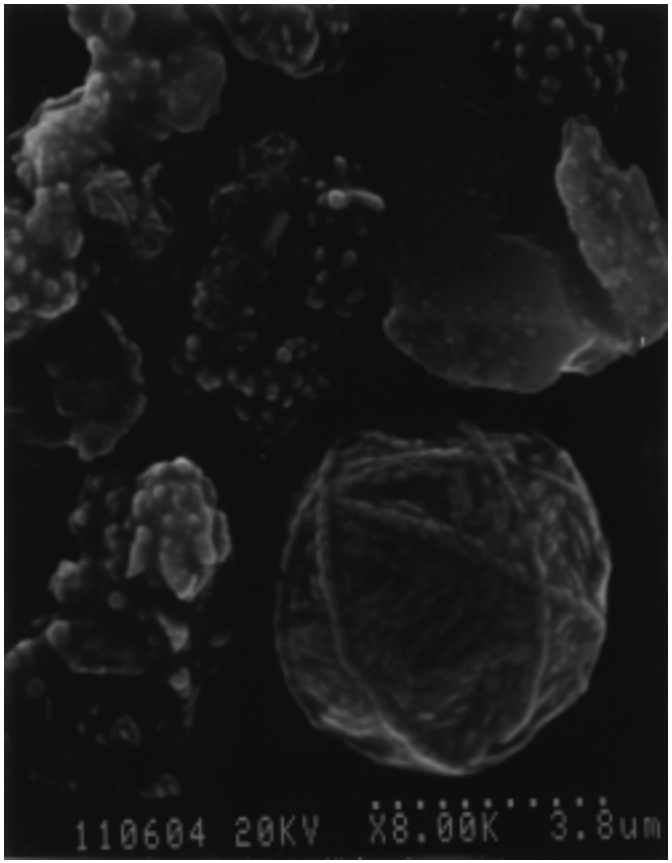
Untreated vs Treated Sediments





Treated Hanford Sediments





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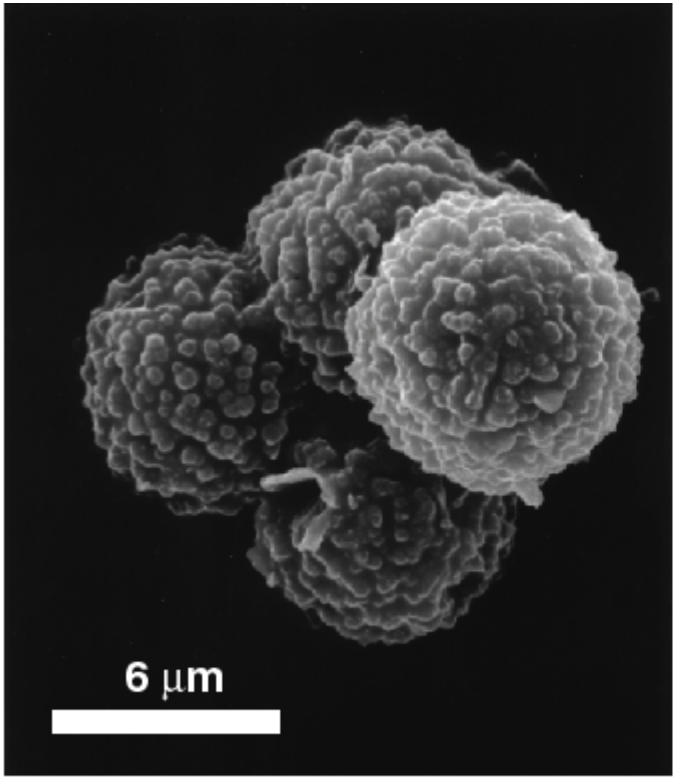
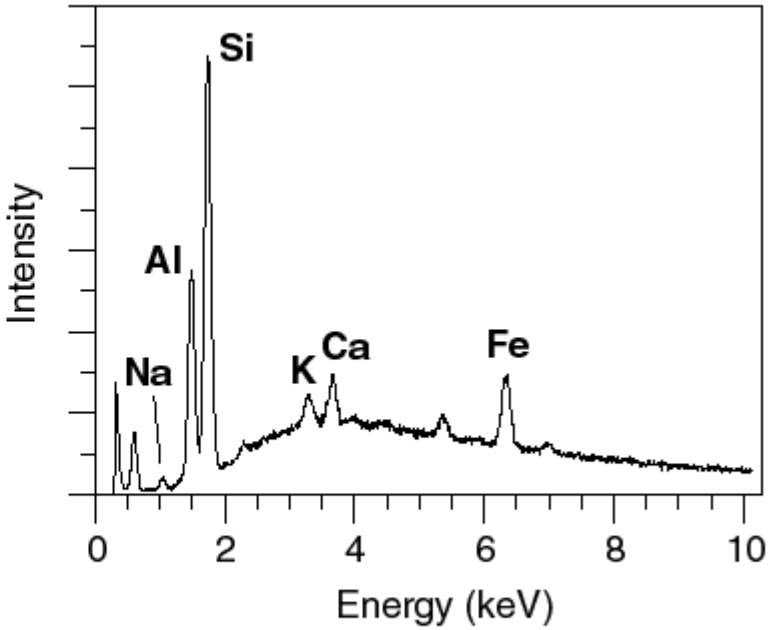
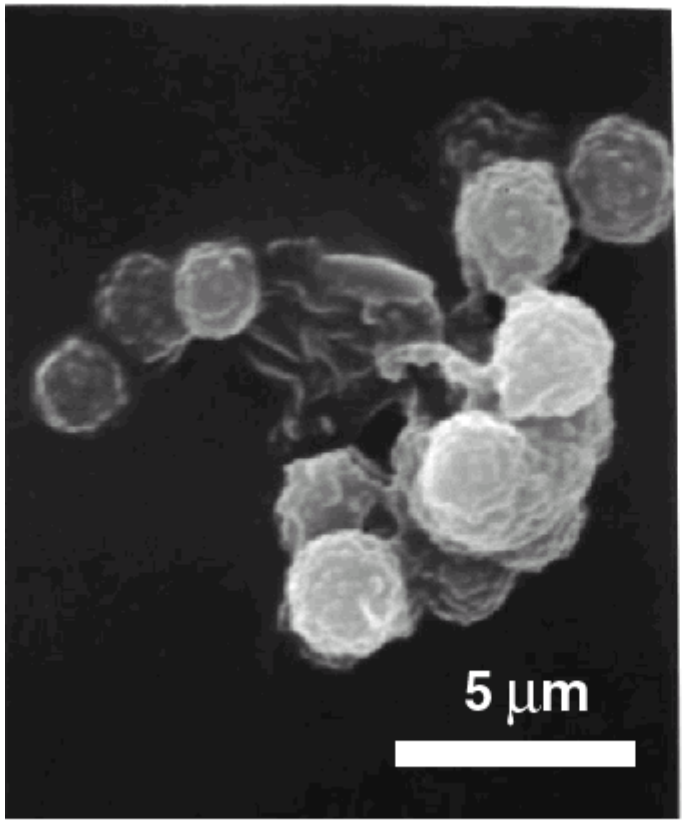
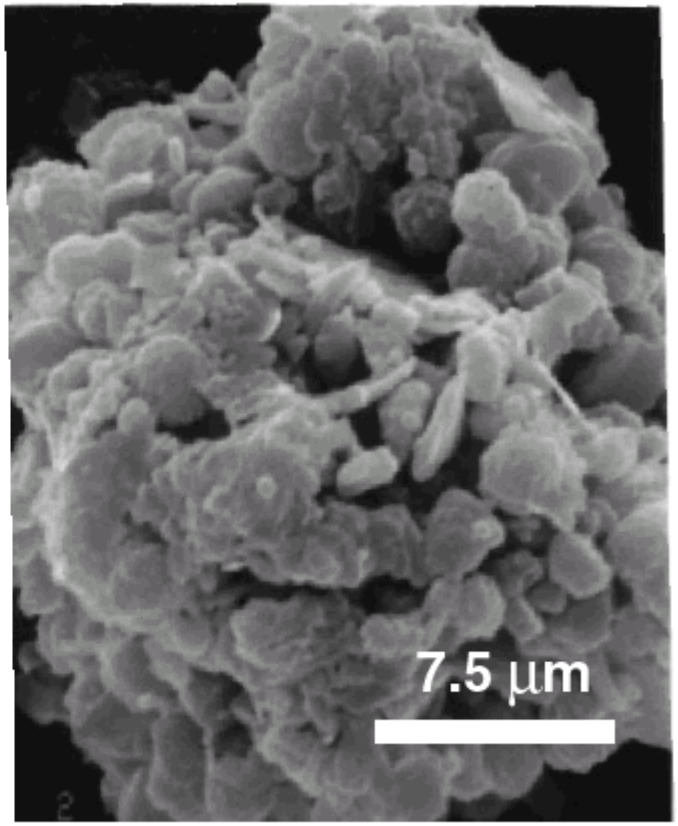




Treated Hanford Sediments

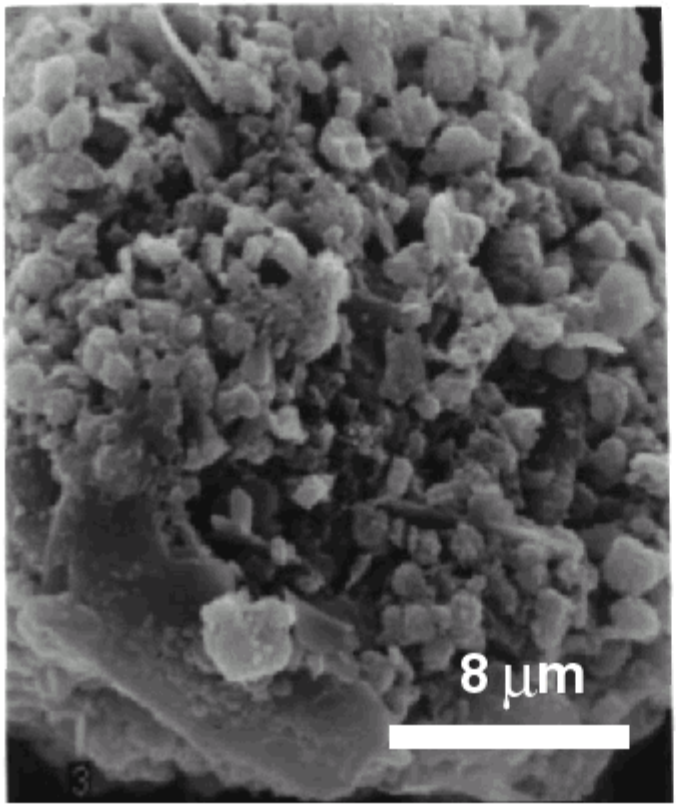


Colloid 1

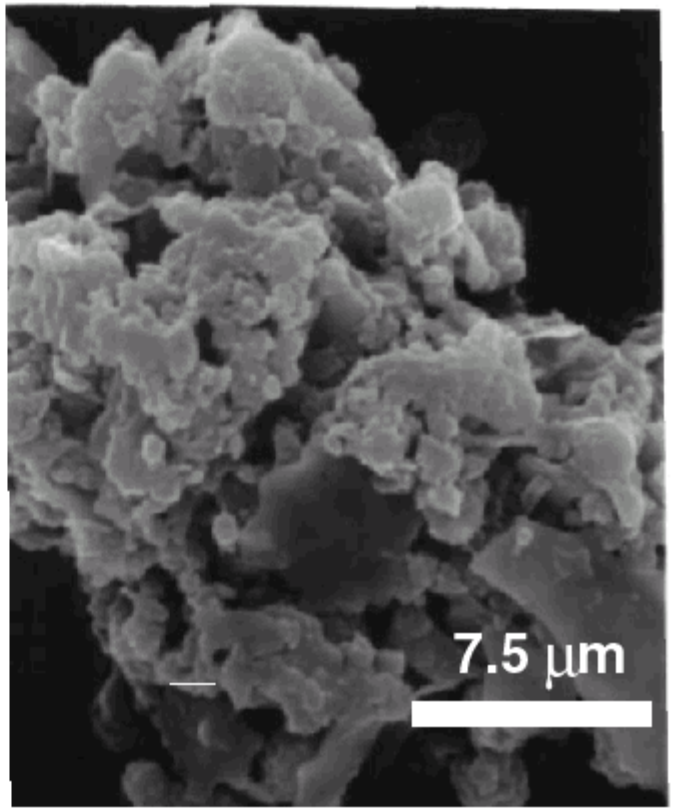


Colloid 2

Treated Hanford Sediments

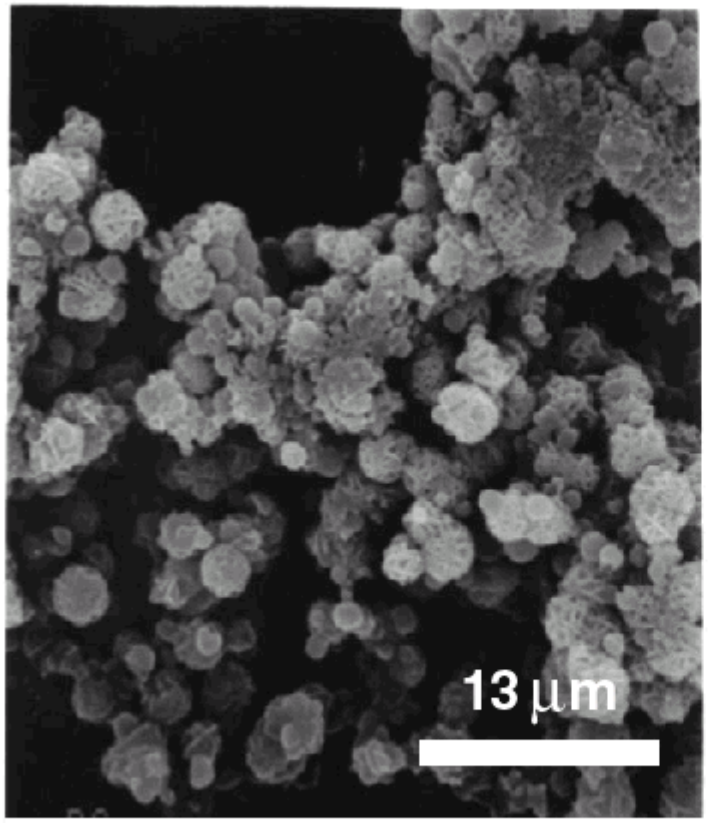


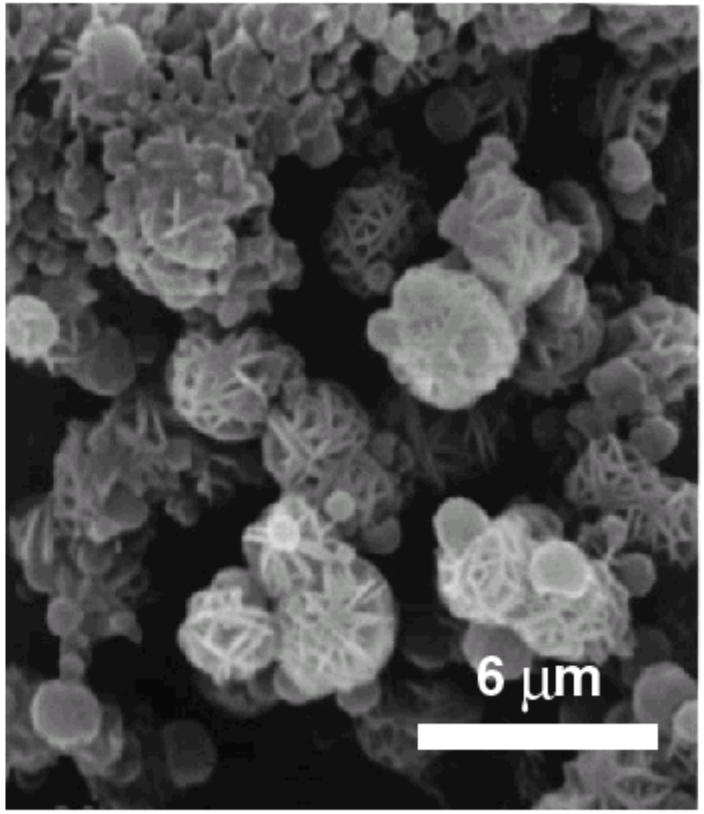
Colloid 3



Colloid 4

Pure Solutions: 0.036 M Si and STW 1





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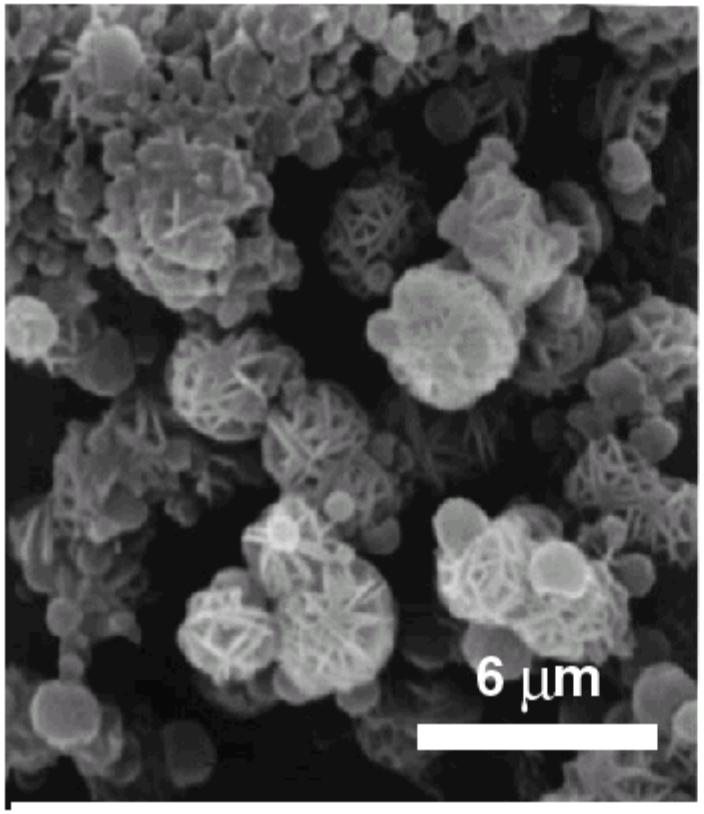
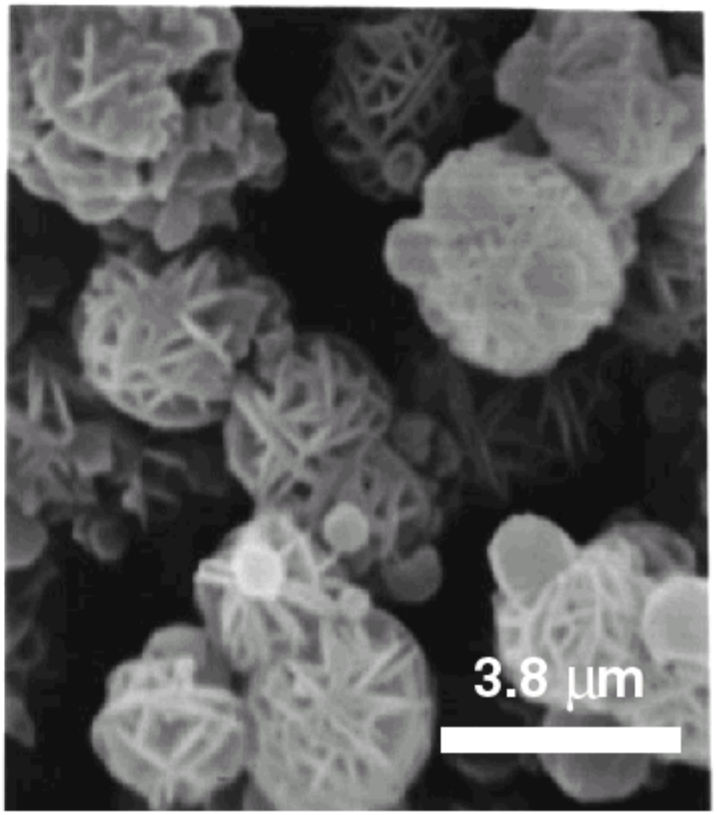
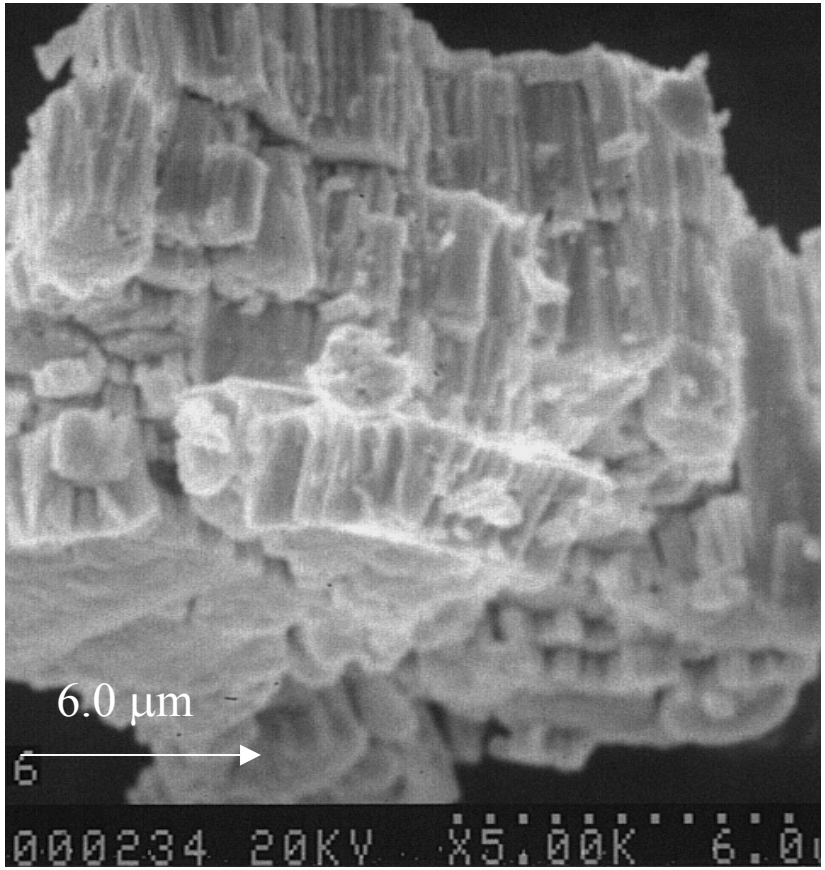
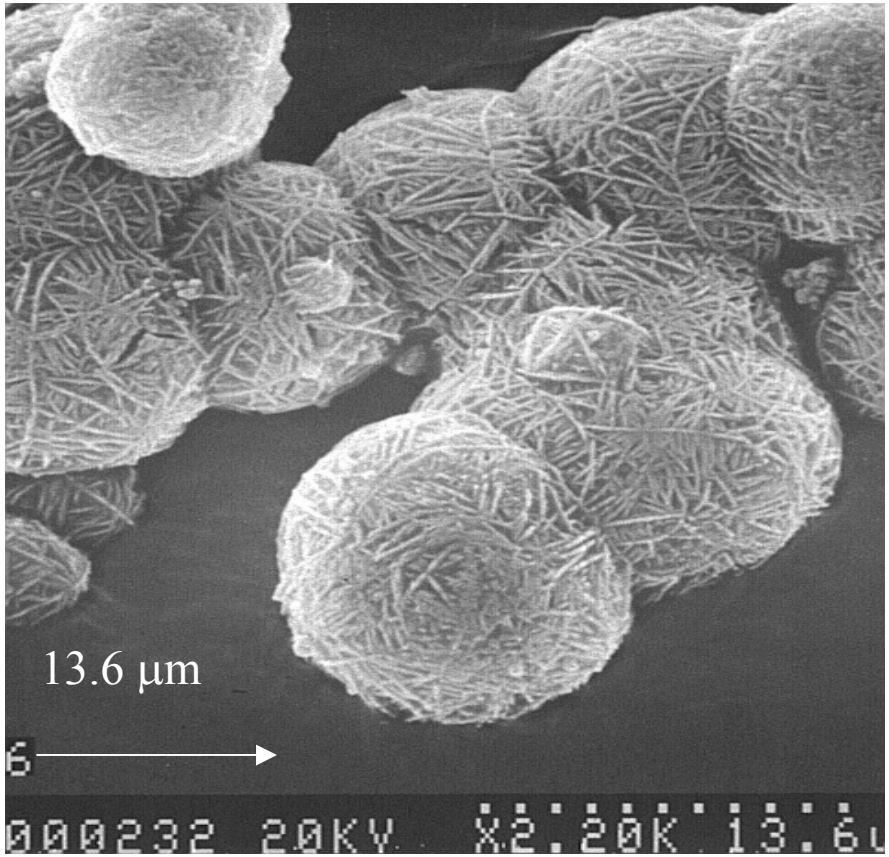


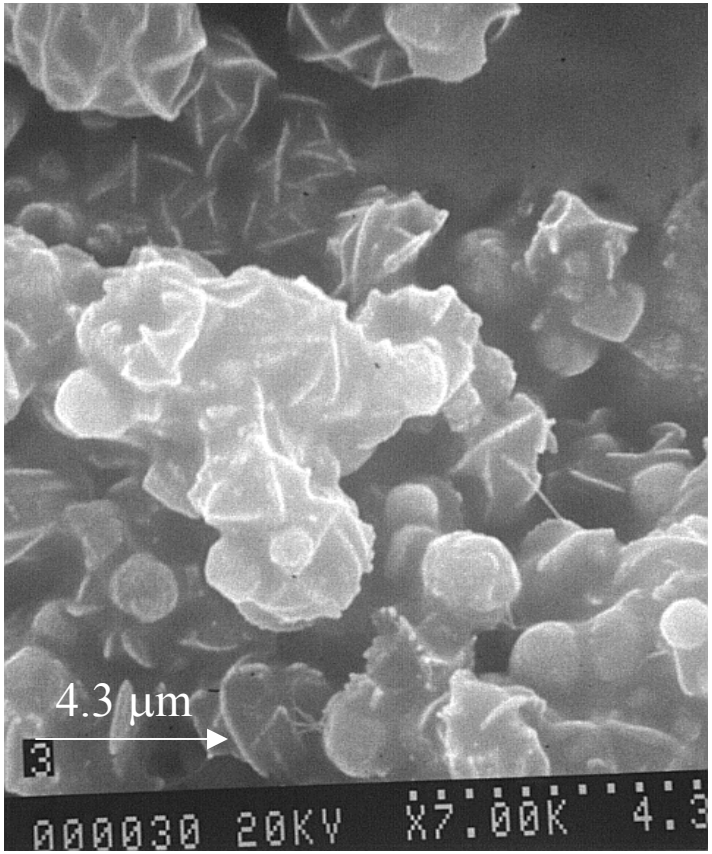


0.036M Si + STW #2

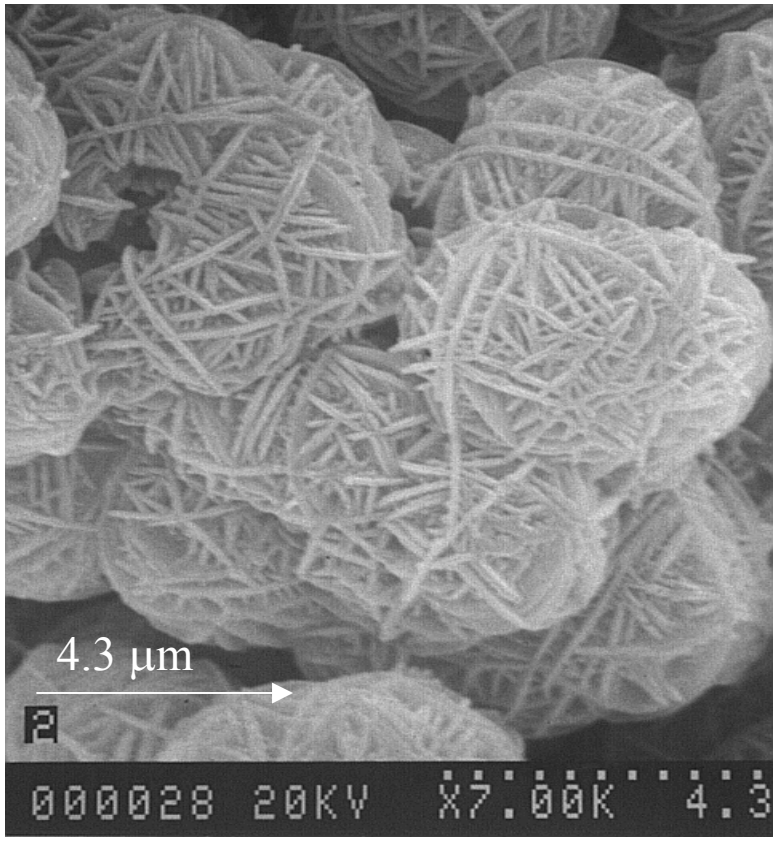




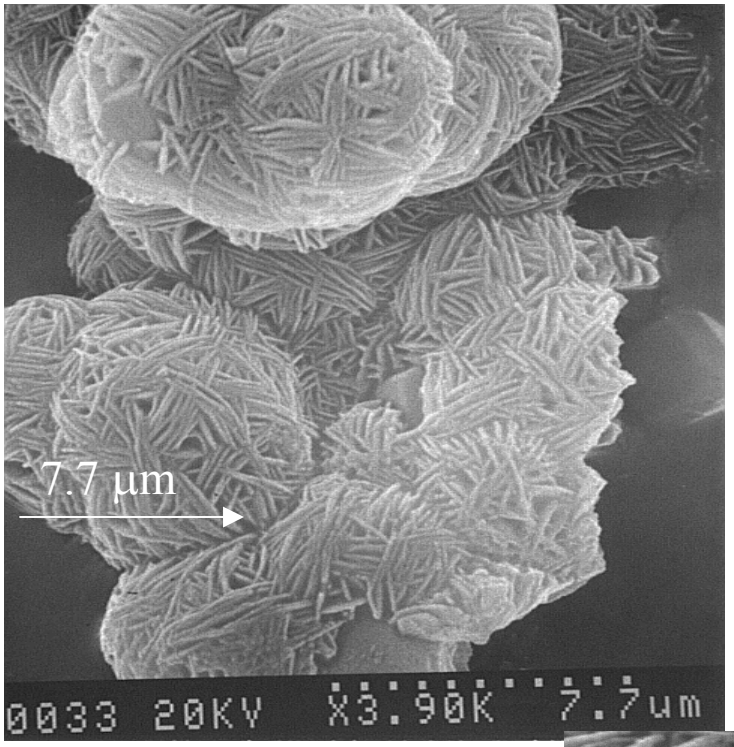
0.036M Si + STW #3

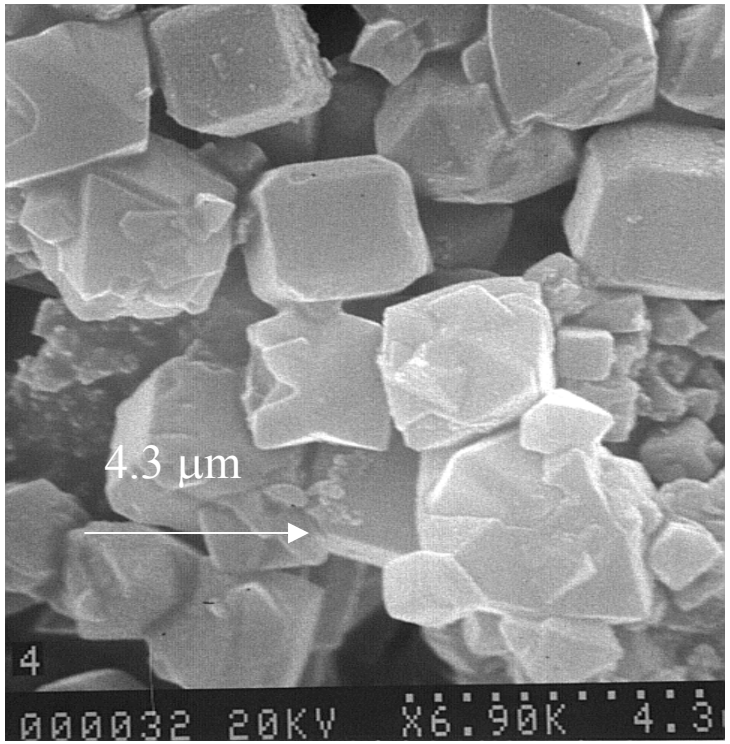


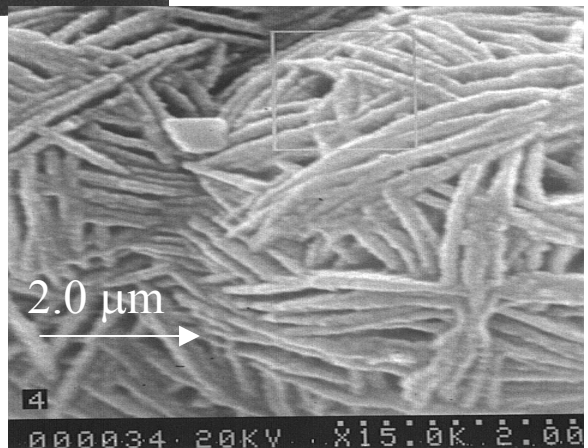
0.036 M Si + STW #4



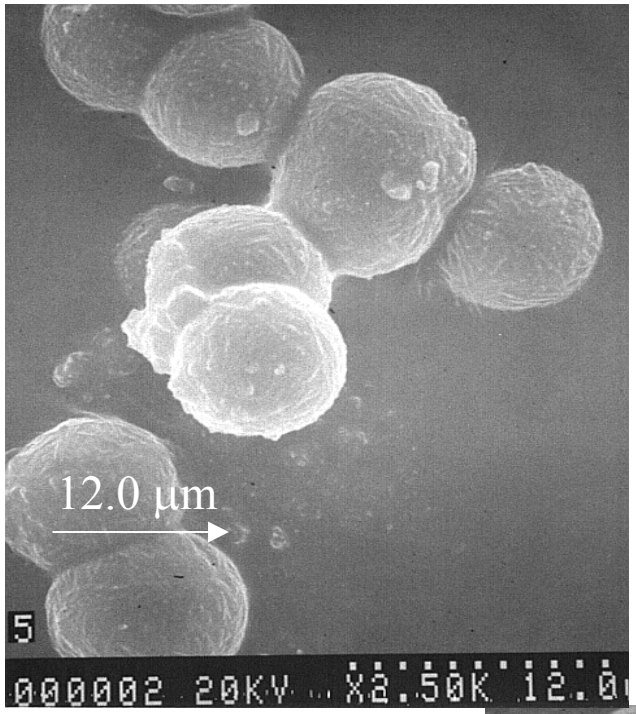
0.036m Si + STW #1 (0.018m Al) 1:2 (Al:Si)

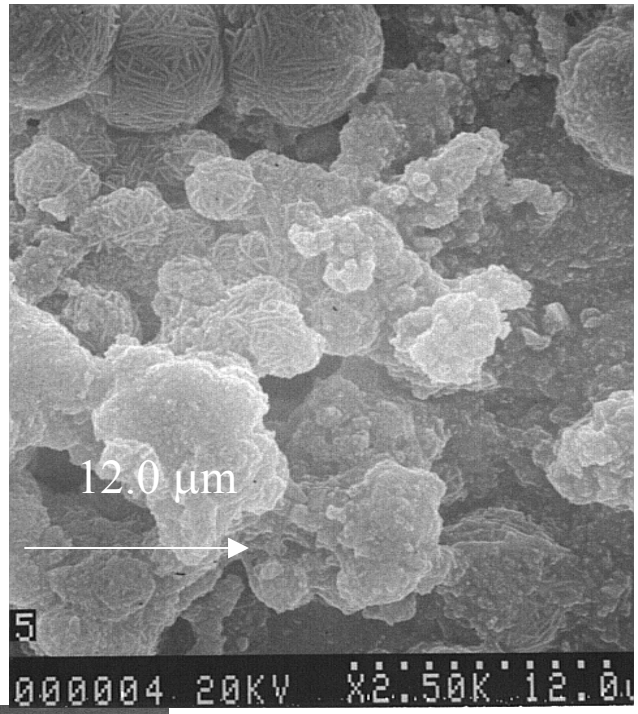


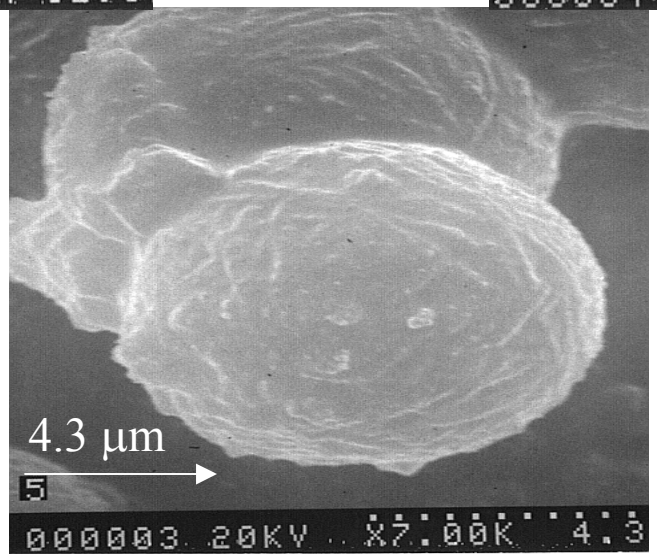




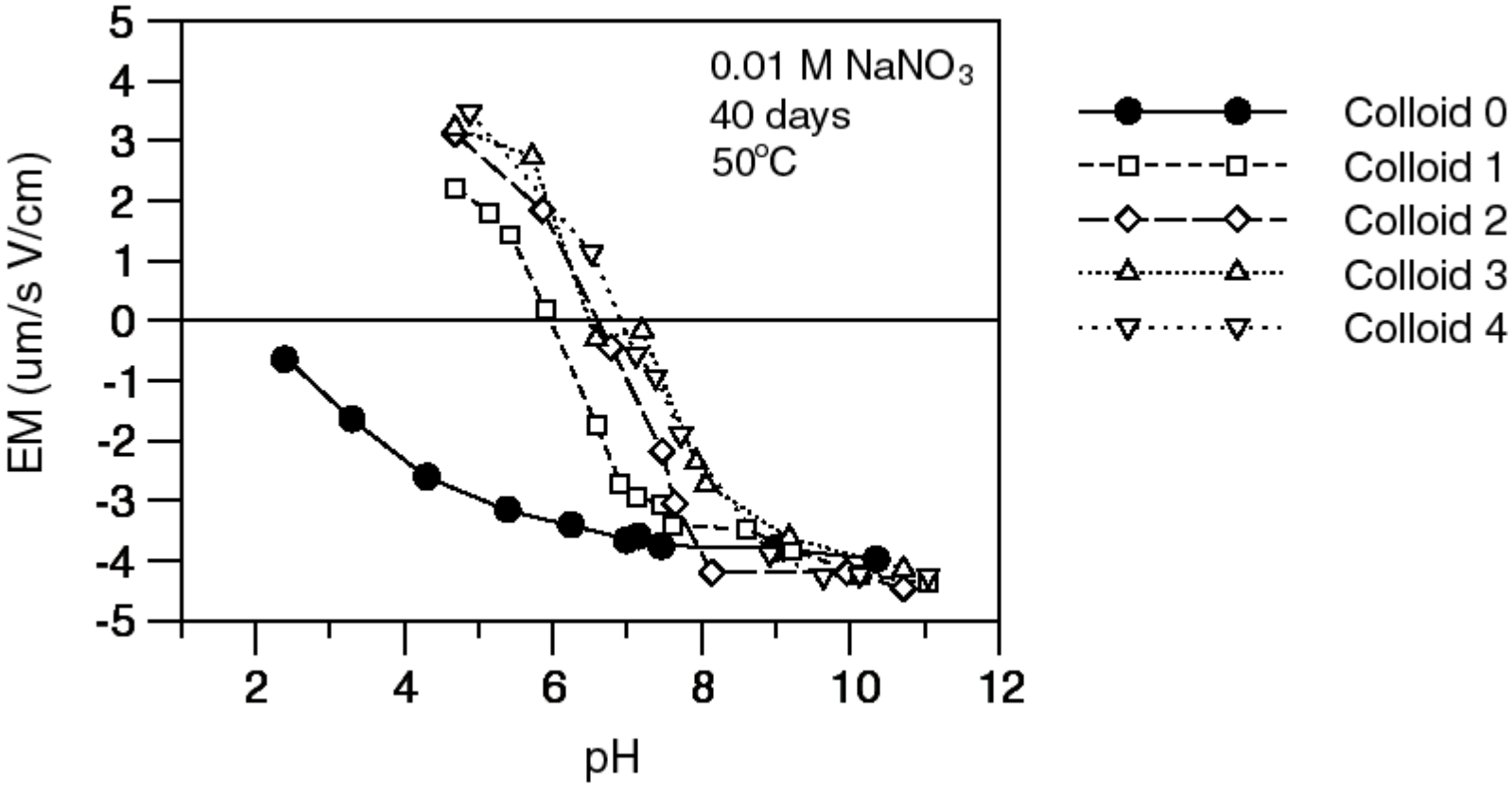
0.036m Si + STW #1 (0.072m) 2:1 (Al:Si)



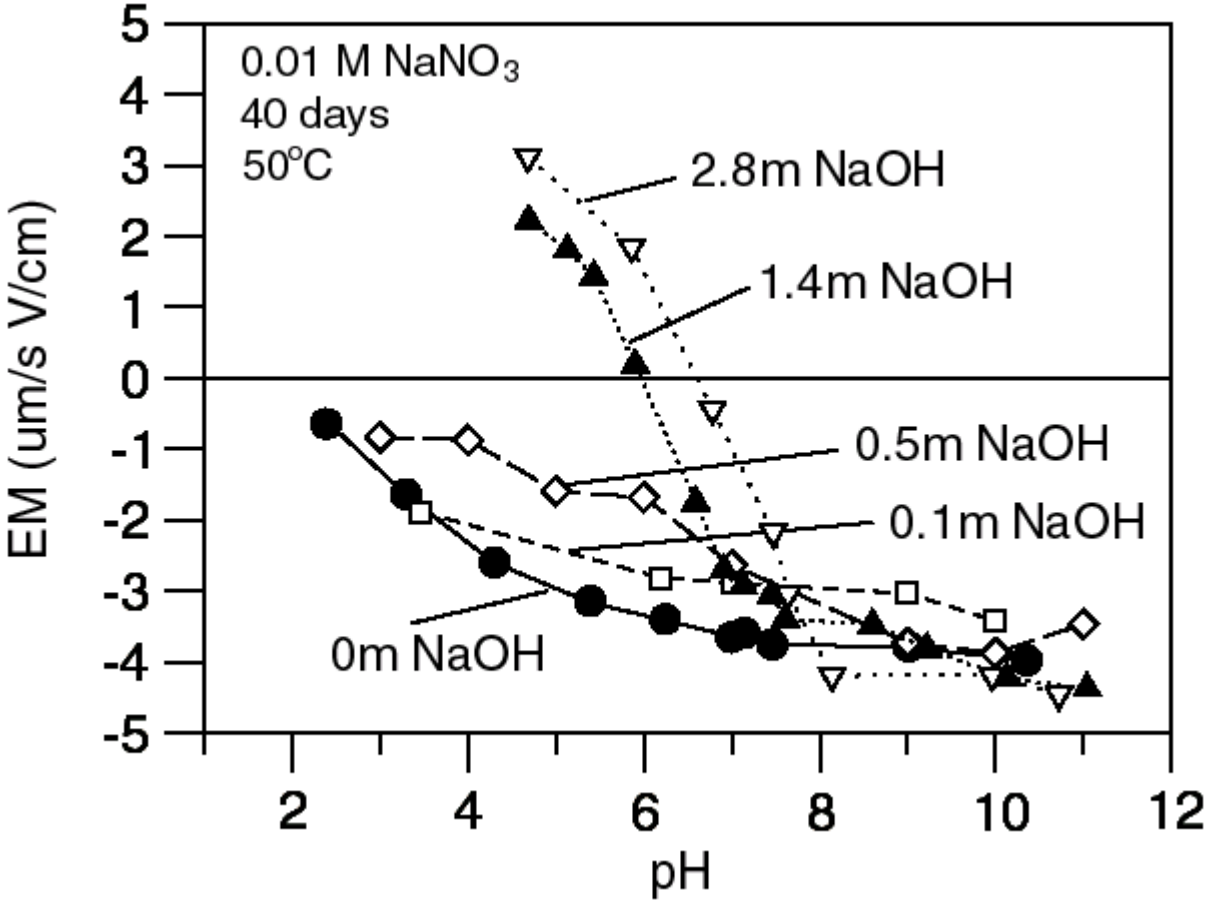




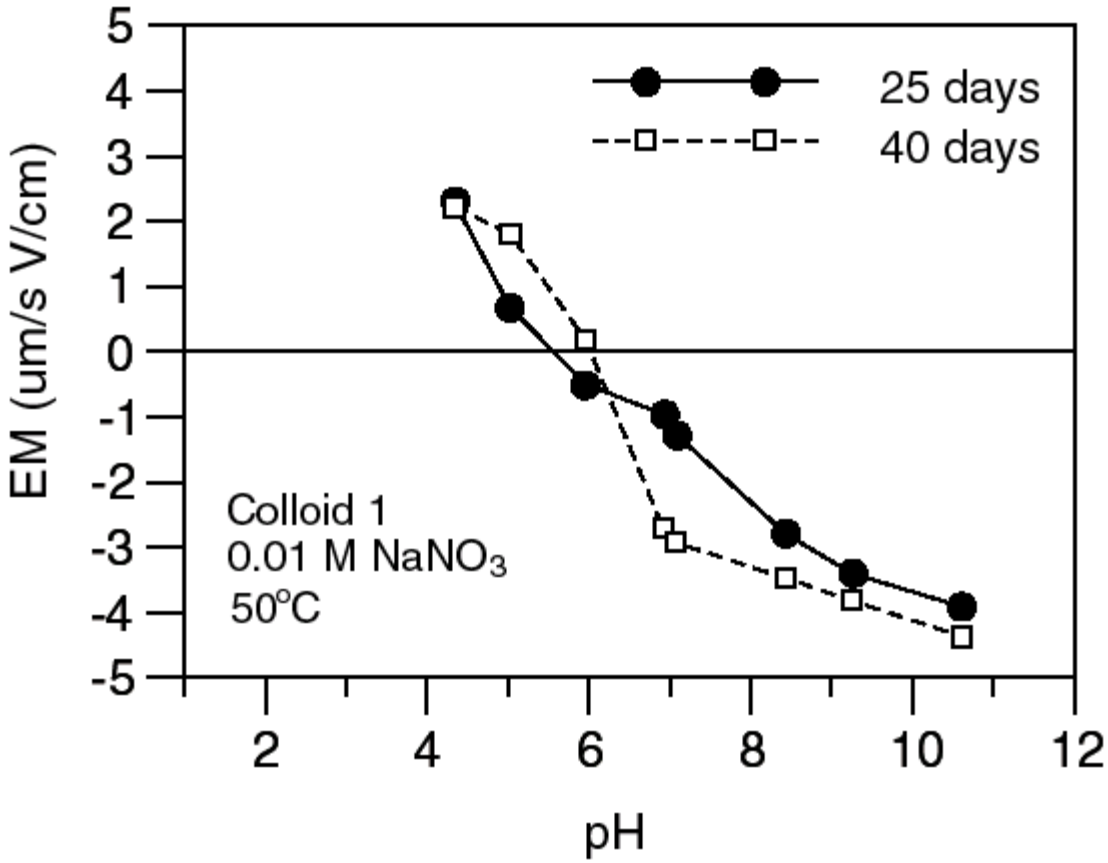
Electrophoretic Mobility



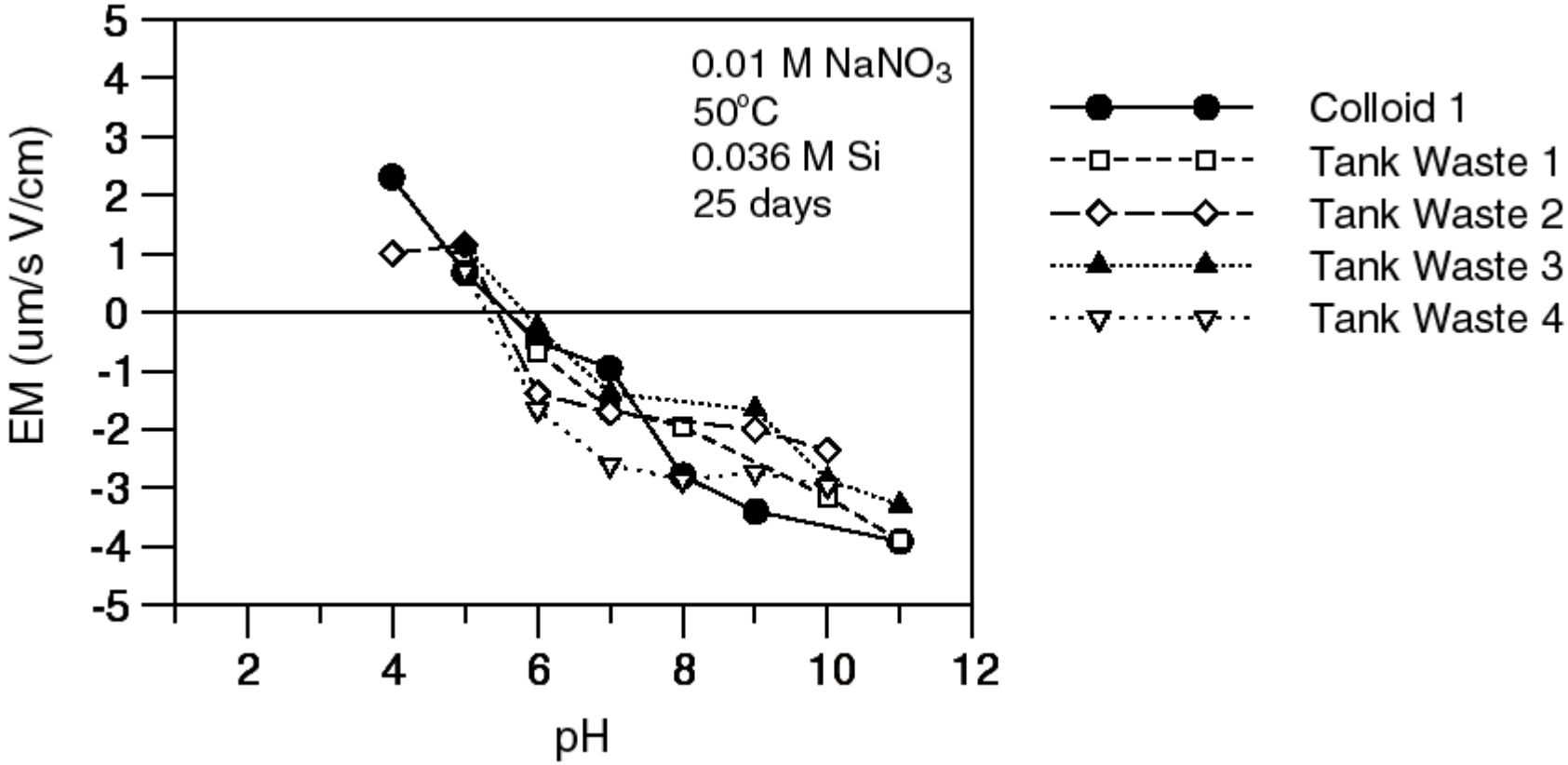
Electrophoretic Mobility



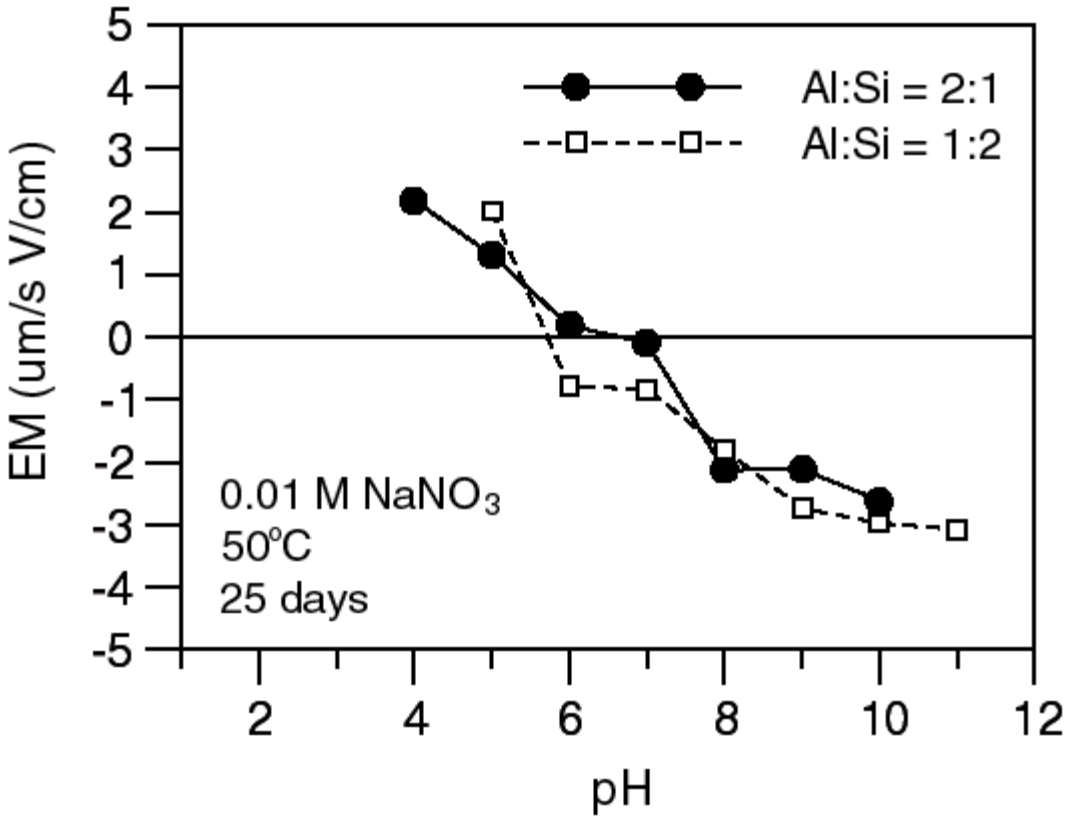
Electrophoretic Mobility



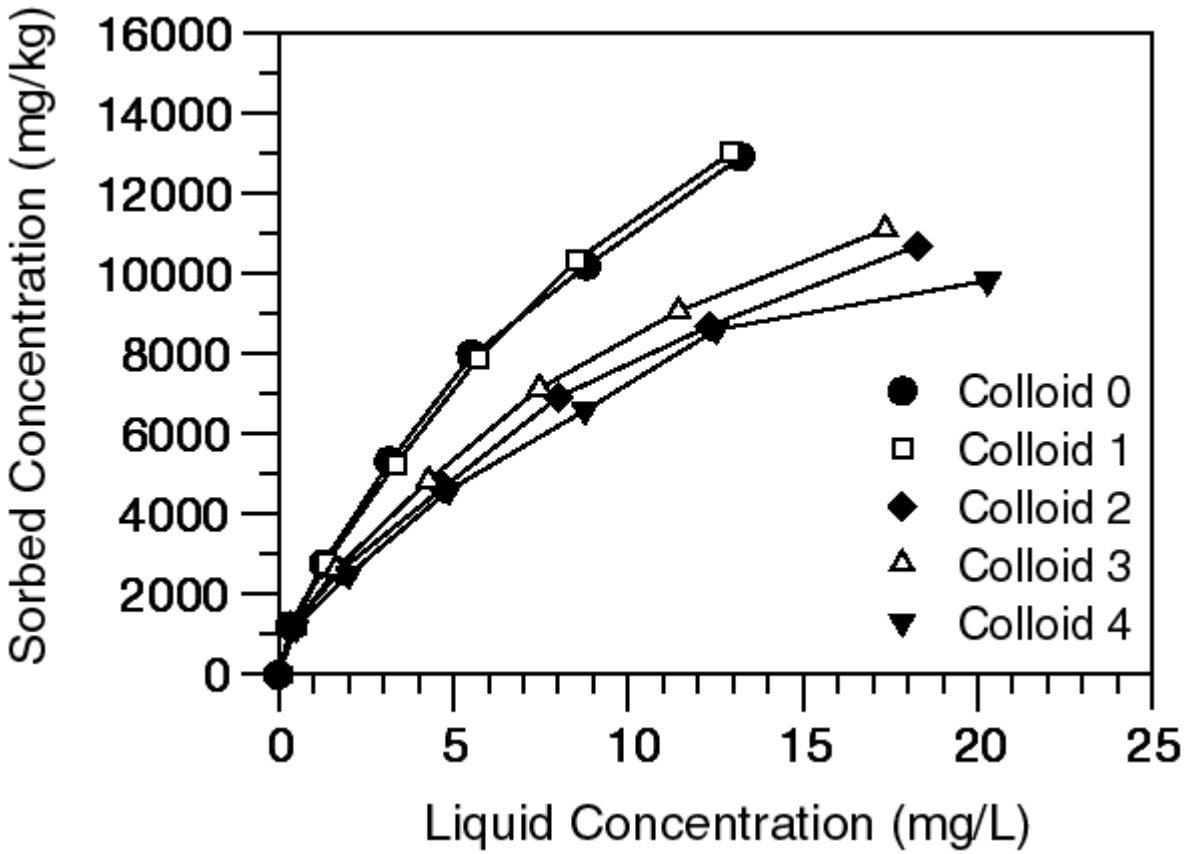
Electrophoretic Mobility



Electrophoretic Mobility



Cesium Sorption on Colloids



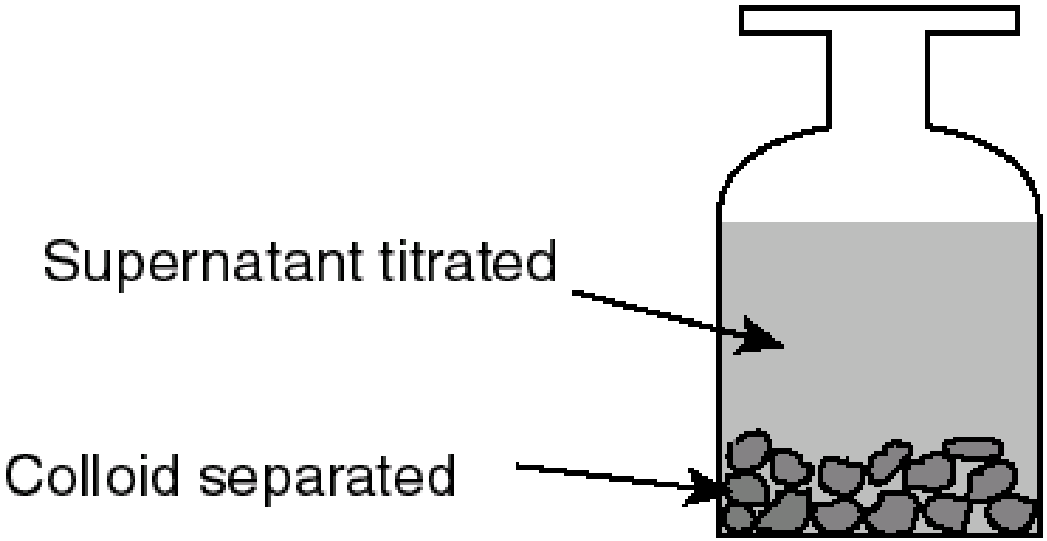
Transformation Mechanism

(Barnes et al., 1999):

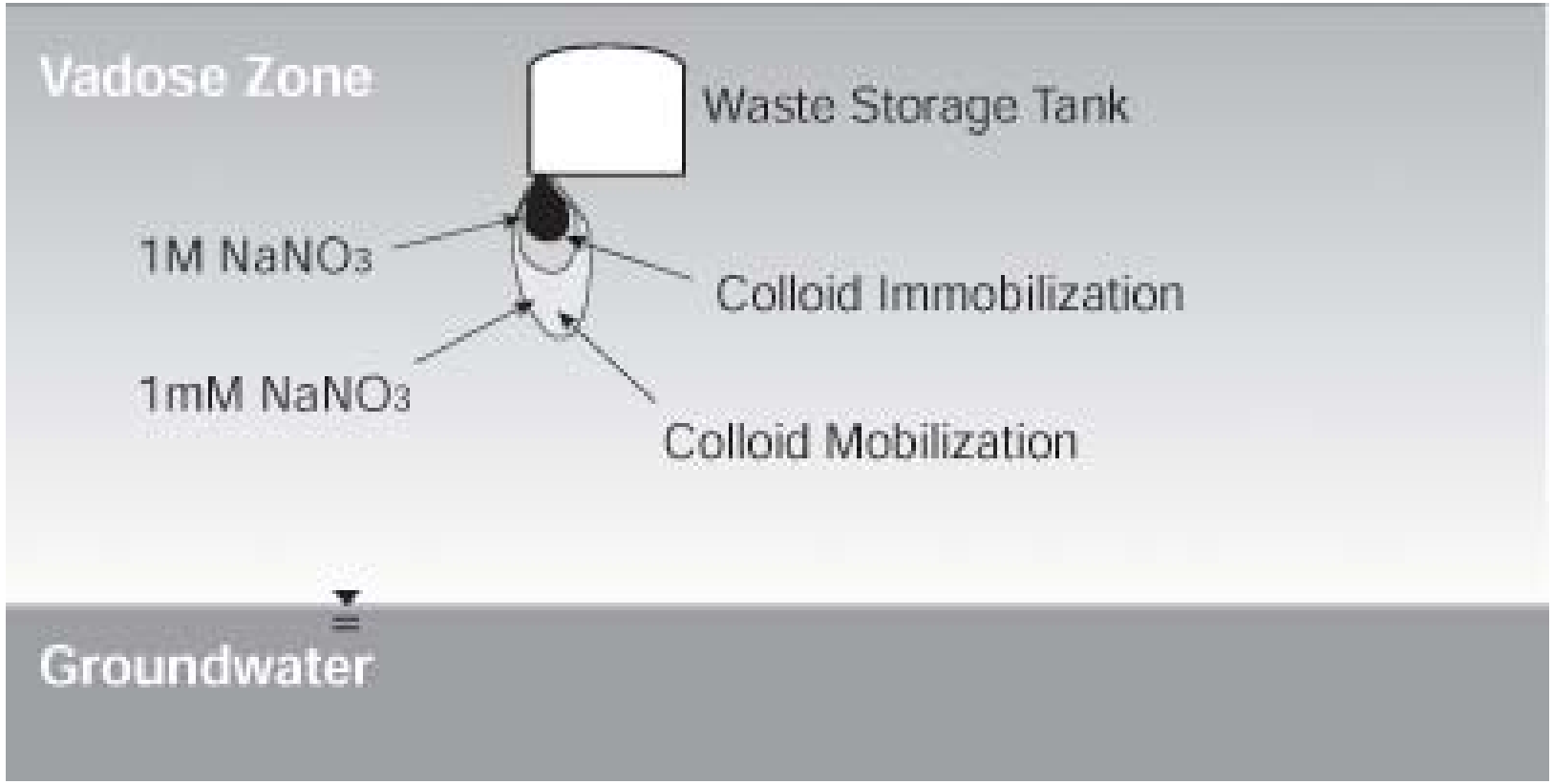
Aluminosilicate species $\Rightarrow$ amorphous phase $\Rightarrow$

zeolite $\Rightarrow$ sodalite $\Rightarrow$ cancrinite

Supernatant Solutions



Colloid Mobilization



Colloid Mobilization

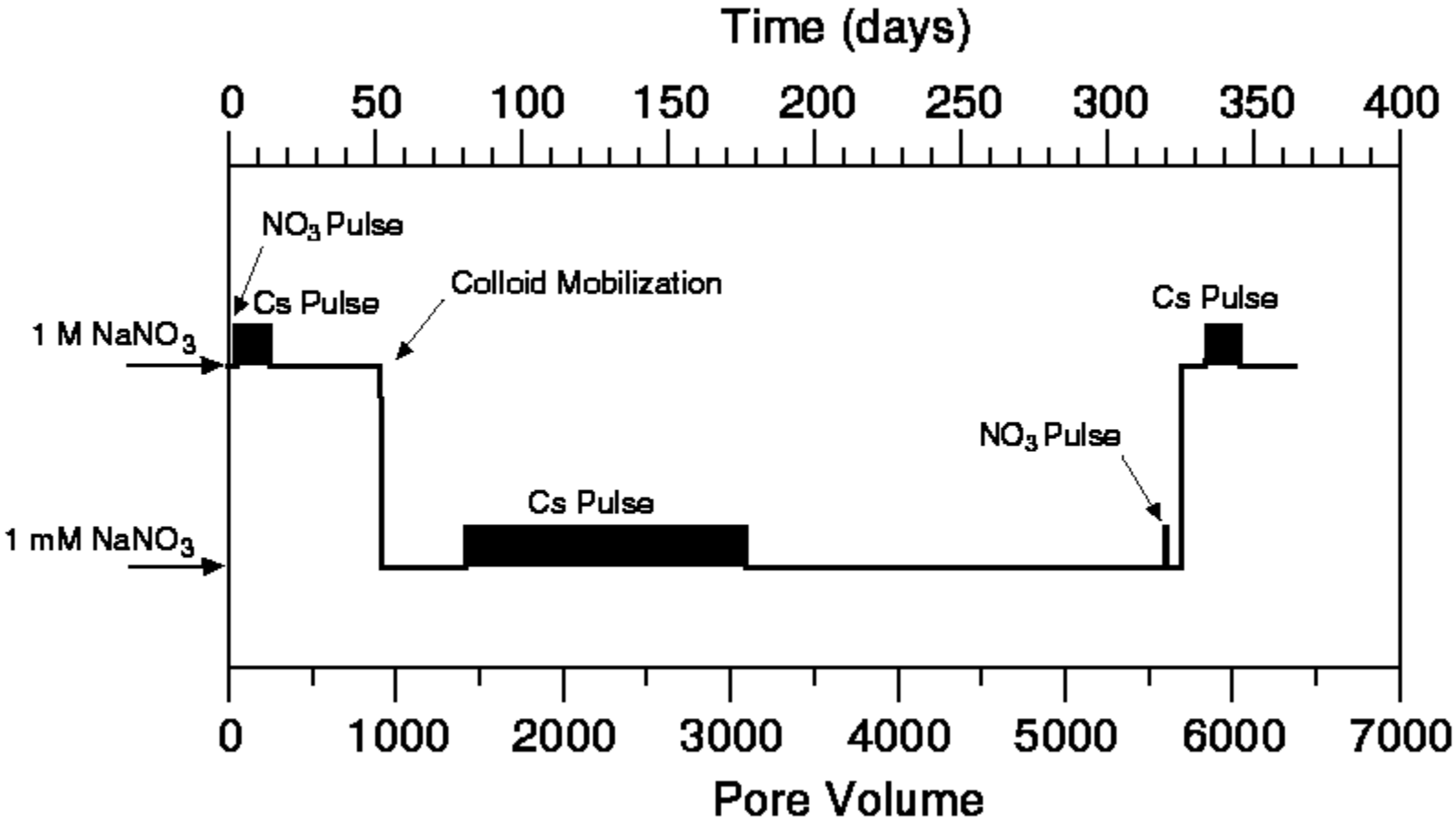
1 M $NaNO_3$
Immobilization

→

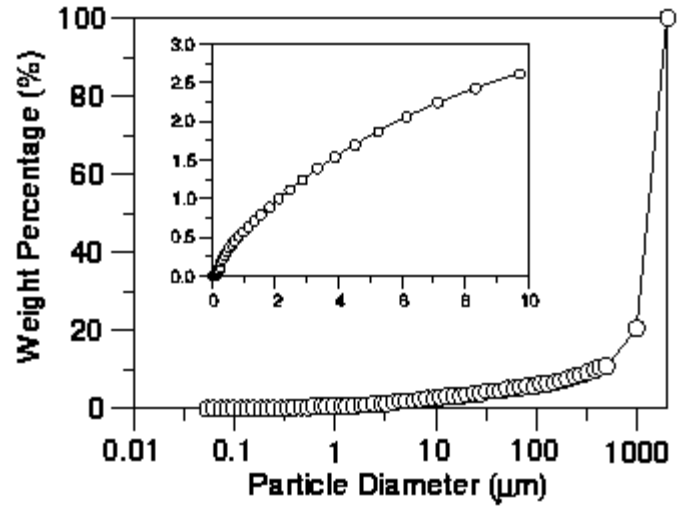
1 mM $NaNO_3$
Mobilization

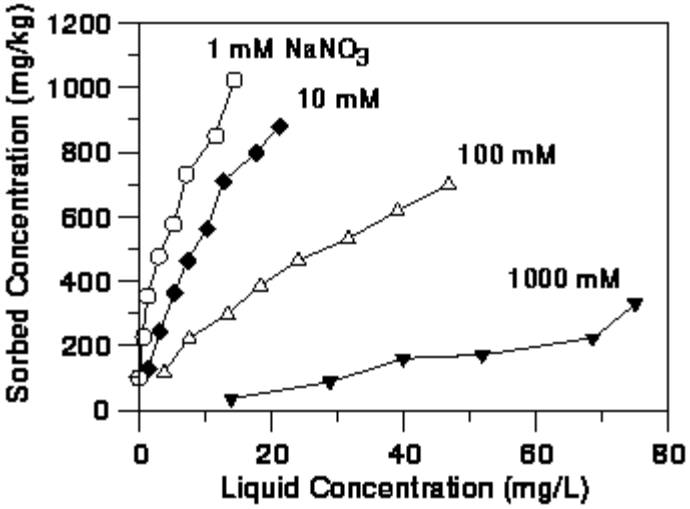
Hydrodynamics
Cs Transport
Colloid Transport
Colloid Characterization

Colloid Mobilization

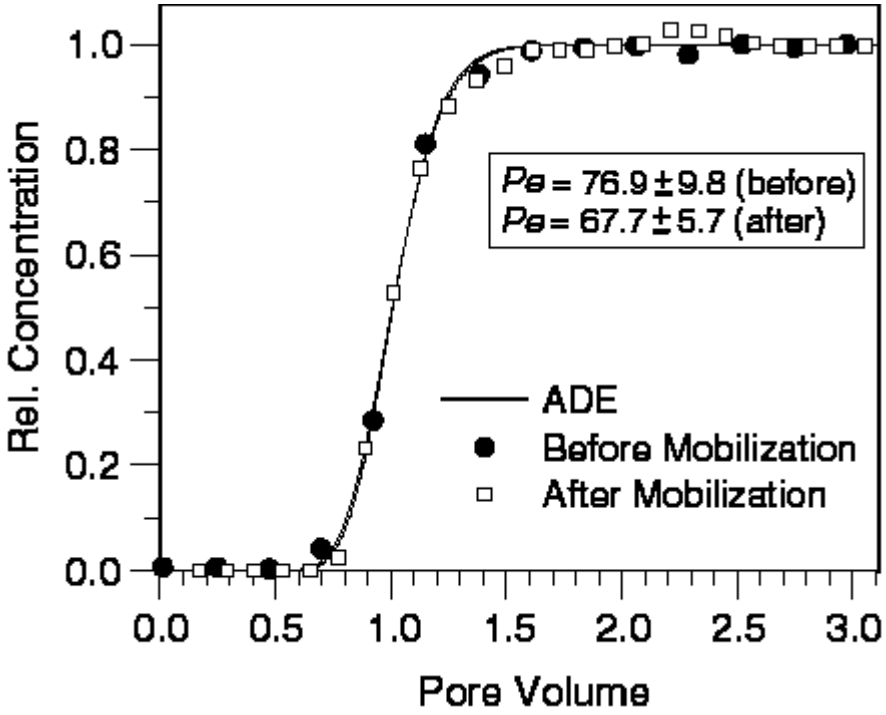


Colloid Mobilization

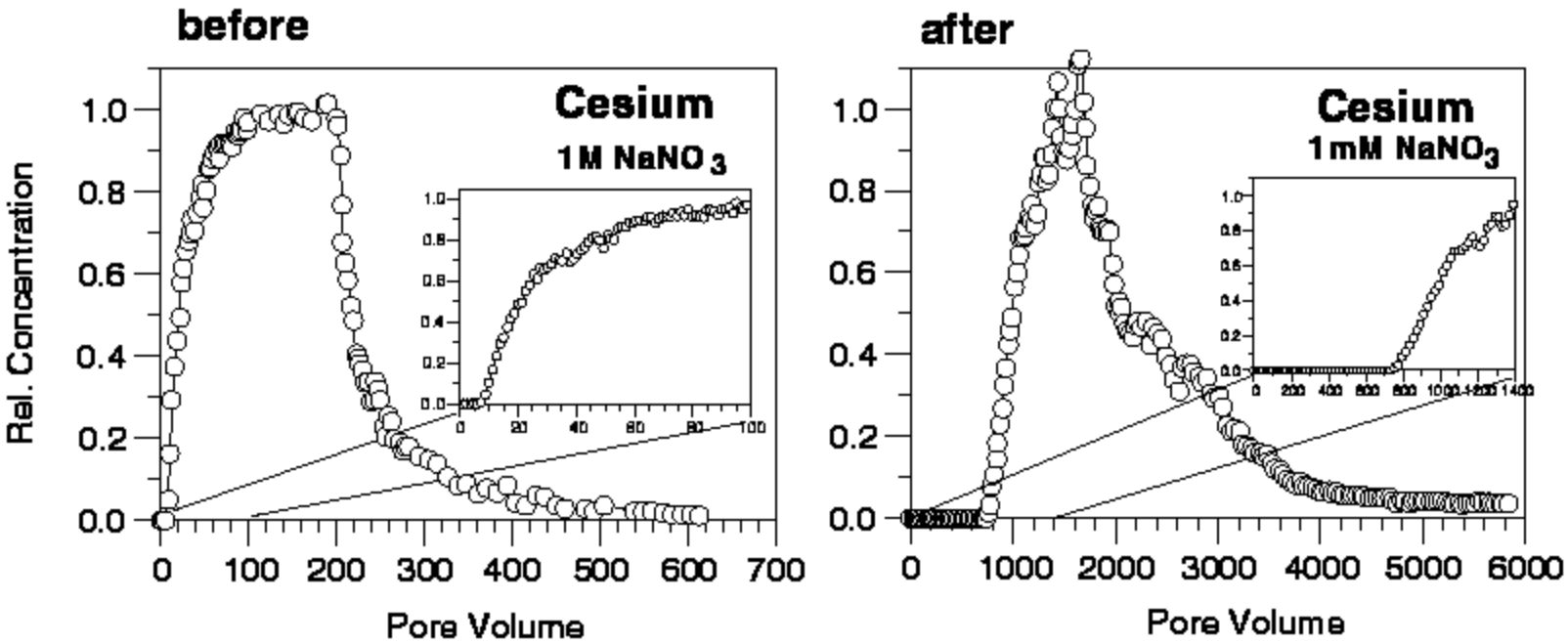




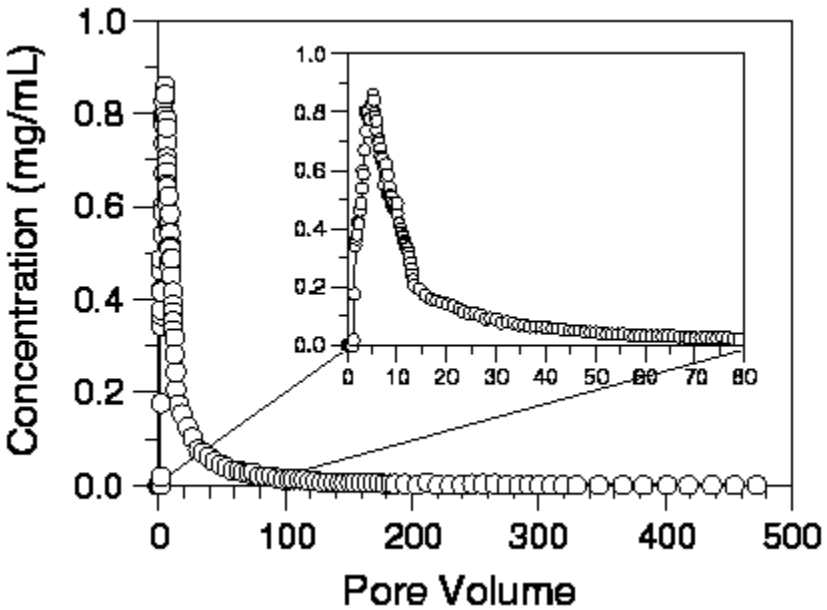
Colloid Mobilization

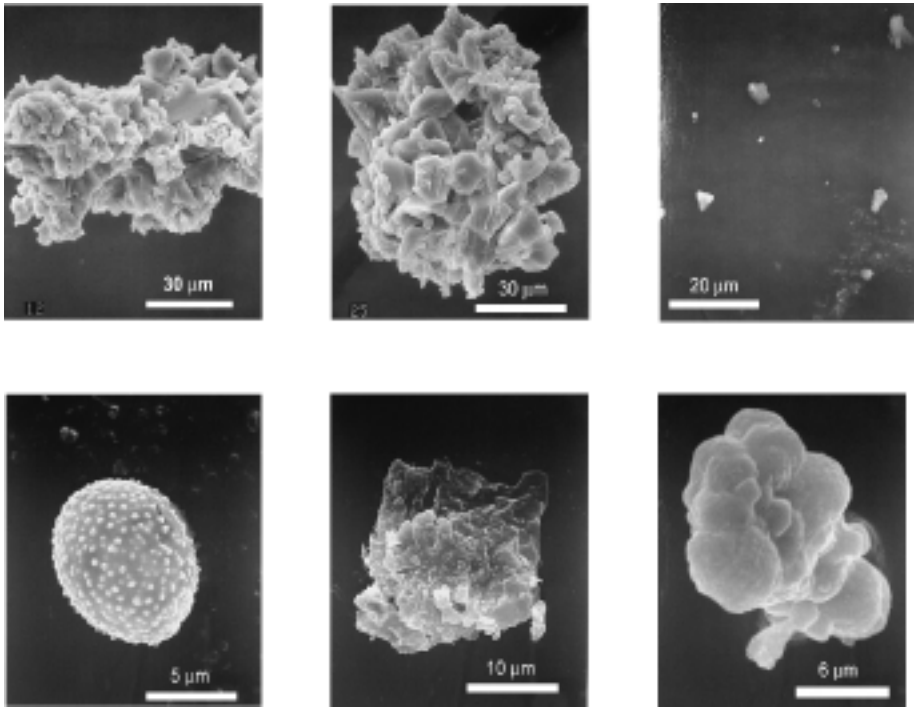


Colloid Mobilization

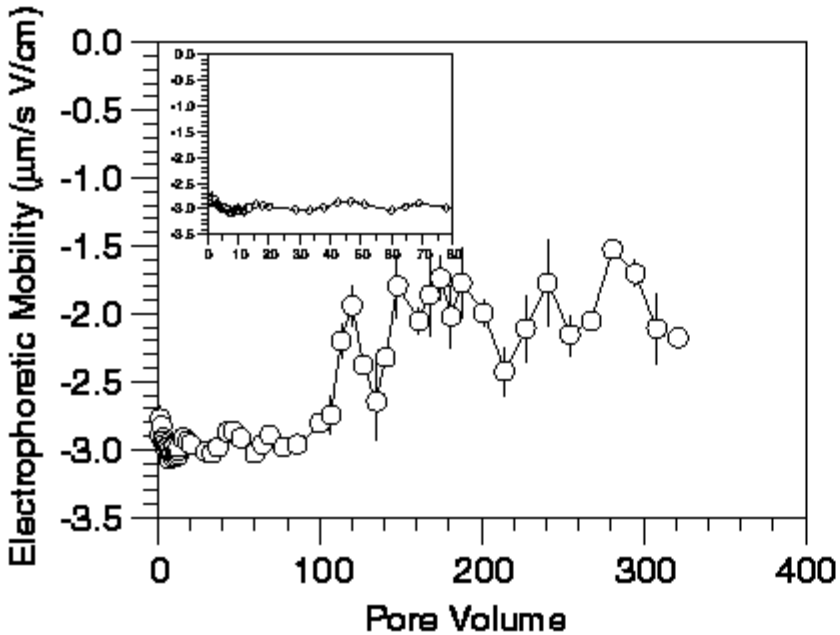


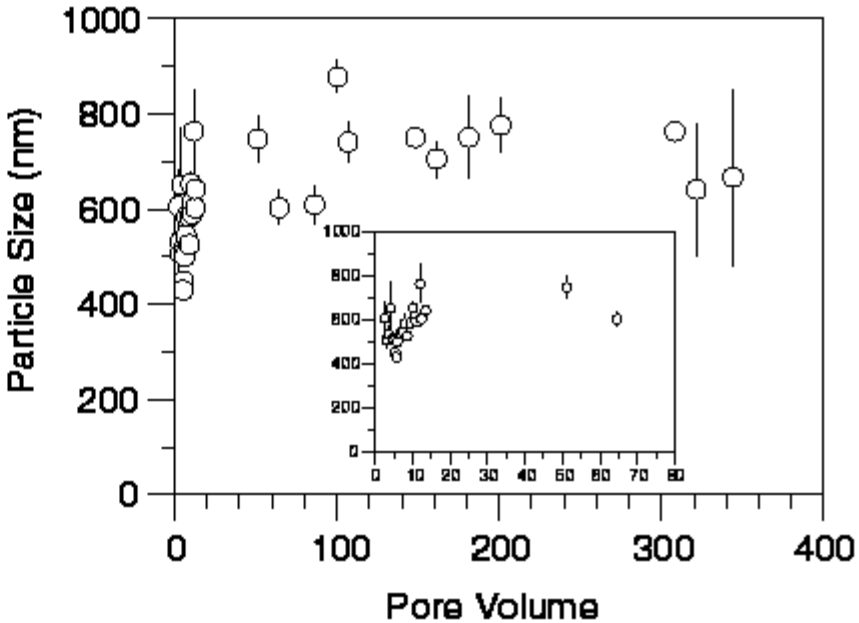
Colloid Mobilization



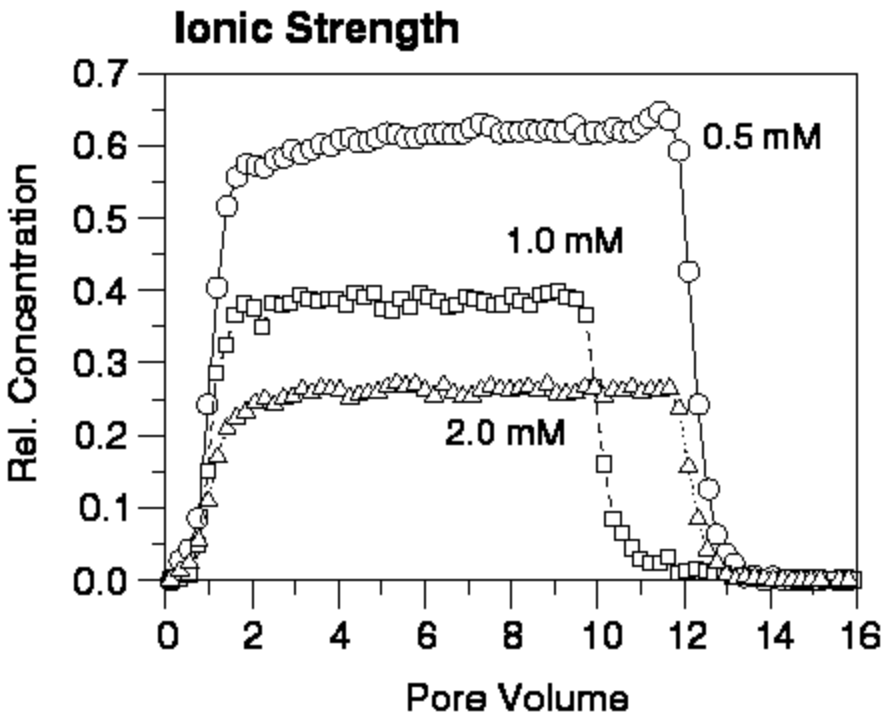


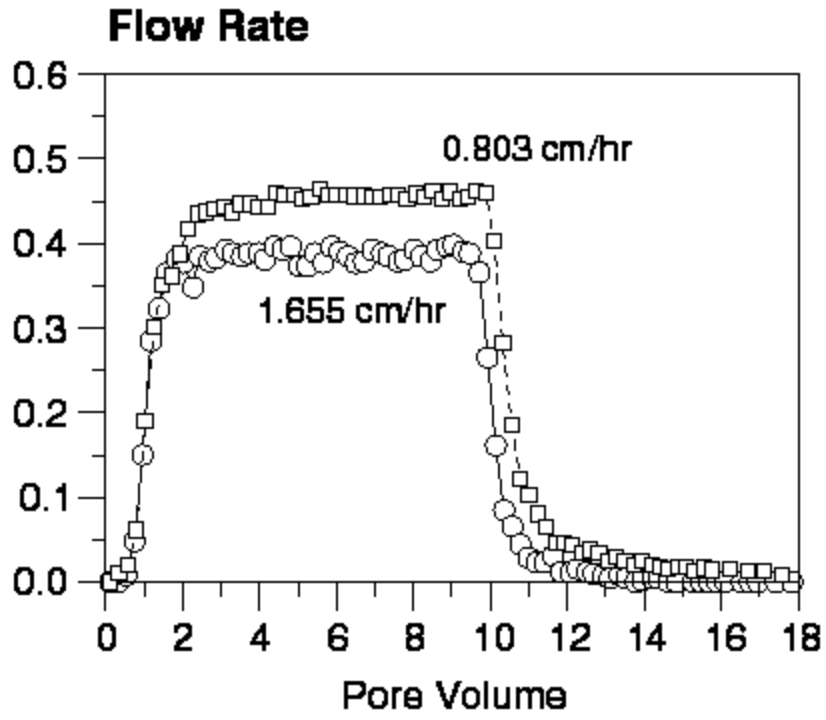
Colloid Mobilization





Colloid Transport





Conclusions

Colloid formation:

- Colloids are forming by:
  1. Alteration of native minerals
  2. Precipitation of new mineral phases
  3. Precipitation from supernatant solutions.
- Colloid yield considerable.

Conclusions

X-ray diffraction patterns:

- The < 2 μm fraction of untreated sediments consists of four major layered aluminosilicates--chlorite, smectite, kaolinite and illite.
- Upon reaction with the alkaline aluminate solutions, the results show that feldspathoids (mixture of cancrinite and sodalite) were formed.

Conclusions

SEM/EDAX:

- Untreated sediment clays have the distinct morphology of phyllosilicate clay minerals.
- Clays extracted from treated sediments show the appearance of spheres with surface features common to cancrinite.
- Reaction of STW with Si solutions produced solids with morphologies common to cancrinite, sodalite, and zeolite.

Conclusions

Electrophoretic mobilities:

- The untreated sediment clays have only negative EM between pH 2 and 11, although mobility becomes less negative at lower pH.
- The treated sediment clays have points of zero charge between pH 5 and 8 depending on time and STW composition.
- Colloids from the Si solutions are similar to those formed from the sediments but show EM depends on pH and Al:Si ratio of the initial solution.

Conclusions

The negative charge on the colloids at pH values typical for the vadose zone at Hanford (pH > 7.0) suggests that:

- they are likely to form stable, mobile suspensions,
- they are not electrostatically attracted to the predominantly negatively charged surfaces of Hanford sediments, and
- Cations such as Sr^{2+} and Cs^{+} will be electrostatically attracted to the colloids.

Conclusions

Colloid Mobility:

- In situ mobilization possible
- Colloids are mobile at low ionic strength
- Cesium transport can be facilitated