Short and long-term behaviour of bentonites
in contact with solutions of different salinities - A safety issue for the long-term closure of repositories?

Results of a 3 years laboratory study

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Background:

•Bentonites, part of EBS

•High salinity, high pH of pore solutions changes in mineralogy and swelling capacity

•Impact on long-term performance of EBS?

Objectives:

- Reactions of bentonite with repository relevant solutions
- Mineralogical changes
- Swelling pressure changes
- Long-term performance

Working hypothesis:

•Pore solutions of different ionic strength influence the swelling pressure of bentonite differently

•The layer charge of montmorillonite affects inversely the swelling pressure of run products

•In closed reaction systems montmorillonite is undergoing a mineralogical alteration, which is causing a reduction of the interlayer charge in the run products. This reduced charge should lead in the short term to increasing swelling pressures

•The theoretical end member of the alteration of montmorillonite in closed reaction systems may be kaolinite or pyrophyllite, leading in the long run to strongly reduced swelling pressures

Materials:

MX-80 bentonite + water
- Opalinus clay pore water
- Äspö URL water
- NaCl saturated water
- Mg-rich IP21 brine
- NaCl-cement solution
- IP21-cement solution
- cement pore solution

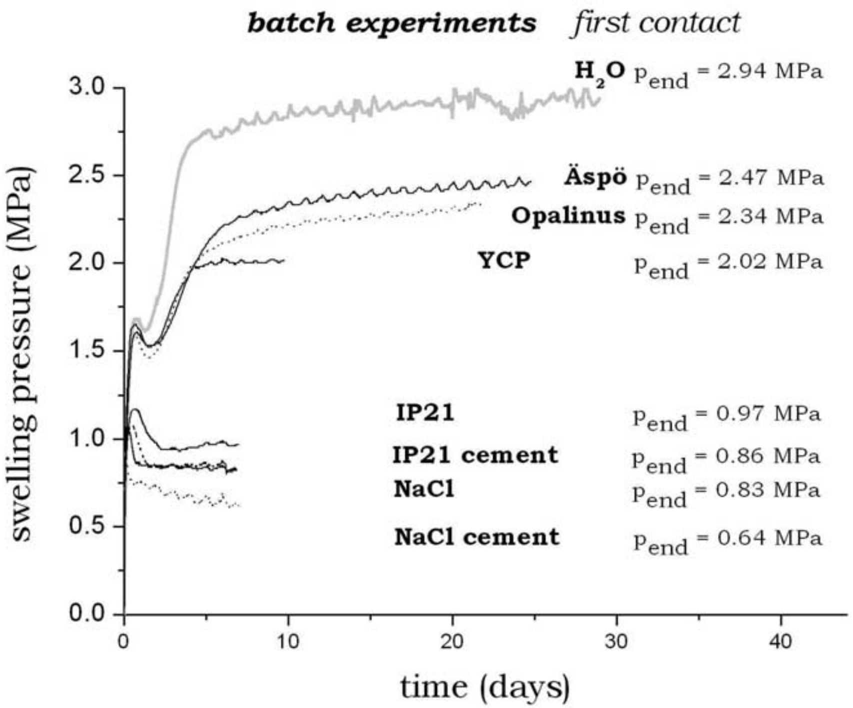
Experimental:

Batch experiments + Experiments with compacted bentonites

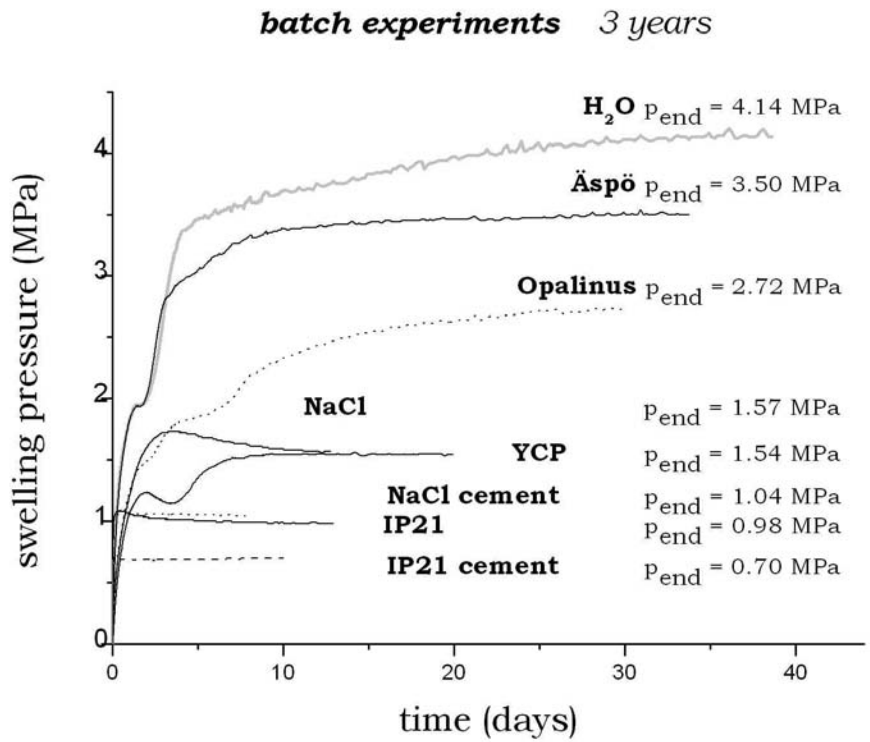
Measurements: 7 days, 1, 2, 3 years

- Swelling pressure
- Interlayer charge
- Cation exchange capacity
- Mineralogical changes (XRD, TEM, TEM-EDX)
  - Glycol expandability
  - Morphology of partivles
  - Order of particles
  - Interlayer spacing
- Fluid composition

Results – swelling pressure



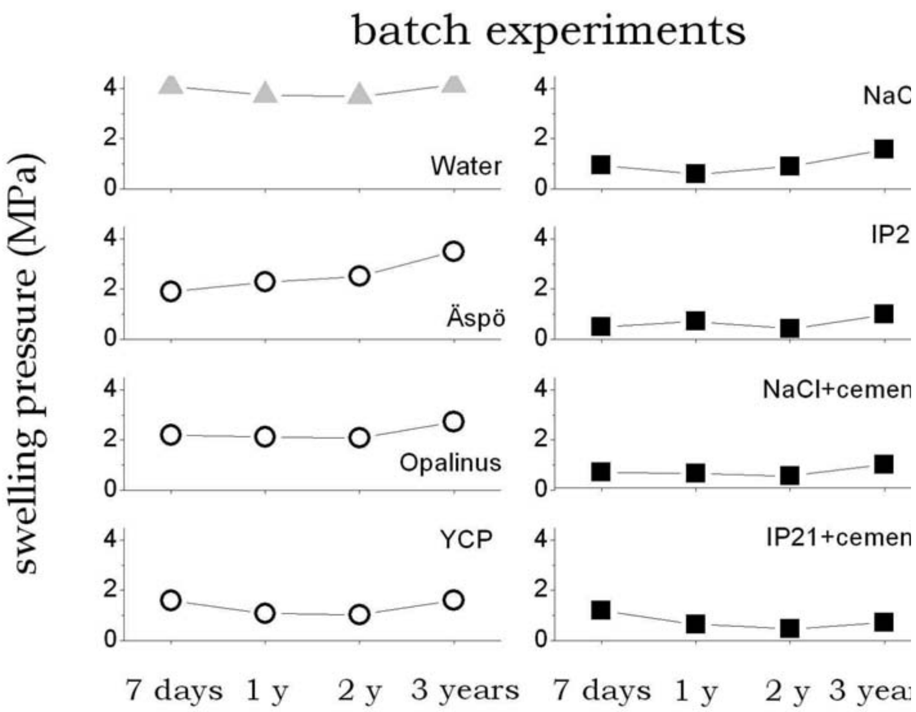
Original MX-80 bentonite

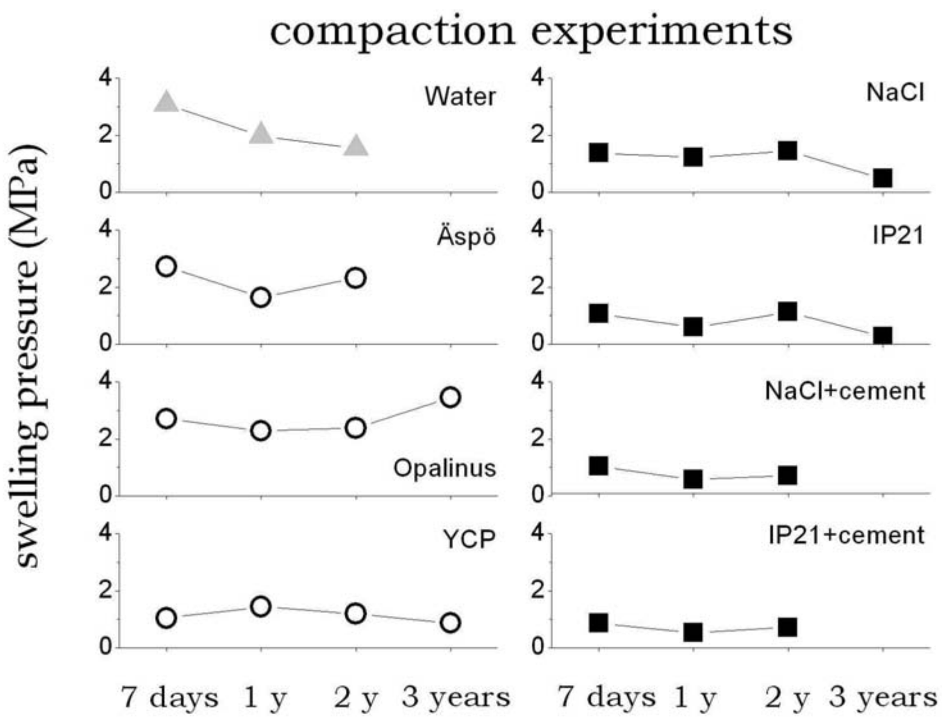


After 3 years of reaction

Results – swelling pressure

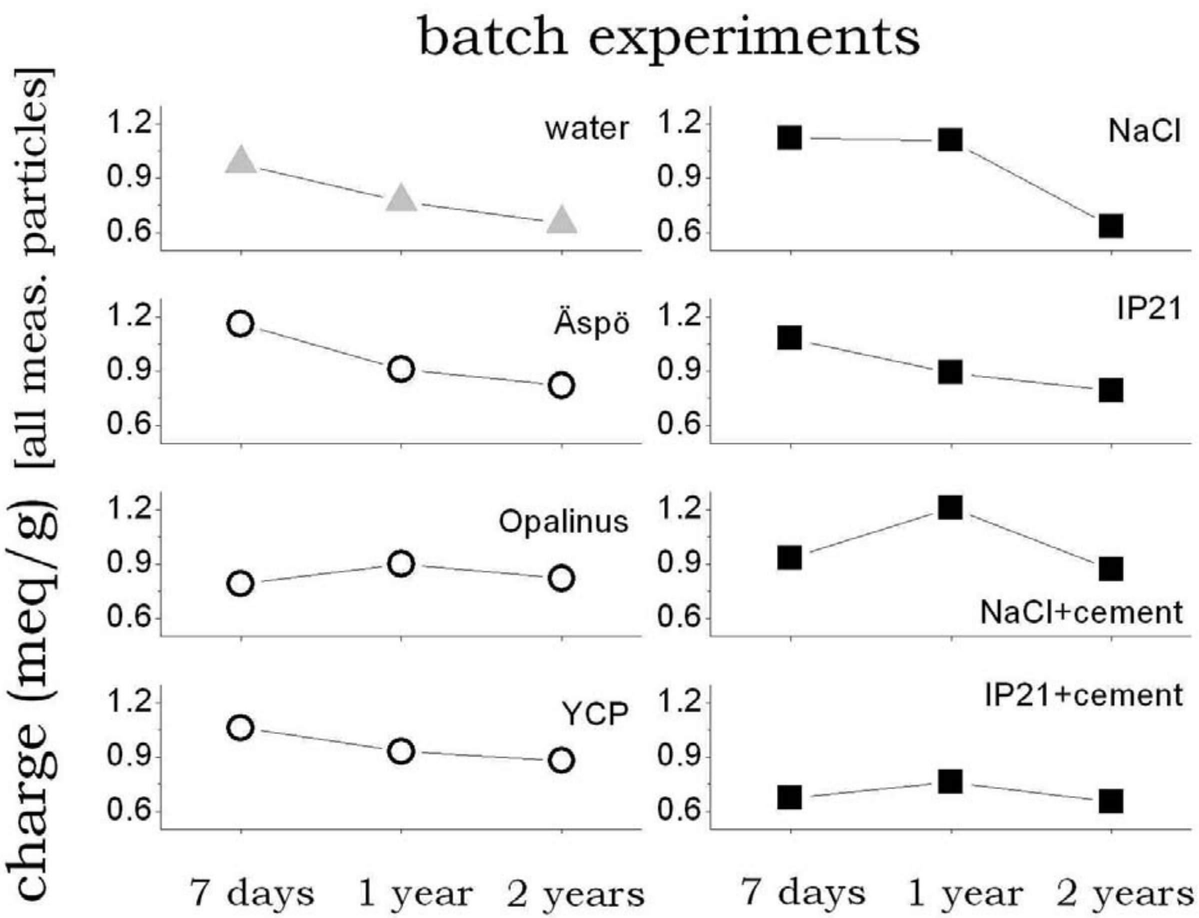
Development of swelling pressures in non-compacted and compacted bentonites





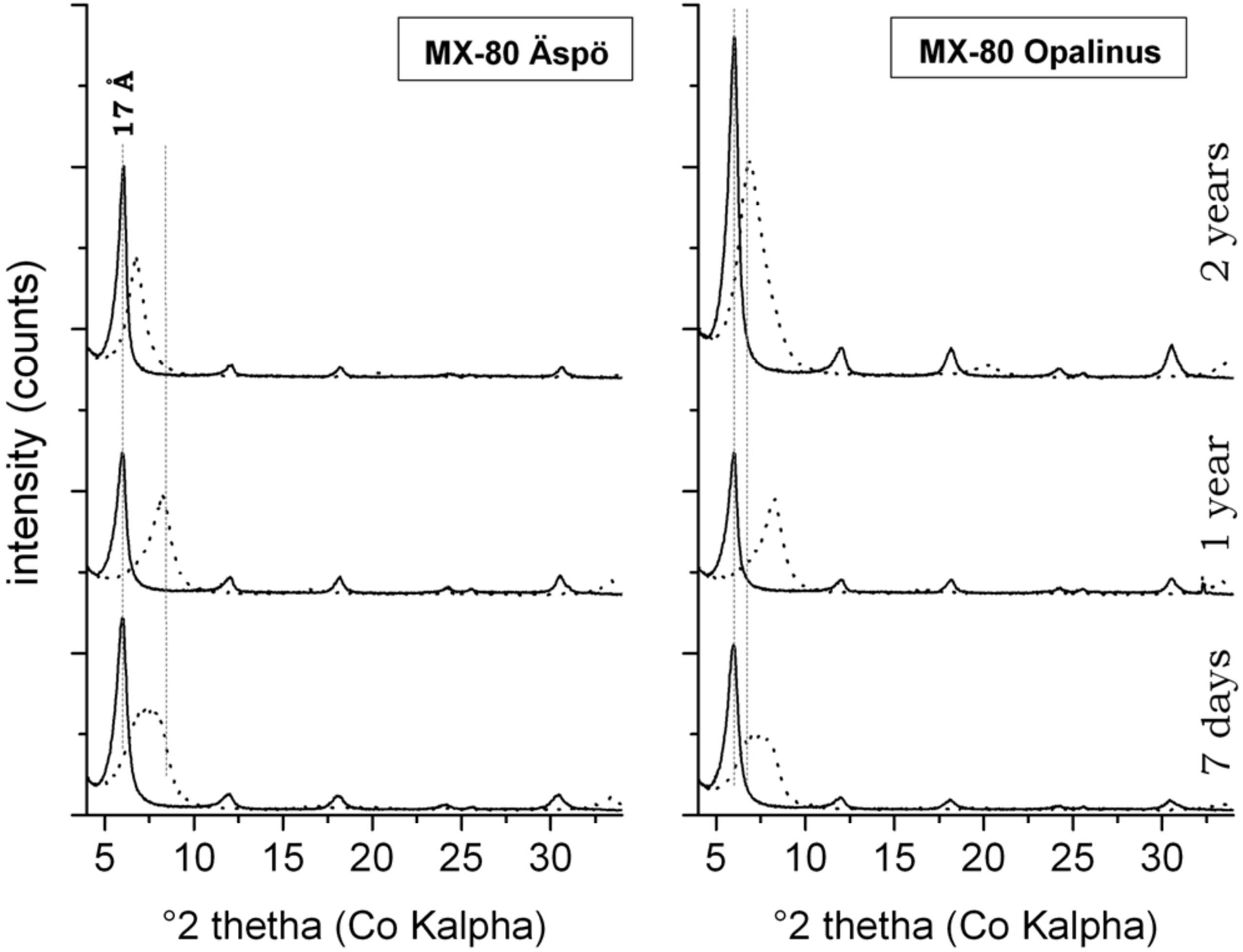
Results – interlayer charge

Development of interlayer charge of montmorillonite – TEM-EDX



Results – mineralogical changes

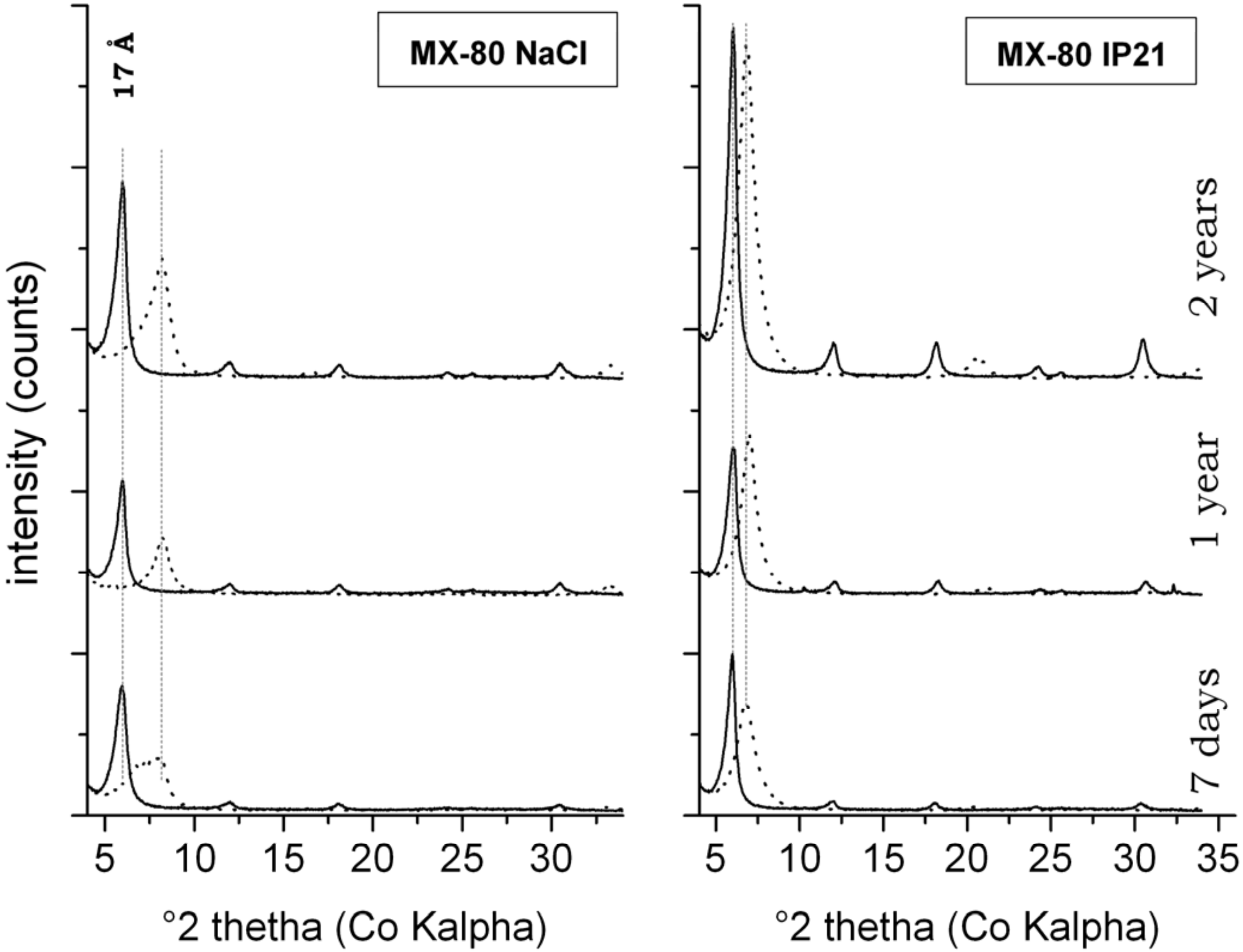
Glycol expandibility



Montmorillonite remains the dominating phase and always full expandable to 17 Å

Results – mineralogical changes

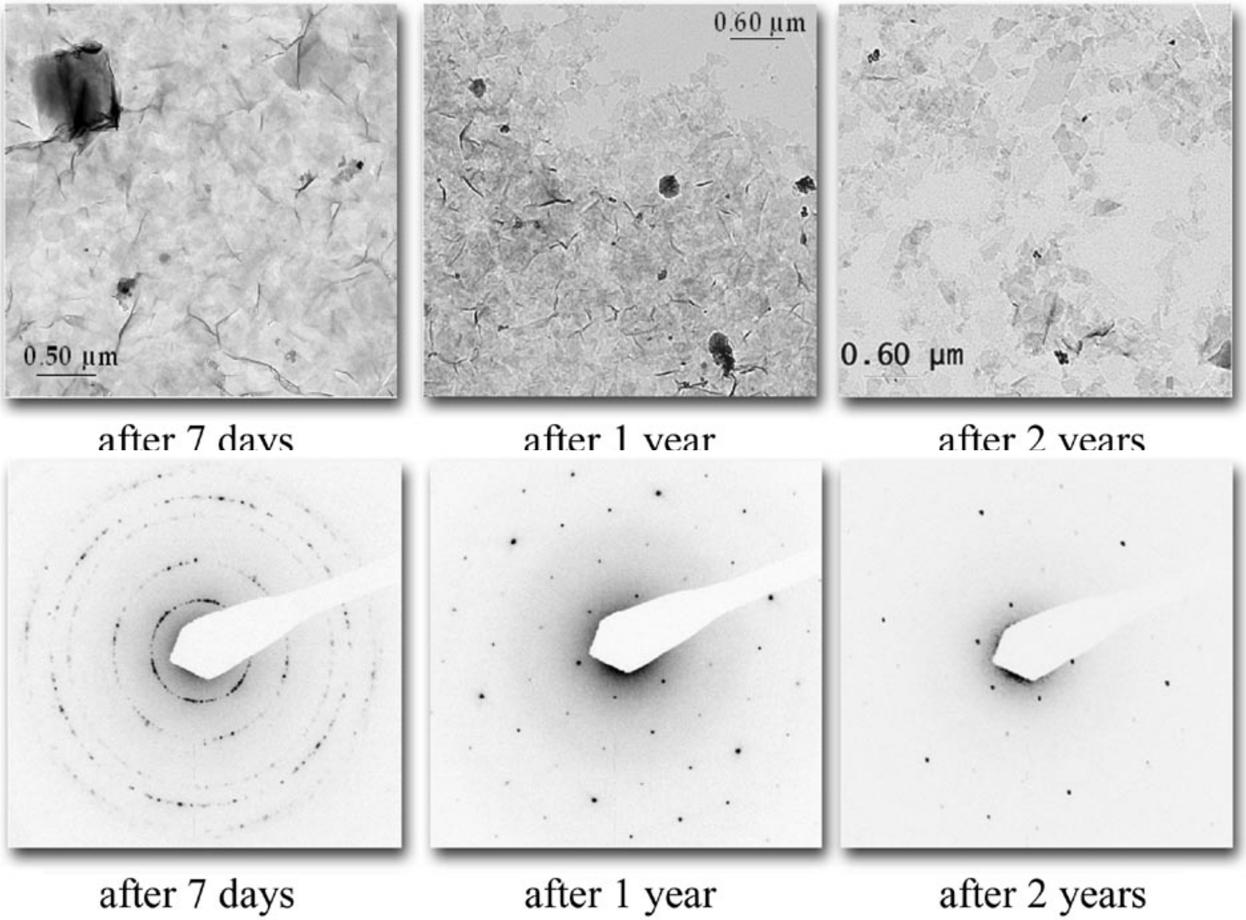
Glycol expandibility



Montmorillonite remains the dominating phase and always full expandable to 17 Å

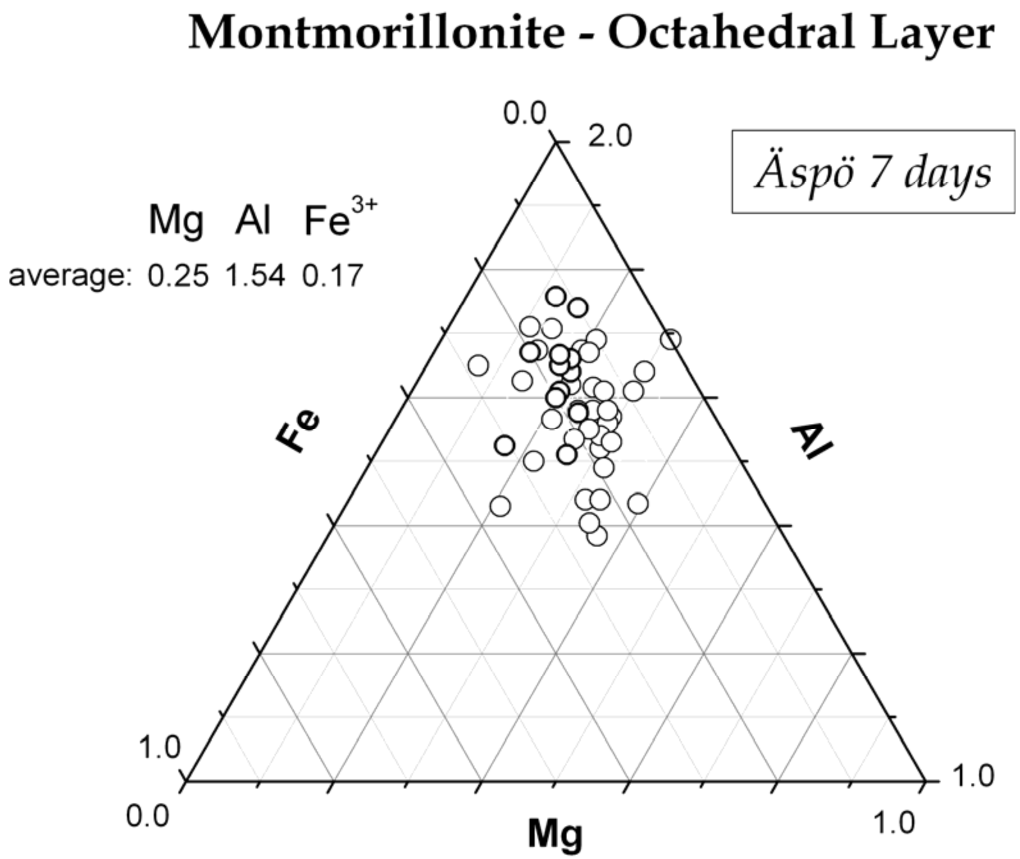
Results – mineralogical changes

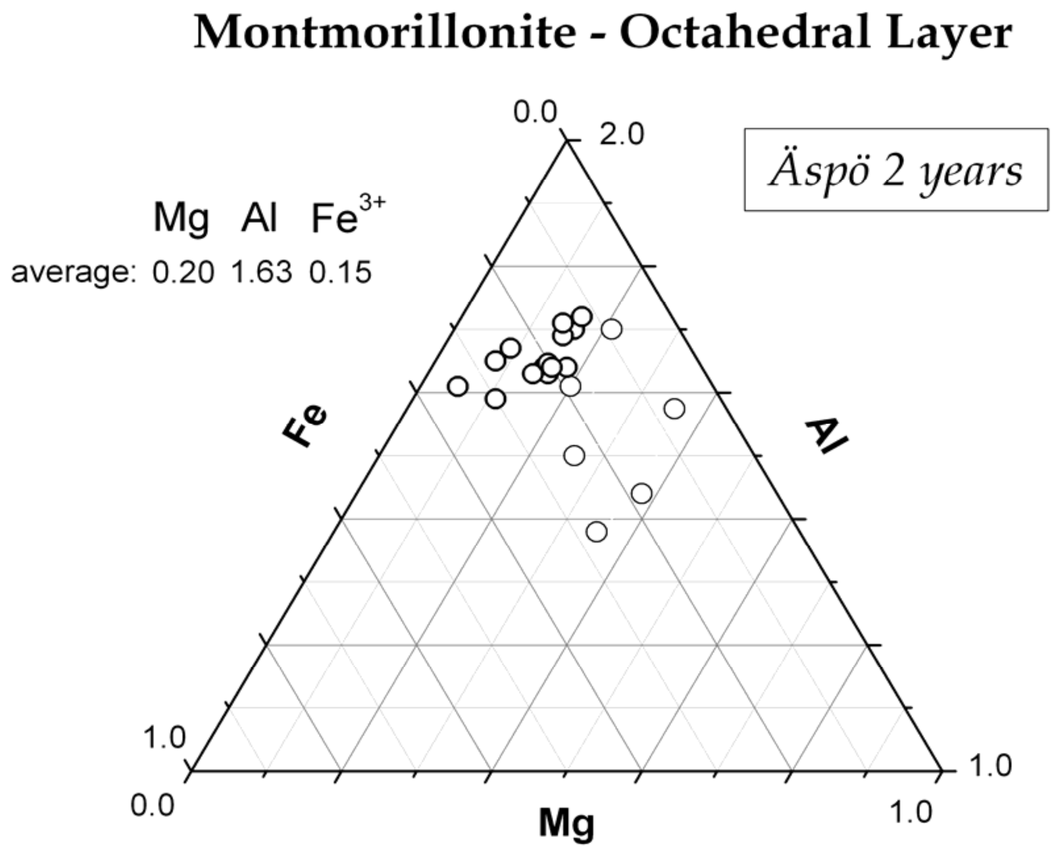
Changes in morphology and orientation of particles



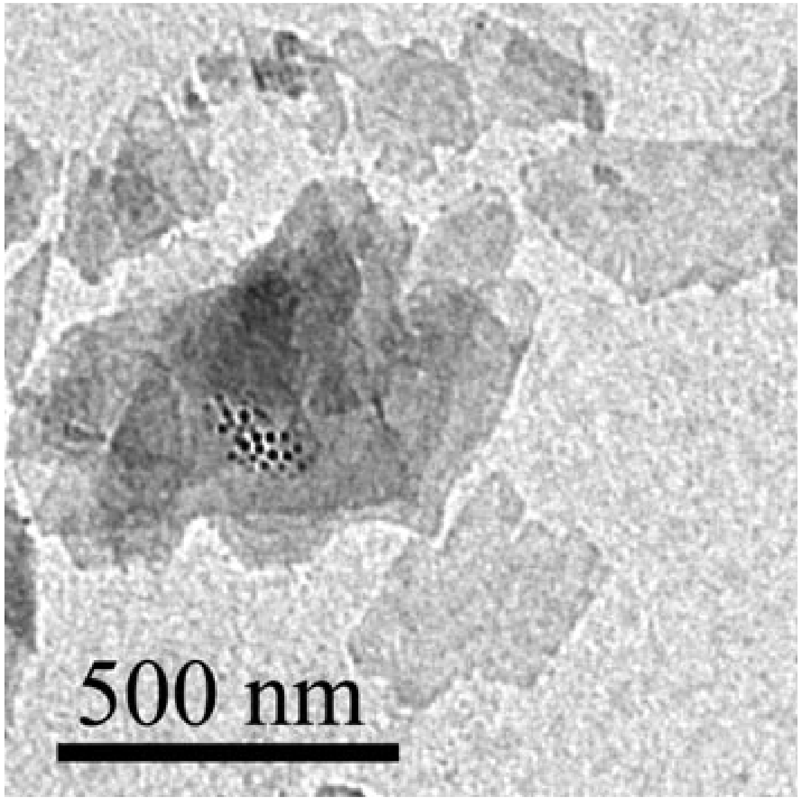
Results – mineralogical changes

Changes in octahedral layers – substitution of Mg by Al

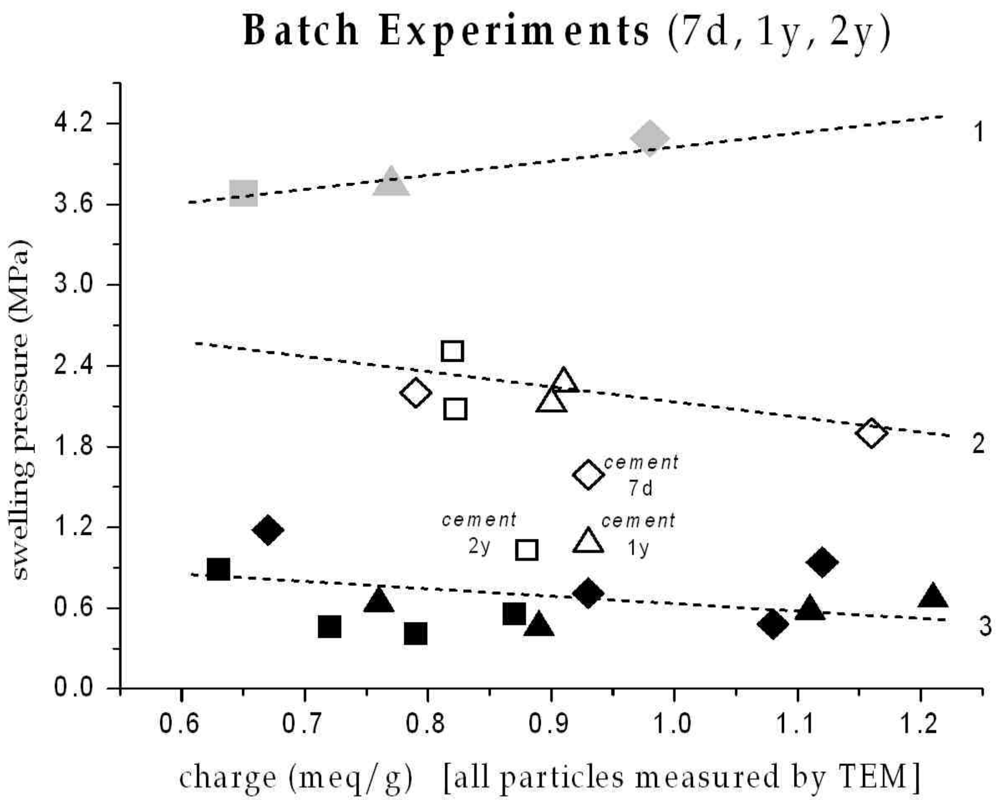


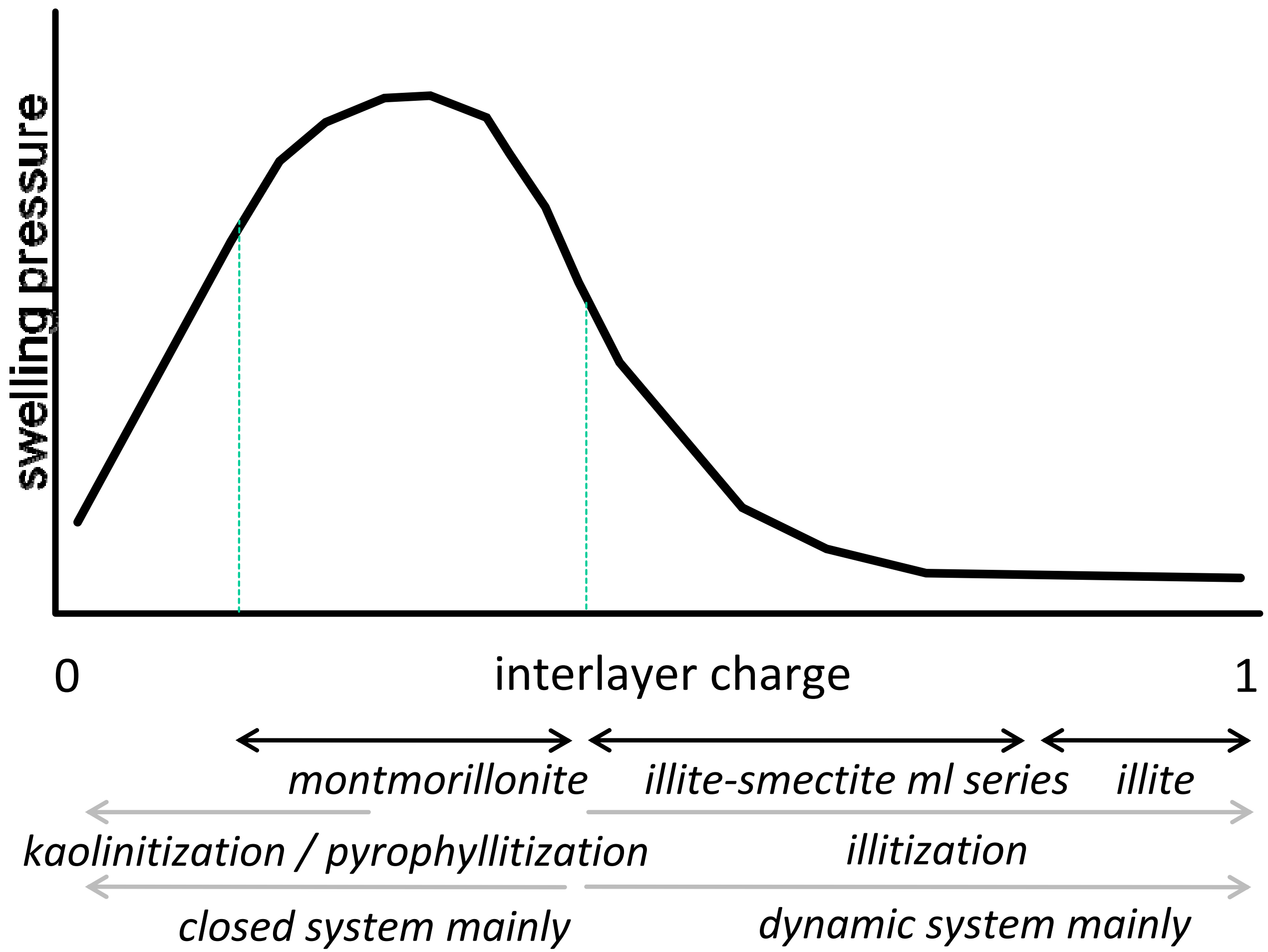


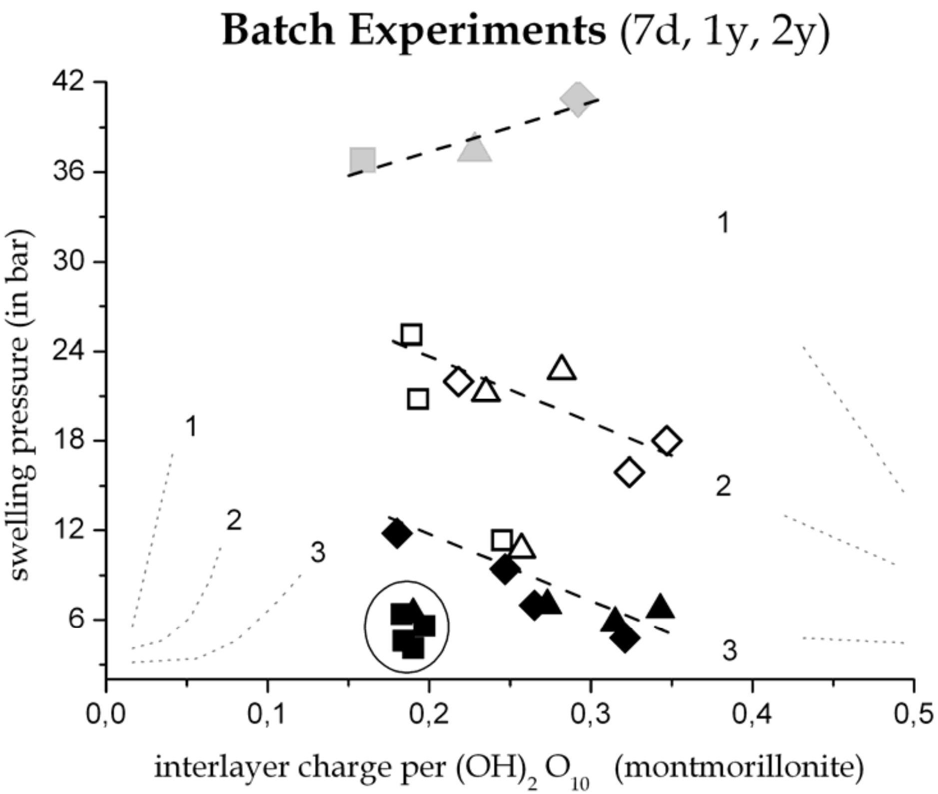
Results – mineralogical changes
Frmation of new minerals

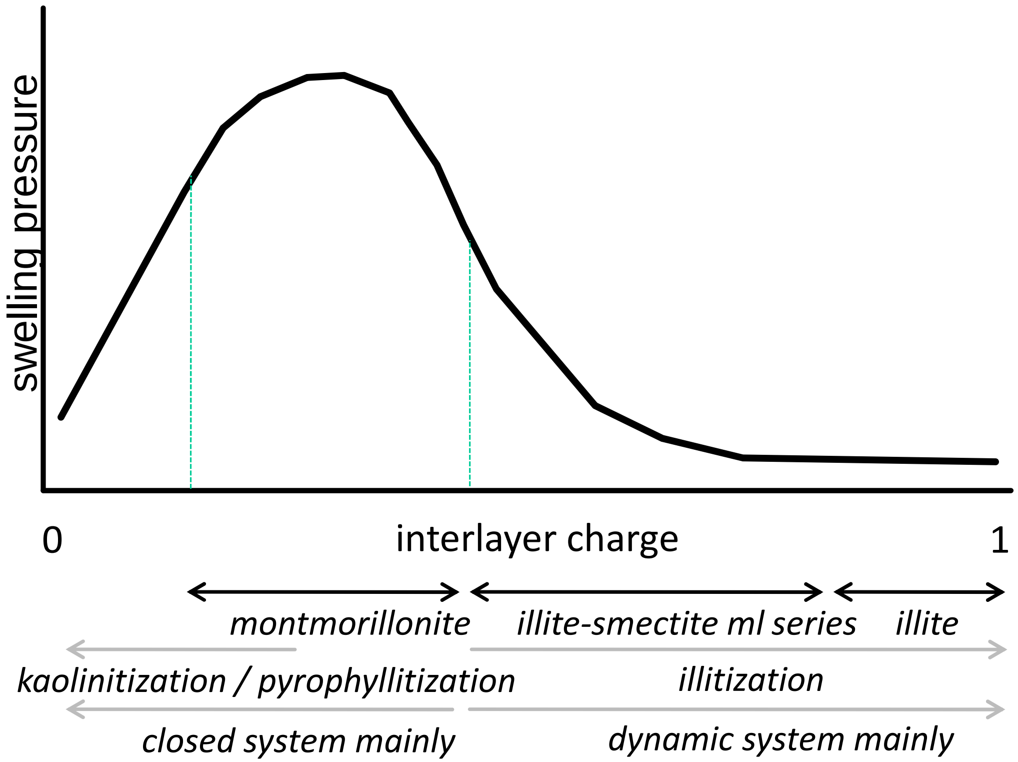


TEM-micrograph with hypidiomorphous particles, kaolinite like habitus (sample IP21, 2 years, compaction experiments)


Batch Experiments (7d, 1y, 2y)
swelling pressure (MPa)
4.2
3.6
3.0
2.4
1.8
1.2
0.6
0.0
0.6
0.7
0.8
0.9
1.0
1.1
1.2
charge (meq/g) [all particles measured by TEM]
1
2
3
cement
7d
cement
2y
cement
1y


swelling pressure
0
interlayer charge
1
montmorillonite
illite-smectite ml series
illite
kaolinitization / pyrophyllitization
illitization
closed system mainly
dynamic system mainly


Batch Experiments (7d, 1y, 2y)
swelling pressure (in bar)
42
36
30
24
18
12
6
1
2
3
1
2
3
0,0
0,1
0,2
0,3
0,4
0,5
interlayer charge per (OH)2 O10 (montmorillonite)


swelling pressure
0
interlayer charge
1
montmorillonite
illite-smectite ml series
illite
kaolinitization / pyrophyllitization
illitization
closed system mainly
dynamic system mainly

Summary and conclusions

•The interaction of waters of different salinities with MX-80 has minor effects on the mineralogical composition of the bentonite, with minor amounts of montmorillonite being dissolved leading to the precipitation of kaolinite/pyrophylite.

•The most relevant changes occur in the octahedral layer of smectites, where Mg is replaced by Al.

•This replacement has two effects:

- 1) a decrease of the interlayer charge; and
- 2) an increase in Mg in solution leads to the replacement of Na by Mg in the interlayer space.

•The swelling pressure of bentonite decreases with the ionic strength of reacting waters, being a correlation between the interlayer charge and swelling pressure observed.

Summary and conclusions (continued)

- High ionic strength solutions seem to accelerate (because of the higher acidity of the interlayer water) the transformation rate.
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- In compacted bentonites the amount of water with higher acidity is higher which increases the transformation rate even more.

- The expected kaolinitization/pyrophyllitization and Si-excess may lead in the long run to a significant or even total loss of swelling capacity of compacted bentonites, if high saline solutions and cement are involved.

- Therefore the question which must be answered is not, whether or not bentonites are stable under the conditions of a repository in salt, but rather, how much swelling capacity is needed and for how long.

- So far the practical relevance of the possible collapse of the SWP due to long term mineral transformation has not yet been finally decided.

- Still open is also the quantitative relationship: alteration – SWP – permeability.