UNDERSTANDING THE MECHANISMS OF CHEMICAL EROSION OF BENTONITE – NEED FOR A HOLISTIC APPROACH

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Contents

Background

- Chemical erosion and its implications need to be assessed in performance assessments (PA)
- To do this, a conceptual model(s) is needed
- Conceptual model is a basis for PA modelling
- Conceptual model can include, in addition to the process chemical erosion, also other relevant and interconnected processes (such as colloidal behaviour, sedimentation etc.)

Background

- Additional to this discussion (this presentation), also modelling development as well as groundwater evolution considerations are needed to fully assess chemical erosion in PA

Sources of information

- Currently we use knowledge based on:
  - Limited amount of small scale experiments
  - Modelling based on the experimental results
- What is missing:
  - State-of-the-art review on chemical erosion in repository environment (past, present and future)
  - Review on chemical erosion considering also simultaneous processes – coupling!
  - Additional experiments to fill the gap between simplified systems and the expected system in the repository
  - URL results – will we get anything from CFM?
  - Detailed knowledge on the natural smectite stability at repository (or scenario) relevant environments

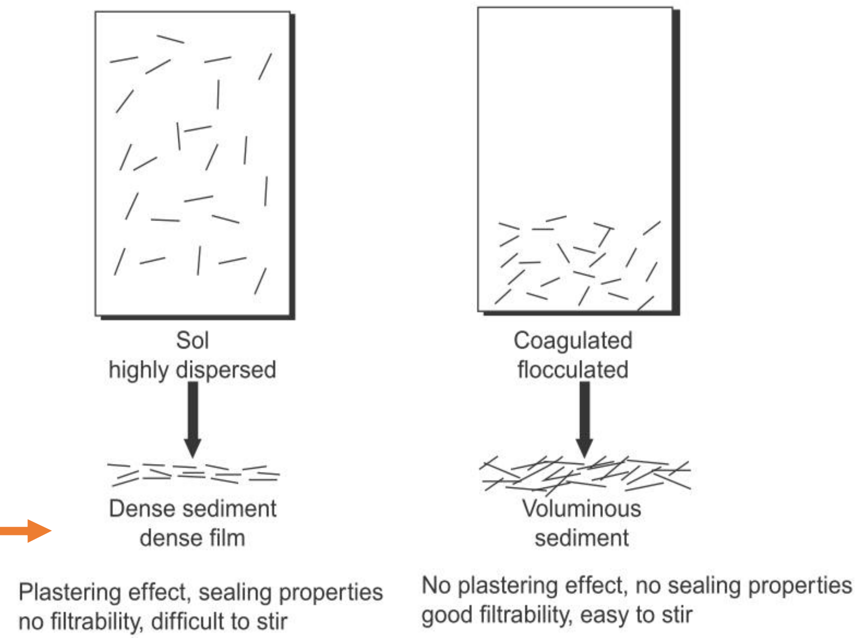
Sources of information

FEW EXAMPLES

Sources of information - mechanisms

1. Sedimentation

- Bentonite behavior has been extensively studied within geotechnical sciences and colloid sciences
  - Colloid science combined the concepts of dispersion, stability of dispersions, aggregation and sedimentation
  - What type of filter cakes are expected in repository environment?



Two types of sedimentation defined for well dispersed clay colloids (left) and aggregates (right) (Lagaly and Dékány 2013).

2. Gravity and available volume

- Gravity has a profound effect for erosion in vertical or inclined fractures (cf. lab experiments)
- Gravity also helps sedimentation
- The fracture volume in natural rock is not infinite → sedimentation will occur – fractures becomes more sparse with depth and their apertures are smaller
- Salinity also increases with depth → colloids destabilisation
- Aim for a conservative approach, but not unrealistic!

Sources of information - mechanisms

3. Filter cake formation

- Geotechnical sciences use the properties of bentonites
    - Filter cakes formation is basis for bentonite drilling slurries performance
    - This information should be reviewed from the safety case point of view

Sources of information - mechanisms

4. Water compositions – natural vs. simulant

- The experimental set ups currently used to study chemical erosion are overly simplified (cf. Na/Ca Cl solutions).
- Natural waters are more complex and buffered by the host rock
- Can we describe smectite stability in dilute conditions only using cation concentration?
- Few examples of ground and Allard waters:

Parameter	Unit	RO (min)[1]	RO (max)[1]	KI (min)[1]	KI (max)[1]	Grimsel[2]	Allard-MO[3]	Allard-MR[3]
TDS	mg/L	61.7	189.9	133.2	240.2	83.0	~220	~220
pH	field	6.8	9.3	7.2	8.5	9.6	8.4	8.8
Eh	mV	-330.0	230	-350.0	400.0	-200.0		
O2	mg/L	0.0	1.1	0.0	0.4			
Na	mg/L	4.3	43.2	4.7	27.0	15.9	52.5	52.5
K	mg/L	0.7	3.0	0.2	2.4	0.2	3.9	3.9
Ca	mg/L	2.7	23.0	11.2	33.0	5.2	10.2	5.1
Mg	mg/L	0.1	5.3	0.9	8.6	0.015	2.8	0.7
Fe(tot)	mg/L	0.0	3.0	0.0	1.4	0.0002		
Cl	mg/L	0.9	7.4	1.0	5.8	5.7	47.5	48.8
HCO3	mg/L	0.0	123.3	82.4	158.6	27.45	90.7	65.0
SO4	mg/L	1.0	6.5	0.1	3.5	5.9	9.6	9.6
SiO2	mg/L					15.0	2.9	1.7
Sr	mg/L					0.18		
Mn	mg/L					0.0003		
F	mg/L					7.2		

1) McEwen & Äikäs 2004

2) as given in Hellä et al. 2014

3) Vuorinen & Snellman 1998

Sources of information - mechanisms

4. Water compositions – natural vs. simulant

- Effect of other cations?
- Effects of anions, esp. HCO_3 and sulphate?

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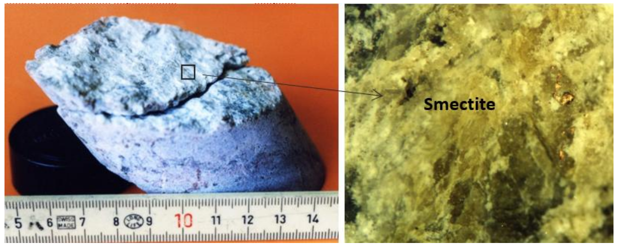
Sources of information - mechanisms

5. Fracture smectites

- Why do we find smectites in dilute conditions?
- Natural smectites could at least tell about:
  - How old thay are? (in situ?)
  - How do they occur? Differences depending on parageneses, smectite+kaolinite, smectite + calcite etc.
  - Are they all exchanged/originally Ca-form?
    - Samples from open and closed fractures
  - Isotopic signatures of groundwater-rock interaction?

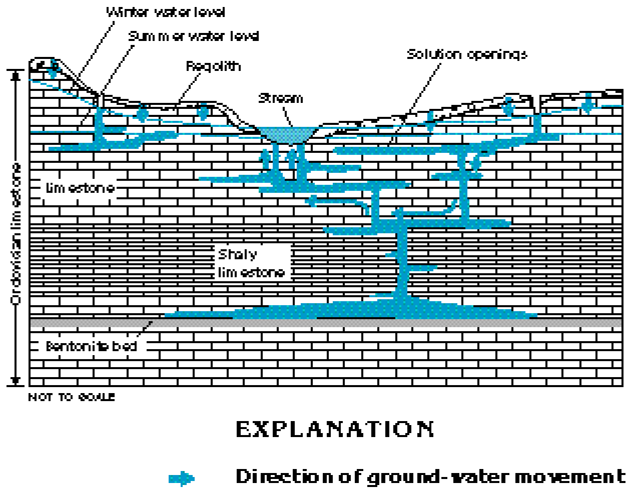


Sources of information - mechanisms

6. Bentonites

- Bentonite deposits located adjacent to water-conducting sedimentary rocks (limestone, sandstone etc.) could provide an answer to the question:
  - what changes are seen in bentonite when fresh water flows next to it?
    - Cation exchange reactions in bentonite deposits
    - Rate of exchange (when local hydrogeological history is known)
    - Do bentonites intrude in the adjacent sediments/fractures in any circumstances?

An example how bentonites function in limestone aquifers:



Modified from Zurawski, Ann, 1978, Summary appraisals of the Nation's ground-water resources—Tennessee region: U.S. Geological Survey Professional Paper 813–L, 35 p.

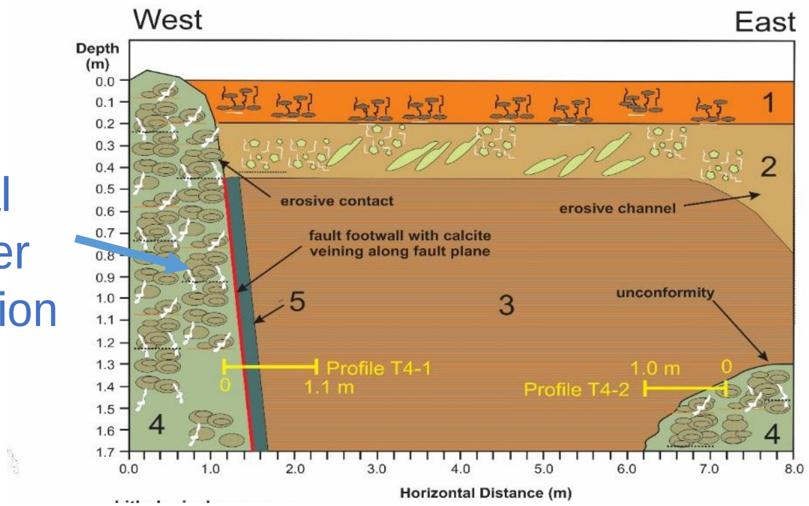
Figure 97. The limestone and dolomite aquifers contain small quantities of insoluble material and, therefore, produce only a thin layer of residuum when weathered. Recharge water percolates through the thin layer of surface material, called regolith, and subsequent-ly moves through vertical frac-tures and horizontal bedding planes in the rocks. The slightly acidic water dissolves some of the limestone and dolomite as it moves to streams and other areas of discharge, such as springs and wells. The vertical movement of the recharge water and, therefore, the depth of de-velopment of solution openings, are restricted by zones of low permeability.

Sources of information - mechanisms

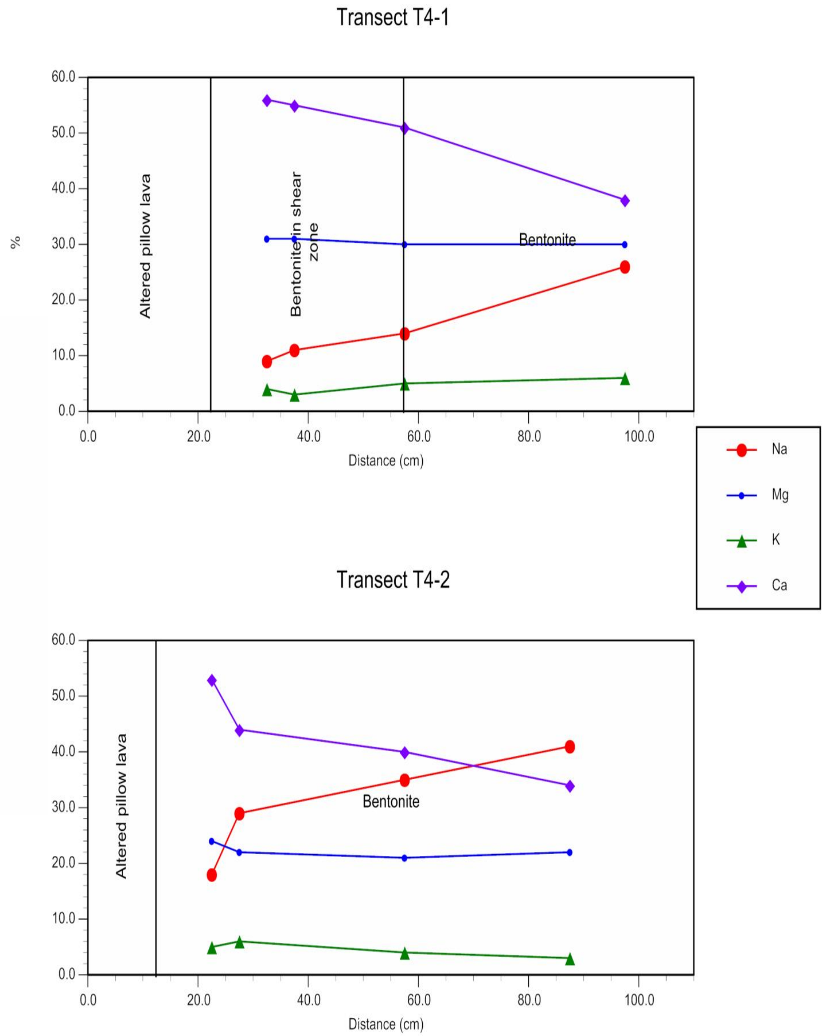
6. Bentonites

An example of cation exchange at a bentonite pillow lava contact surface:

Local aquifer formation



- chlorine concentration 2.4mM and the total charge equivalent of main cations is about 7mM



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Conclusions

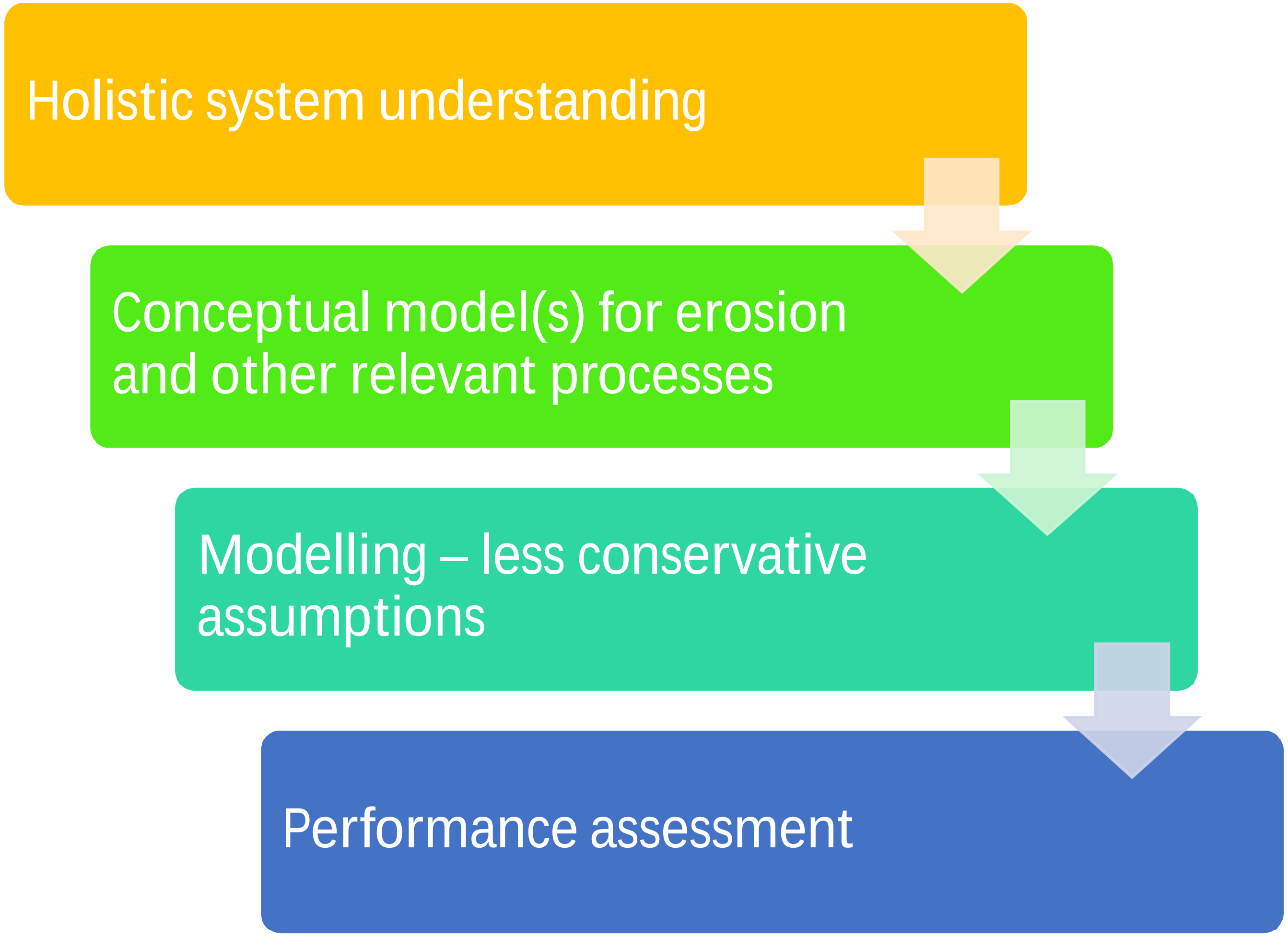
- A more holistic approach is needed to perform a reliable assessment of the bentonite stability in repository conditions.
- The mismatch between laboratory experiments and observations in nature is due to a lack of understanding on the overall parameters and processes affecting the erodability of smectite and how these interact with each other.

Main implications:

1. The information in the already existing scientific literature should be considered more thoroughly
2. The experimental set ups to study chemical erosion should cover also more complex groundwater compositions (accounting for more complex anion/cation compositions) and smectite/bentonite compositions (including presence of various types of accessory minerals)
3. The scaling effects of the process should be better understood when moving from laboratory scale to in-situ conditions. The lack of data at larger scale from underground research laboratories is an obvious challenge.
4. Natural montmorillonites should be further studied to understand their mode of occurrence as stable phases in repository-relevant environments.

Components for holistic analysis – what to do next:

From system understanding to PA



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