Coupled THMC interactions at temperature beyond 130 °C : experimental investigations and modelling of materials alteration, swelling processes and overpressure build-up

F. Claret, S. Gaboreau, J. Tremosa and C. Tournassat

Géosciences pour une Terre durable

brgm

CONTEXT

Available online at www.sciencedirect.com
ScienceDirect
Physics and Chemistry of the Earth 32 (2007) 780–788
PHYSICS and CHEMISTRY of the EARTH
www.elsevier.com/locate/pce

Performance of the bentonite barrier at temperatures beyond 100 °C: A critical review

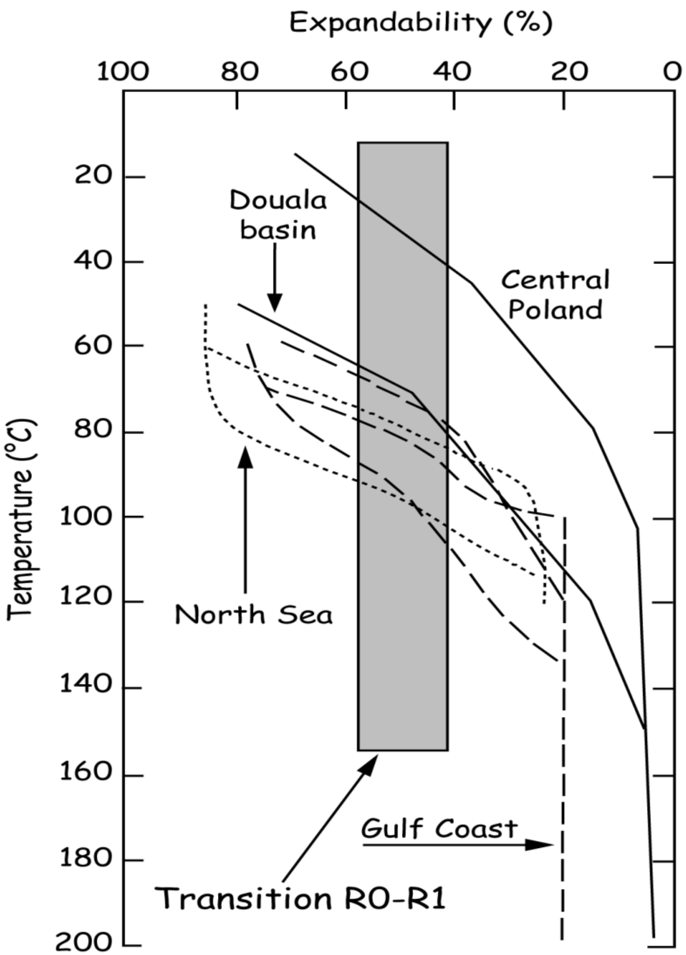
P. Wersin *, L.H. Johnson, I.G. McKinley

National Cooperative for the Disposal of Radioactive Waste (Nagra), Hardstrasse 73, 5430 Wettingen, Switzerland

Received 15 April 2005; received in revised form 17 January 2006; accepted 23 February 2006
Available online 9 October 2006

> Lack of reliable information at temperatures beyond 130°C regarding hydraulic, mechanical and mineralogical changes

Mineralogical transformation: Illitisation?



Modified from Srodon et Eberl (1994)

American Mineralogist, Volume 96, pages 207–223, 2011

A reinvestigation of smectite illitization in experimental hydrothermal conditions: Results from X-ray diffraction and transmission electron microscopy

ERIC FERRAGE,[1,2,*] OLIVIER VIDAL,[3] RÉGINE MOSSER-RUCK,[4] MICHEL CATHELINEAU,[4] AND JAVIER CUADROS[1]

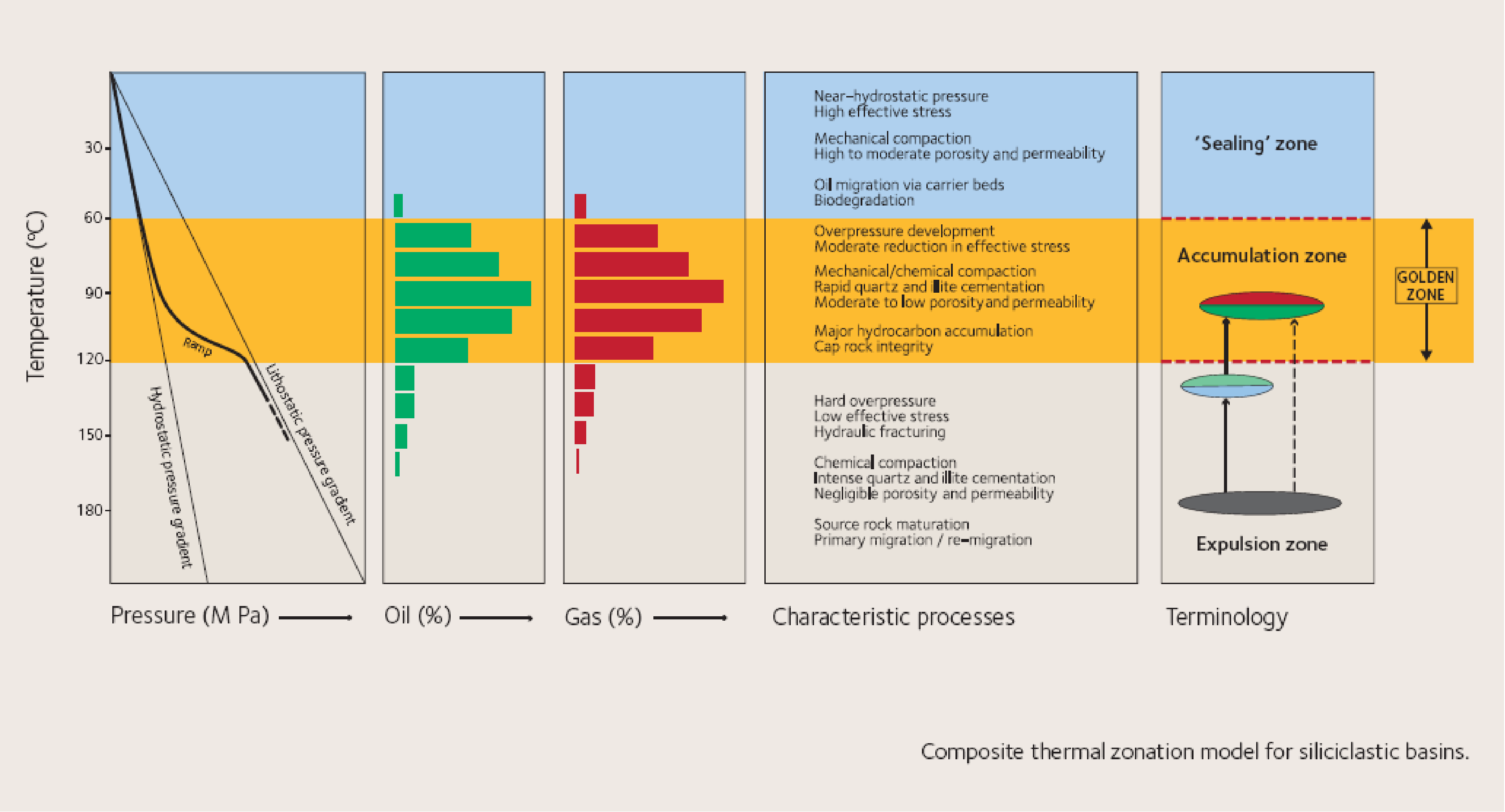
[1]The Natural History Museum, Department of Mineralogy, Cromwell Road, London SW7 5BD, U.K.
[2]Laboratoire Hydrogéologie, Argiles, Sols et Altérations, UMR6269-CNRS, Université de Poitiers, 40 avenue du Recteur Pineau, 86022 Poitiers Cedex, France
[3]LGCA, UMR5025-CNRS, Université Joseph Fourier Grenoble, 1381 rue de la Piscine, BP 53, 38041 Grenoble Cedex, France
[4]UMR G2R 7566, CREGU, Université Henri Poincaré, BP 239, F-54506 Vandoeuvre-lès-Nancy Cedex, France

"However this treatment overestimates the amount of illite layers because of the presence of smectitic non-expandable layers. This was revealed by calcium exchange of the products, which causes re-expansion of the apparent illite layers"

Are only clay concerned by the temperature issue?

- **In some concept, the canister is not surrounded by bentonite but by *cementitious materials* in order to reduce the corrosion rate. Hydraulic and mineralogical changes of these cementitious materials at elevated temperatures could be detrimental to the safety of radioactive waste disposal, and they certainly need to be investigated**

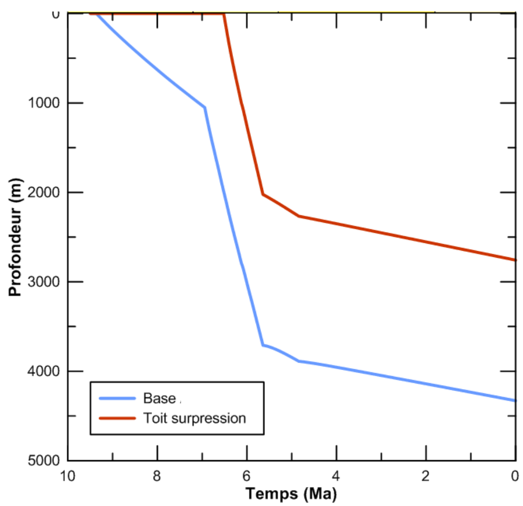
Overpressure development



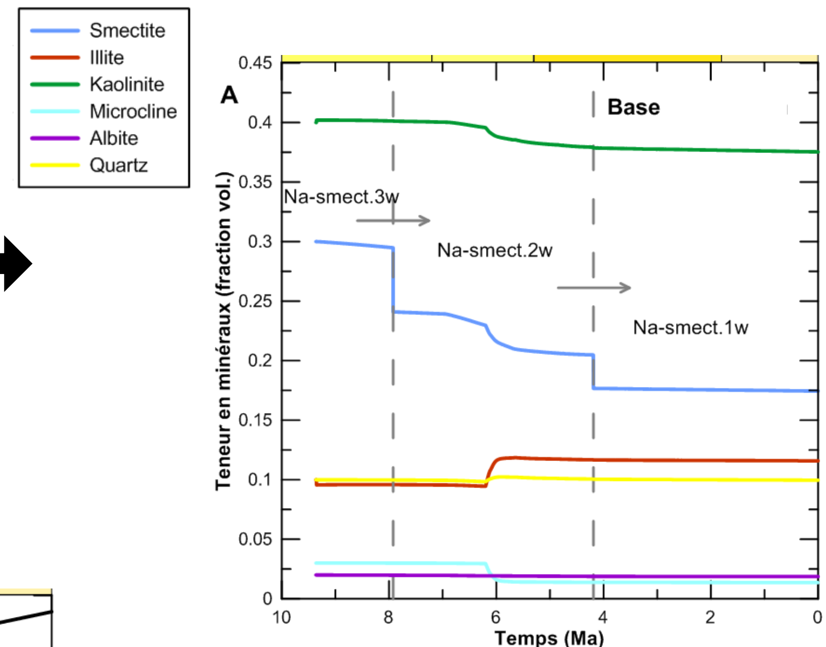
Composite thermal zonation model for siliciclastic basins.

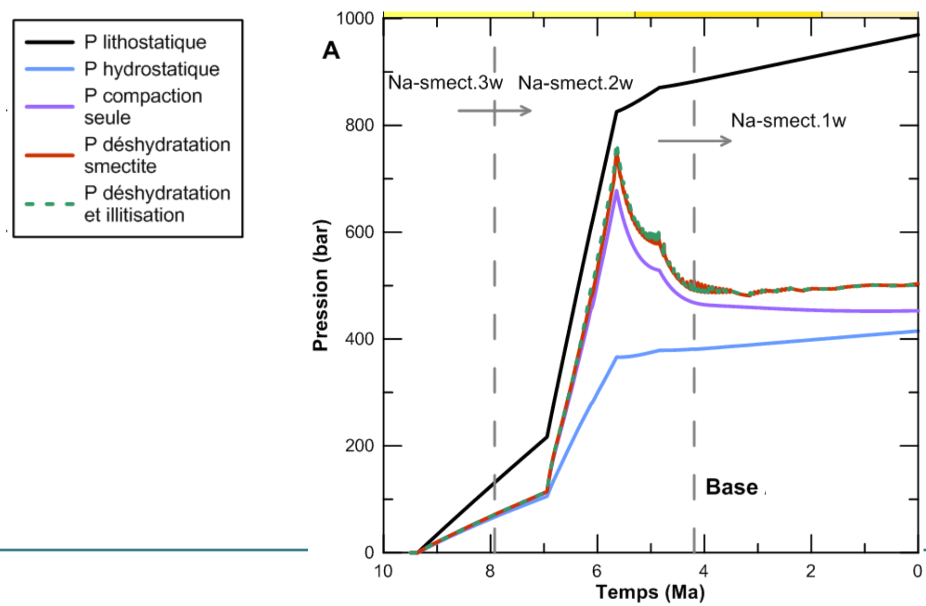
Modified from Nadeau Clays Minerals V46 (2011)

Burial of a sediment leading to porosity decrease and temperature increase



Diagenetic evolution including smectite deshydration (5 to 135°C)

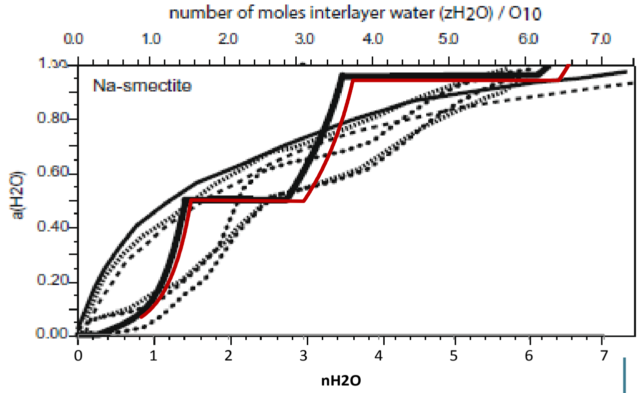




Coupled effect on pore pressure

Modelling the thermo-hydro-mechanical evolution of a clayrock together with smectite deshydratation

- An example of clayey sediment burial and diagenesis (smectite deshydration and smectite to illite transformation) with feedback on the pore pressure in a passive margin
- Smectite deshydration through a solid solution model, leading to fluid expulsion in the porosity
- Fully coupled with the porosity evolution due to compaction and the related fluid circulations (Python-Phreeqc program developped @ BRGM)
- Such modelling can be applied to the near-field evolution of a nuclear waste storage, during the thermal transient phase



How much thermo-osmosis flow is contributing to the pressure regime and the water flow in a nuclear waste storage?

- **Thermo-osmosis is a water flow under a temperature gradient. It is due to modifications of the properties of water sorbed on the clay surfaces**
- **Possible effect of thermo-osmosis on the flow in a clay-rock formation (Gonçalvès et al., 2012) or in an exothermal radioactive waste disposal (Soler, 2001)**
- **But, it remains a poorly characterized process: thermo-osmotic permeability can be predicted (Gonçalvès et al., 2012), but very few experiments (Tremosa et al., 2010)**

Journal of Colloid and Interface Science

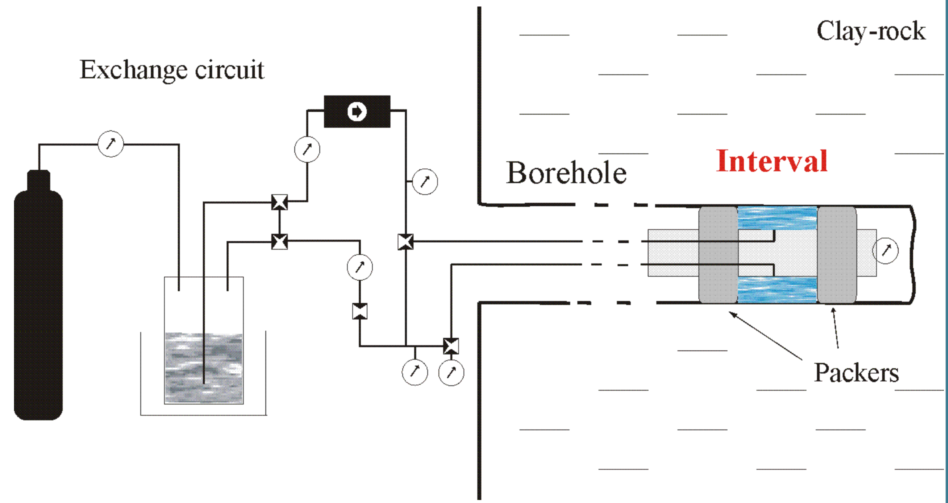
Volume 342, Issue 1, 1 February 2010, Pages 175–184

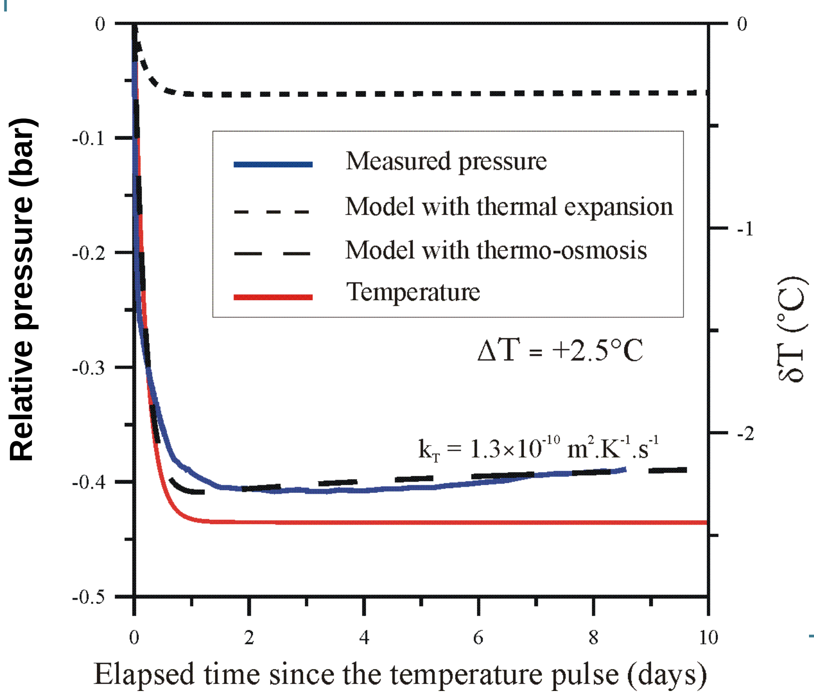
Estimating thermo-osmotic coefficients in clay-rocks: II. In situ experimental approach

J. Trémosa[a, b], J. Gonçalvès[b, c], J.M. Matray[a], S. Violette[b, d]

An illustration of the effect of thermo-osmosis on flow and pressure under a thermal gradient (Tremosa et al., 2010)

- In-situ experiment at Tournemire URL
- Thermal pulse in a borehole interval filled with water
- THM evolution inversion by a numerical model

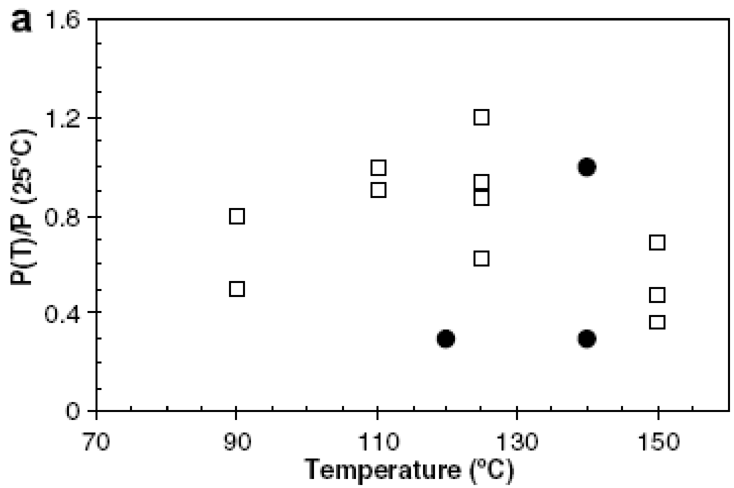




Pressure decrease due to water thermal expansion effect

Pressure decrease due to thermo-osmotic flow and water thermal expansion effect

Impact of temperature on swelling pressure?



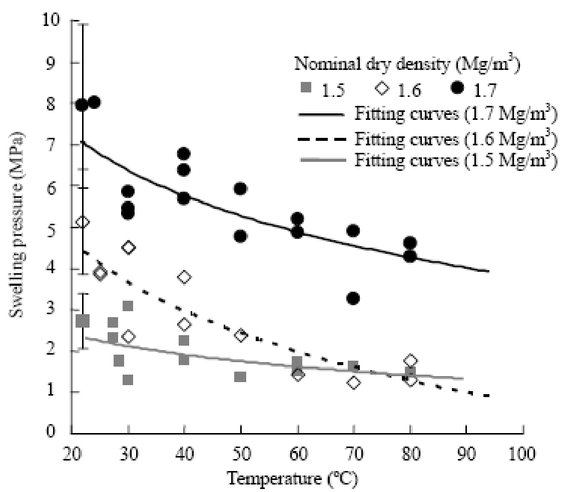


Fig.5 Swelling pressure as a function of temperature for saturated compacted samples.

ELSEVIER

Available online at www.sciencedirect.com

ScienceDirect

Physics and Chemistry of the Earth 32 (2007) 780–788

PHYSICS and CHEMISTRY of the EARTH

www.elsevier.com/locate/pce

Performance of the bentonite barrier at temperatures beyond 100 °C: A critical review

P. Wersin *, L.H. Johnson, I.G. McKinley

National Cooperative for the Disposal of Radioactive Waste (Nagra), Hardstrasse 73, 5430 Wettingen, Switzerland

Received 15 April 2005; received in revised form 17 January 2006; accepted 23 February 2006
Available online 9 October 2006

Journal of Rock Mechanics and Geotechnical Engineering. 2010, 2 (1): 71–78

CSRME

Journal of Rock Mechanics and Geotechnical Engineering

Journal online: www.rockgeotech.org

Experimental investigation into temperature effect on hydro-mechanical behaviours of bentonite

M. V. Villar[1*], R. Gómez-Espina[1], A. Lloret[2]

[1] *CIEMAT, Madrid, 28040, Spain*

[2] *UPC, Barcelona, 08038, Spain*

Received 15 May 2009; received in revised form 10 October 2009; accepted 24 November 2009

Impact of temperature on swelling pressure?

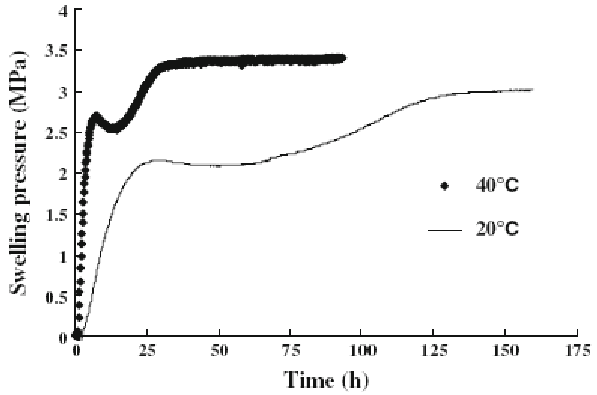


Fig. 3 Evolution of swelling pressure with time at different temperatures

Environ Earth Sci (2013) 68:281–288
DOI 10.1007/s12665-012-1738-4

ORIGINAL ARTICLE

Temperature effects on the swelling pressure and saturated hydraulic conductivity of the compacted GMZ01 bentonite

W. M. Ye · M. Wan · B. Chen · Y. G. Chen · Y. J. Cui · J. Wang

Several assumptions have been made to describe the nonmonotonic swelling behavior of smectite-rich material:

- Pusch (1982) related the swelling pressure peak to the formation of gel structures upon wetting that modify the mechanical characteristics of aggregates/particles (shear strength) and decrease the swelling pressure.
- This has been integrated in the Barcelona Expansive Model (BExM, Alonso; with the progressive reorganisation of the internal structureof samples upon wetting. Progressive filling of the macrostructure(inter-aggregate void ratio decrease) occurred.

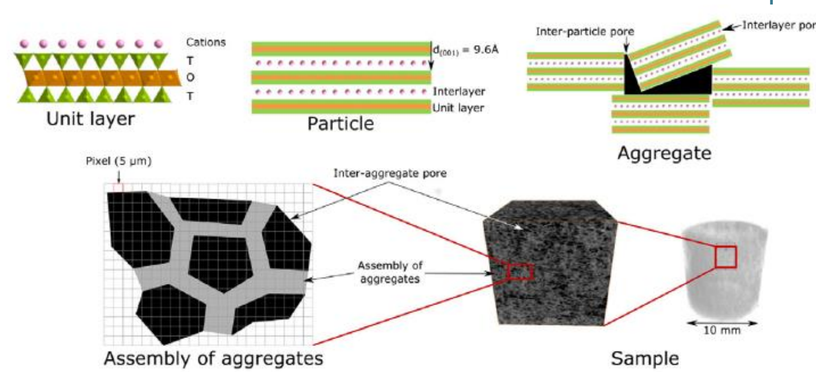


Fig. 1. Organisation of Kunipia-G across the different scales (from unit layer up to centimetre scale).

Development of an oedometer cell transparent to X-Ray to follow inter-aggregate porosity

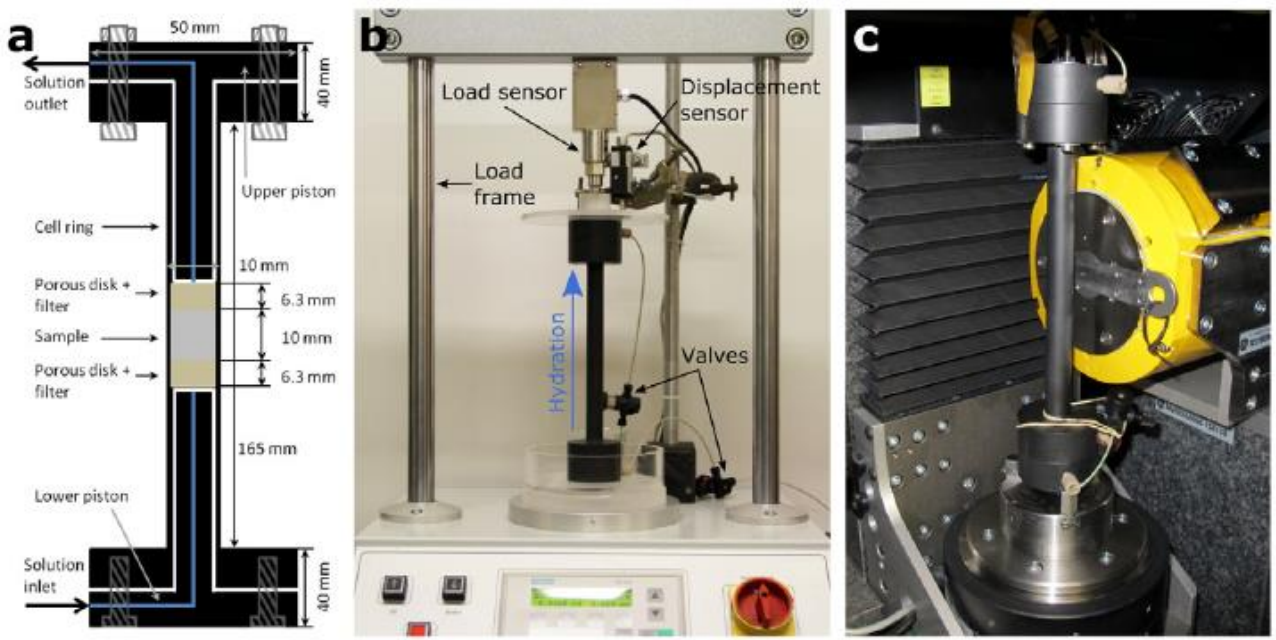


Fig. 3. Representation of the new designed oedometer cell at constant volume: (a) schematic layout, (b) cell with upper piston unscrewed into load frame and (c) cell with upper piston screwed and installed into X-ray tomograph.

Applied Clay Science 124–125 (2016) 197–210

Contents lists available at ScienceDirect

Applied Clay Science

journal homepage: www.elsevier.com/locate/clay

ELSEVIER

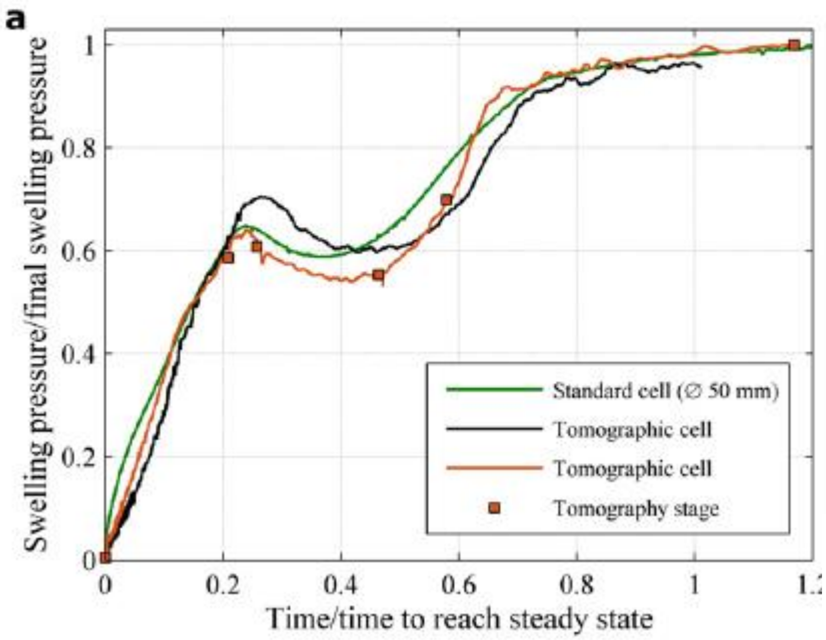
Research paper

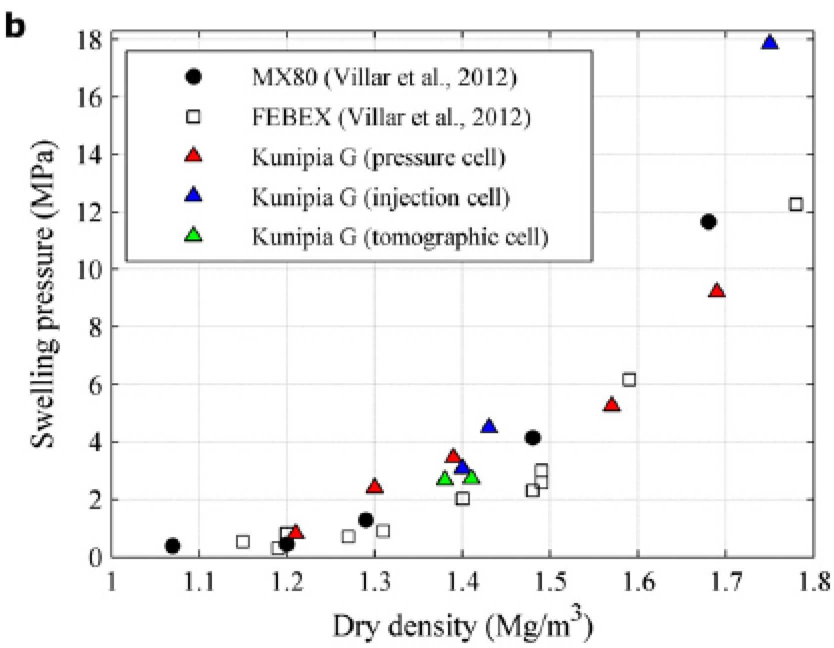
Swelling pressure development and inter-aggregate porosity evolution upon hydration of a compacted swelling clay

CrossMark

Luc Massat [a,b,c], Olivier Cuisinier [a,b,*], Isabelle Bihannic [d,e], Francis Claret [c], Manuel Pelletier [d,e], Farimah Masrouri [a,b], Stéphane Gaboreau [c,**]

Tomography cell gives consistent results





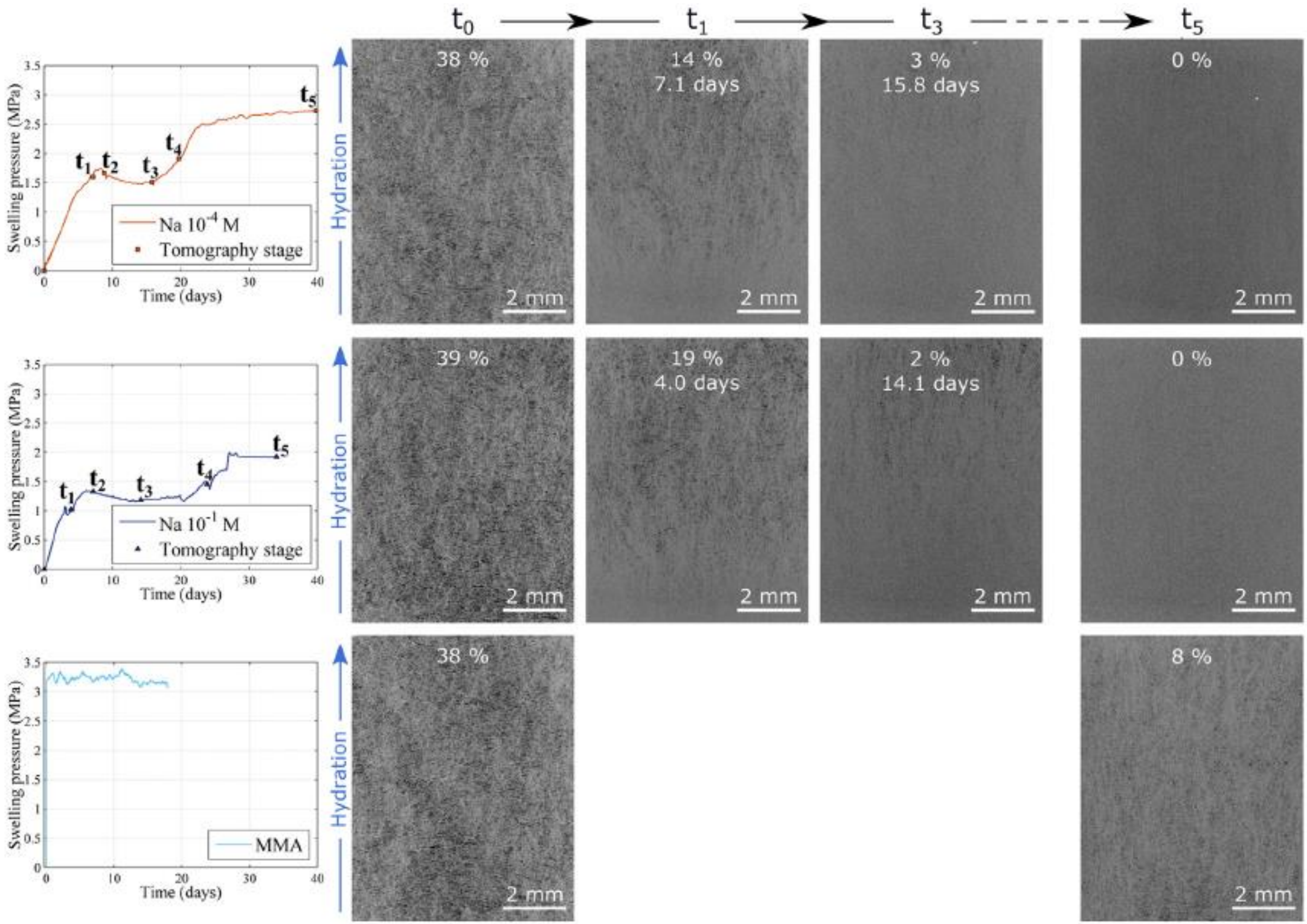
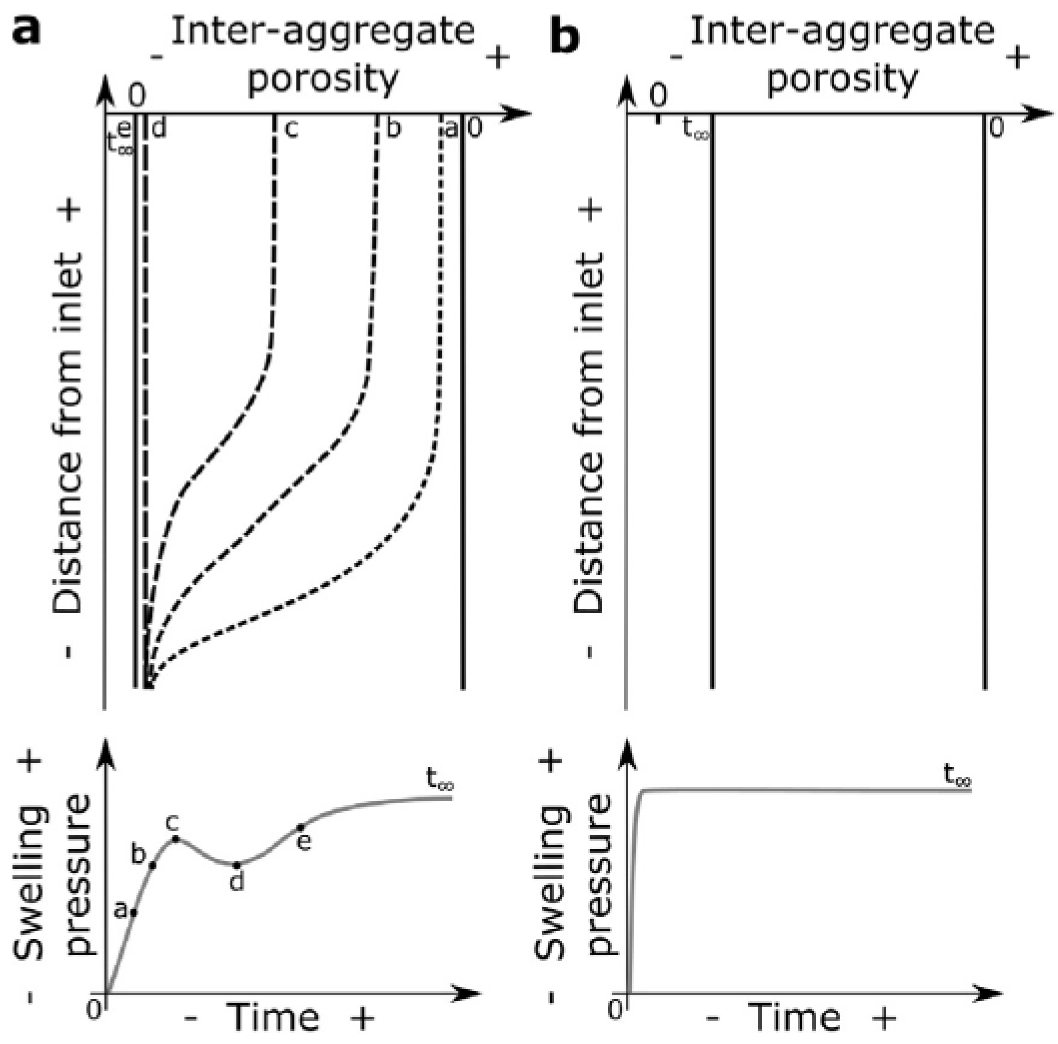
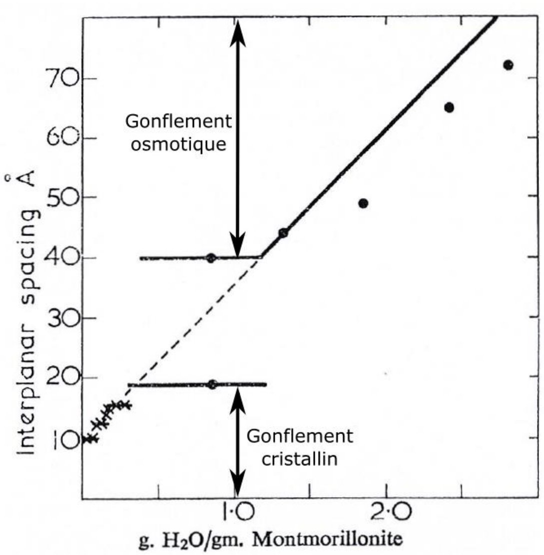


Fig. 8. Inter-aggregate porosity evolution of specimens wetted with NaCl 10^{-4} M, NaCl 10^{-1} M solutions or MMA at specific stages using tomographic cells: X-ray tomography picture, scan time after hydration start, mean porosity.

Inter-aggregate porosity evolution upon hydration



Insights given by other fluid



The main processes that generally operate in concert to control bentonite swelling in aqueous systems are crystalline (interlayer) swelling and osmotic swelling (both double-layer at the inter-particle and the inter-aggregate level);the second process leading to break-up the initial dry particles into thinner ones

Colloids and Surfaces A: Physicochem. Eng. Aspects 434 (2013) 303–318

Contents lists available at SciVerse ScienceDirect

Colloids and Surfaces A: Physicochemical and Engineering Aspects

journal homepage: www.elsevier.com/locate/colsurfa

Prediction of swelling pressures of different types of bentonite in dilute solutions

Longcheng Liu*

Department of Chemical Engineering and Technology, Royal Institute of Technology, S-100 44 Stockholm, Sweden

- The osmotic pressure can be derived by applying the Gouy–Chapman theory according to the following equation issued from Liu (2013) :

$$P_{DDL} = 2cRT(\cosh y^{m} - 1)$$

- ym is the scaled midplane potential at the midpoint between unit layers.
- Depends on the Debye length which is square root dependent on the relative dielectric constant of the pore solution.
- MMA's relative dielectric constant is about 12 times lower than water's (6.32 versus 78.54, see Table 2),
- The osmotic pressure developed with MMA is one order of magnitude lower than that obtained with water (NaCl, both 10−4 and 10−1 M)

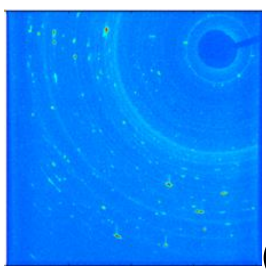
> **NaCl : both crystalline and osmotic swelling**
>
> **MMA : crystalline swelling only**

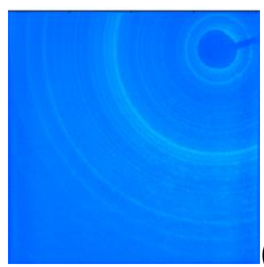
To sum up

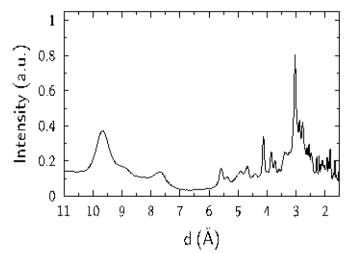
- **Hydration of Kunipia-G specimens by saline solutions induces particle breakup due to osmotic component with major aggregate reorganisation. This could explain partly the inter-aggregate porosity reduction seen by µCT (also confirmed by SEM and TEM).**
- **In contrast, for specimens hydrated by MMA, only crystalline swelling occurs which does not lead to particle breakup, probably explaining the slighter inter-aggregate porosity decrease observed by µCT**

RELEVANCE OF SUCH A DEVICE @ High T?

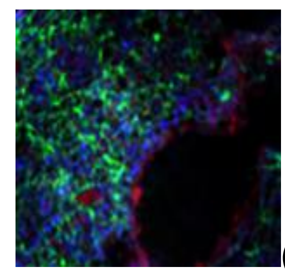
- **Very preliminary design & discussion give us a good confidence in the fact that such a device can be used in temperature (e.g. micro cracking pathway)**
- **Cell design should also allow gas sampling**
- **Complementary technique can be used (e.g. XRD-tomography)**


(a)


(b)



©


(d)

(a) and (b): Details of a single diffraction image (the same image contrast levels have been set) from the cement paste showing the effect of the filtering procedure. (a): Original image; (b): “Filtering” image. (c): Average XRD diffraction pattern. (d): Reconstruction of the distribution of (some) minerals phases in red C-S-H, in green Ettringite, in blue AFm. (Claret et al. in prep)

OTHERS PERSPECTIVES

- **In 2018, an experimental plateform MIMA'ROC will be available @ BRGM**
  - THMC cell coupled to Geophysical tomography (useful for upscaling)

THANK YOU