A chemo-mechanical understanding model for smectite stacks from nanoscopic scale

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Introduction

Deep geological storage is generally considered to be the only viable solution to safely dispose nuclear waste. Different countries have different concepts, but it is widely planned to use smectite based materials, either as buffer material between the host rock and the metallic canister containing the waste, or as seal of the depository gallery. In particular, in the Swiss concept for High Level Waste Storage it is expected to use MX-80 bentonite, principally in the form of compacted pellets. MX-80 bentonite is composed of about 85% of smectite clay (Plötze & Weber 2007). As buffer, the MX-80 pellets will be set up at hygroscopic water content, and then progressively saturated by the water uptake from the host rock. In a first time, MX-80 pellets are foreseen to expand in the different technological gaps, ensuring therefore the closing of preferential flow paths and the overall low-permeability of the buffer, limiting the ionic transport to diffusion. In a second time, the pressure inside the buffer will rise due to the swelling restriction, possibly up to several MPa. This so-called swelling pressure should however not exceed a critical value that will endanger the integrity of the host rock (Sellin & Leupin 2013). It is therefore a key point for the overall safety of the repository to understand the different mechanisms at play during bentonite hydration and model them properly. The goal of the presented work is to investigate these mechanisms at the scale of the water Clay-Ions-Water system, i.e. the nanoscopic scale, to obtain an understanding model of the clay stacks mechanical behaviour.

Clay stacks modelling from nanoscopic scale interactions

The extensive swelling and pressure build-up in compacted bentonite during hydration is principally due to the swelling of the smectite stacks, itself caused by the electrochemical interaction between the smectite platelets, the adsorbed water and the dissolved ions. Cations are attracted in the interplatelets space because of the negative charge carried by the smectite platelets, around 0.12 $C.m^{-2}$ (Karnland et al. 2006), while anions are repelled.

In compacted states, smectite stacks have a *mille-feuille* structure, and the interlayer spacing between smectite platelets is determined by the water molecular diameter (around 0.3 nm) and is usually around one, two or three water diameter(s). Upon hydration, the water intrusion is discrete and done layer by layer. This mechanism is often called crystalline swelling and is the main mechanism at play during clay stacks hydration. Because of the interaction with the smectite platelets, the pressure of the water adsorbed in the interlayer space varies from the one in bulk homogeneous water. This pressure difference called hereafter disjoining pressure, can be determined by computing the water density profiles in-between two smectite platelets. This is done using Integral Equation Therory. The details of the method used, as of the interactions pair potential adopted and the numerical implementation done, are not precised in this abstract, but will be given in a future publication of the authors (in preparation).

The computed disjoining pressure – for a temperature of 300K – is displayed as a function of the basal spacing between smectite layers in figure 1. This curve represents the mechanical behaviour of a single interlayer space. The clay stacks *mille-feuille* is made of tens of them piled together. To interpret the single interlayer pressure curve and to generalise it to clay stacks, the associated swelling free energy is computed. It highlights the role and importance of the cation during the hydration and swelling mechanisms. The final result is an average layer spacing – pressure relationship that exhibits, among two of its most interesting features, hysteresis and free swelling.

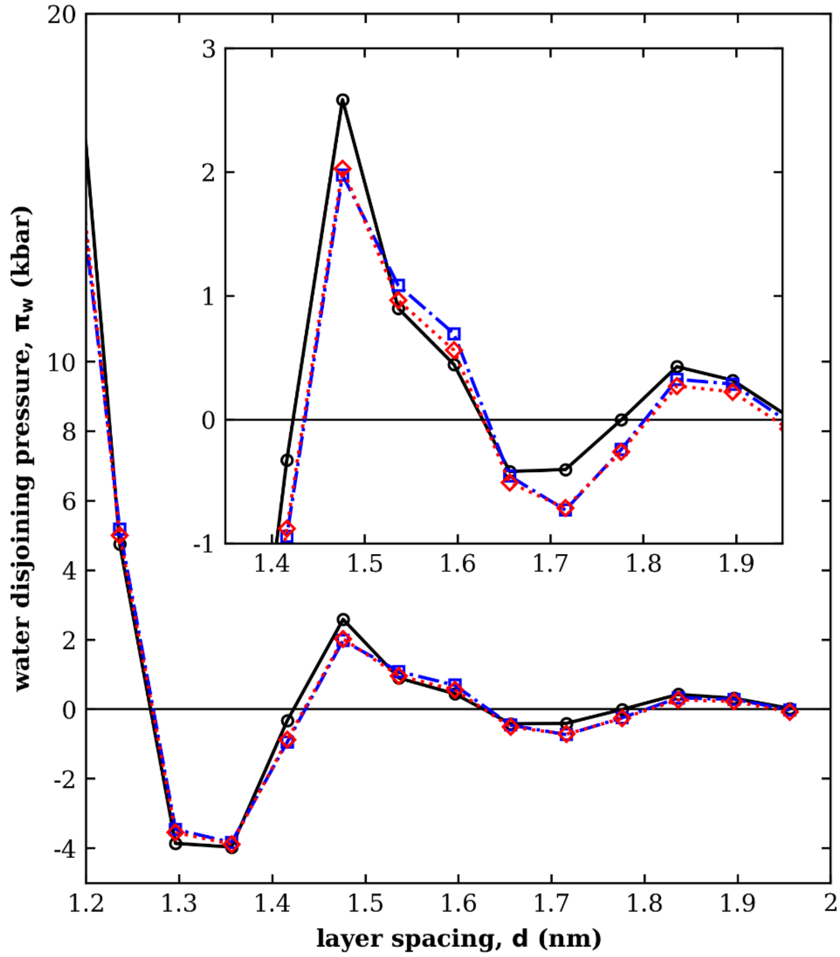

Fig. 1: Disjoining pressure in water confined between smectite platelets computed at 300K with Integral Equation Theory and different approximation closures. (black, full curve, circle) Hyper-Netted Chain approximation. (others) Self-consistent approximation closures.

Acknowledgements

The authors would like to thank SwissNuclear for the financial support.

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