



X-ray studies of interlayer water absorption and mesoporous water transport in a weakly hydrated clay

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The swelling of layered smectite clay particles causes changes in the interlayer repetition distance (d -spacing) as a function of temperature and humidity. For the synthetic clay sodium fluorohectorite, hydrodynamically stable hydration states with zero, one and two intercalated monolayers of water have previously been reported, with discrete jumps in d -spacing at the transitions between the hydration states.

Keeping the temperature fixed and varying the relative humidity surrounding a powder sample, we find small reproducible d -spacing changes also within the hydration states. These changes are monotonous as a function of relative humidity, and one order of magnitude smaller than the shift in d -spacing that is typical of the transition between two hydration states.

We have used this observation to monitor the humidity content of a clay sample *in situ*, in a non-invasive manner. Imposing a humidity gradient between the two ends of a quasi-one-dimensional weakly-hydrated sample, we use space- and time-resolved X-ray diffraction to follow the transport of water by monitoring the swelling of clay particles.

From the series of diffractograms we are able to extract profiles of the relative humidity along the sample length. The time evolution of these profiles describes the transport of water through the mesoporous space inside the clay: An analysis of the measured humidity profiles based on the Boltzmann transform, under certain simplifying assumptions, yields a diffusive behavior that is either normal or possibly weakly anomalous.

The humidity profiles also provide the dependence of the effective diffusion coefficient D of the water vapor in the clay mesoporosity as a function of the local concentration of water molecules. The concentration-dependence of D impacts the transport process significantly. Another effect that potentially alters the process is the removal of water molecules from the mesoporosity by intercalation inside the clay grains. Based on 2D finite element simulations we discuss the mechanisms and consequences of those two effects.