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# Studying metamorphic phase transformations and nano-porosity evolution in situ with Synchrotron X-ray scattering: the example of gypsum dehydration

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We demonstrate how nano-scale metamorphic processes can be unravelled with small- and wide-angle transmission scattering of Synchrotron radiation (SAXS and WAXS, respectively) in concert with a novel loading cell. We studied the dehydration of Volterra alabaster, a polycrystalline gypsum rock, at four temperatures (128, 144, 152, and 173˚C) and two axial strains.

Cylindrical samples with a diameter of $\sim$ 13 mm and a thickness of 1 mm were confined axially by the Beryllium (Be) windows of the loading cell. Prior to heating, the Be windows were screwed into the cell up to hand-tightness (“low strain”) or up to 54 Nm (“high strain”) using a torque wrench, thus keeping the sample discs in place. Radially, the discs remained unconfined with respect to the cell interior. A high-powered electric heater clamped around the cell supplied heat conductively. Numerical models of the thermo-elastic response of the cell and the sample suggest that dehydration onset occurred at temperatures above $\sim$ 105 ˚C. Hence, the transient heating period of the sample was so short compared to the observed dehydration kinetics that the experiments were essentially isothermal. During each experiment, three or five 100x100 micron interrogation points along the disc radius were irradiated with Synchrotron radiation (12 or 16 KeV) for 5 s per scan. These points were distributed equidistantly between disc centre and margin. WAXS and SAXS detectors collected patterns over a scattering vector length range between 1.17 to 4.13 per Å and 0.003 to 0.22 per Å, respectively. The dehydration product was always hemihydrate.

The WAXS data provide the dehydration kinetics. The conversion curves are well described by an Avrami model. However, at the two intermediate temperatures of the low-strain experiments, a clear transient “bump” in the conversion curves suggests a switch in mechanism, possibly from nucleation-and-growth to diffusion control. An Arrhenius fit to low- and high-strain data yields activation energies of 74 and 50 kJ/mol, respectively, lower than those for powders at T $\leq$ 120˚C ($\geq$ 90kJ/mol). However, a single low-strain experiment at 120˚C converted ten times more slowly than extrapolated from the aforementioned Arrhenius fit and powders dehydrated at the same temperature. This observation probably indicates another change in dehydration mechanism at lower temperatures.

The SAXS data yield novel insight into the evolution of pores in the size range 1 to 80 nm. Due to instrument geometry, the Guinier region could not be observed. Thus, absolute porosity could not be measured. Nevertheless, the SAXS signal experienced its most significant modification well after the phase conversion was completed. Therefore, pore-space geometry and porosity on the nano-scale kept on evolving well past reaction completion, indicating two different characteristic timescales for both processes. This observation contradicts the general assumption that phase conversion and porosity evolution proceed hand-in-hand during gypsum dehydration.

Our experiments highlight the tremendous impact of microstructure (powder versus polycrystal) and mechanical loading on dehydration. These insights are only possible because the experiments allowed the coeval monitoring of phase changes and nano-porosity evolution under geological loading conditions.