



Gypsum dehydration as a pore pressure diffusion process - insights from time-series Synchrotron X-ray microtomography, thermodynamic analysis and finite element modelling

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The dehydration reaction of gypsum, an analogue for prograde devolatilisation in Earth sciences, is an anomalously slow process. We conducted a 4-dimensional microtomographic experiment at the Advanced Photons Source to investigate the slow dehydration of gypsum to hemihydrate and anhydrite in situ. The experiment reveals that the sample dehydrates across a narrow dehydration front which propagates to the centre of the sample cylinder, covering 1.3 mm in more than four hours. No reaction was observed ahead of the front. The front separates a permeable outer shell from an undrained central core.

We combined infrared and Raman spectra to calculate the heat capacity and entropy of gypsum, hemihydrate and anhydrite. These data were combined with information on the thermodynamic behavior of water to calculate phase diagrams of the gypsum dehydration reaction for the undrained and permeable parts of the specimen. These phase diagrams show that, at 388 K, Gypsum is stable at pressures higher than 26 MPa, a condition that we consider satisfied ahead of the reaction front.

We analyzed the stress state in the sample and argue that thermal expansion-, pore fluid-, sintering- and chemical pressures suppress the pressure-dependent reaction ahead of the dehydration front. The reaction progresses where water can be drained and the stresses relaxed, giving rise to pressure gradients across the dehydration front. We demonstrate that the propagation of the dehydration front can be described by the diffusion of a pore fluid pressure perturbation in a poroelastic medium, similar to a Biot-Frenkel wave. The analysis of the tomographic data yields a mean linear diffusion coefficient of $5 \times 10^{-11} \text{ m}^2 \text{ s}^{-1}$.

We tested the effects of temperature and pressure on the progress of the dehydration reaction under unconfined conditions with a coupled thermo-hydro-mechanical finite element code. The numerical results agree excellently with the physical observations. We therefore conclude that thermodynamic coupling governs the dehydration process. This introduces temperature- and pressure-sensitive source terms to the classical pressure-diffusion equation. The magnitude of these source terms, the intrinsic transport characteristics of gypsum, and the phase diagram explain the characteristic dehydration time scale observed in the physical experiment.