

The role of temperature on the pressure-mediated adsorption in natural zeolites: the case of leonhardite

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Abstract text

While the high-pressure and high-temperature behavior of natural zeolites has been intensively studied in the last decades, to the best of our knowledge, no *in-situ* X-ray diffraction studies have been performed combining the effects of both. Experiments at these conditions could have crucial geological implications and potential applications at the industrial level (e.g., high-*P/T* adsorption of alcohols compounds in zeolites to promote methanol to olefins reaction). In this study, we present the results from the first pilot experiments, obtained with an easy and reproduceable experimental set-up, performed with one of the most common natural zeolite, *i.e.*, laumontite ($[(Ca_{4-x}Na_x)K_x][Al_8Si_{16}O_{48}]\cdot(H_2O)_n$, with $n \leq 16$). This zeolite occurs in a wide range of natural environments, including sedimentary deposits or volcanoclastic sequences interested by burial diagenesis/metamorphism, as well as in hydrothermal vugs of volcanic rocks. Partially hydrated laumontite (*i.e.*, with 15 H₂O molecules per unit cell) is often referred to as “leonhardite”. The behavior and adsorption mechanisms of these minerals have been (already) well characterized at high-pressure by several authors (Gatta et al. 2018; Comboni et al. 2018), leaving unexplored the effect induced by temperature increase. *In-situ* high-pressure+high-temperature single-crystal synchrotron X-ray diffraction experiments were performed at the ID15-b beamline, at the ESRF, Grenoble (France). Saltwater (0.35 wt% NaCl) was used as hydrostatic pressure-transmitting fluid. The DAC was placed in a resistive heater, which allowed to increase the *T* up to 100(2)°C. Temperature was defined using a thermocouple placed very close to the *P*-chamber; *T* value was consistent with what obtained by the analysis of the Au-powder pattern. In leonhardite, the temperature seems to enhance the H₂O adsorption, giving rise to a volume expansion at *P* < 5 kbar. Above this pressure, the compressibility becomes similar to that of fully hydrated laumontite [2]. Previous experimental findings proved that leonhardite experiences a full hydration, at ambient *PT* conditions, only after about 24 hours, whereas each data-point at high-*PT* required not more than 20 minutes: this further highlights the role played by temperature on the kinetics of the *P*-mediated adsorption process (*i.e.*, speeding up the adsorption process).

References

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