In operando inelastic neutron scattering and pair distribution function analysis of water and carbon dioxide adsorption by calcium-based layered double hydroxide

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Layered double hydroxides (LDHs) have gained significant attention across various industries due to their enhanced specific surface area and catalytic activity. Among this family of materials, calcium-based LDH has emerged as a promising candidate for carbon capture applications due to a high density of basic adsorption sites. It is known that H₂O has a significant influence on the CO₂ capture capacity of the material, however, the fundamental mechanisms controlling this behaviour remain elusive. Here, in operando inelastic neutron scattering, in operando X-ray and neutron pair distribution function analysis, and molecular modelling are used to investigate (i) the H₂O adsorption-desorption behaviour of calcium-based LDH and (ii) the impact of adsorbed H₂O on the CO₂ capture mechanism, and (iii) thermal regeneration of the capture material (Fig. 1). Our results reveal that the exposure of calcium-LDH to H₂O and CO₂ can lead to changes in LDH interlayer spacing, coordination environment, and crystallinity. Moreover, by combining density functional theory normal mode analysis with inelastic neutron scattering data we uncover the mechanism by which adsorbed H₂O molecules affect adsorption of CO₂ molecules, and their subsequent molecular interactions with the Ca-based LDH.

Figure 1. Synchrotron-based X-ray total scattering data of calcium-based layered double hydroxide exposed to elevated temperature (ambient to 700 °C).