Clay minerals under the microscope

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Clay minerals such as montmorillonite (MMT) play an integral role in myriad natural processes involving H_2O, CO_2, nutrient, and contaminant transport and storage in soils, sediments, and clay-rich suspensions. Here, we show that MMT particles in mixed-cation systems dynamically equilibrate between phase-separated states that are near the thermal energy, exchanging both ions and MMT layers between particles. Interlayer forces and thermodynamic relationships between hydrated particle swelling states that control MMT structures were quantified using cryo-transmission electron microscopy (cryo-HRTEM). Direct evidence of cooperative chemical-mechanical interactions between layers during Na^+/K^+ ion exchange that promote rotational ordering lead to a new model of MMT swelling/collapse that accurately predicts selectivity coefficients and swelling states. The three-dimensional nature of the interactions between layers and particles were elucidated by cryo-electron tomography and found to be controlled by layer size polydispersity, leading to a panoply of topological and kinetic defects that are expected to have a dramatic impact on transport in clay-rich media. This new perspective will help reduce the complexity of clay mineral systems and improve predictions of ion exchange and fluid transport in many natural systems.