Electronic Supplementary Material (ESI) for Nanoscale.

Supplementary Information

Atomic structure of surface defects at mineral-water interfaces probed by in situ Atomic Force Microscopy

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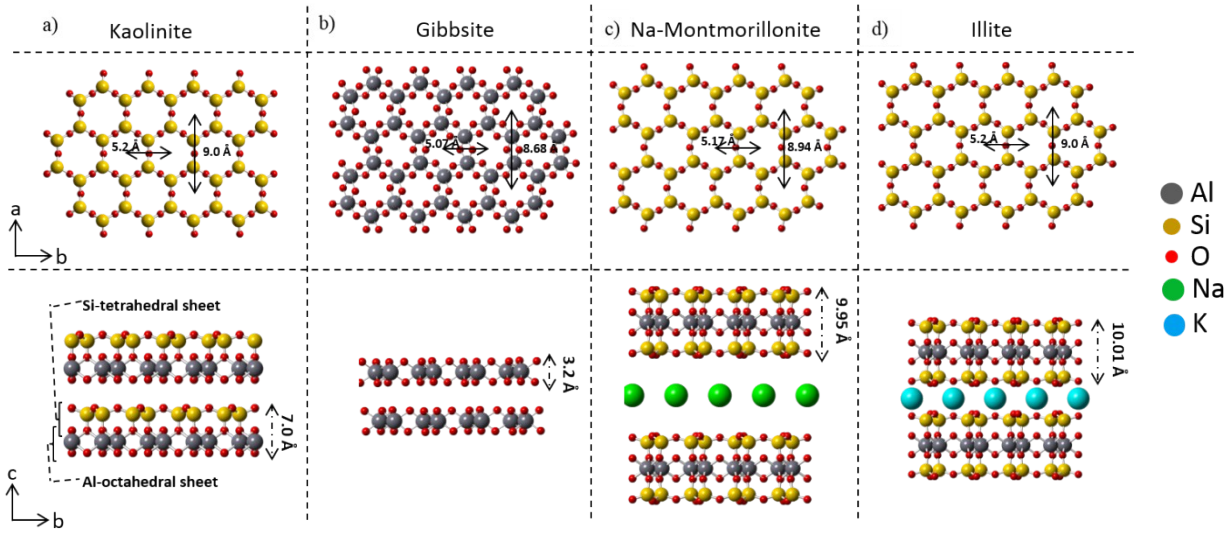
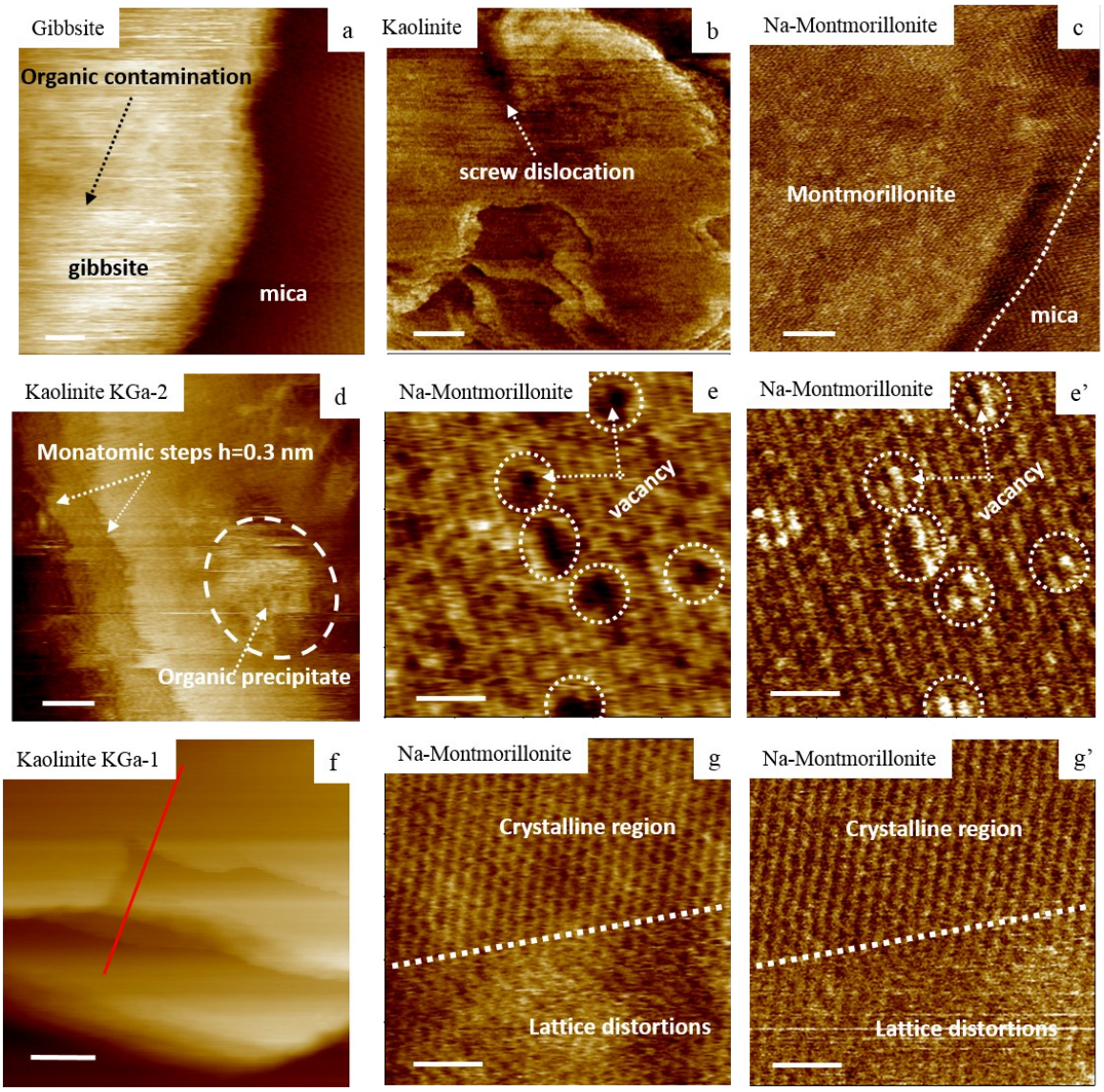


Supplementary Figure 1. (a) The kaolinite, (b) gibbsite, (c) Na-montmorillonite and (d) illite crystal structures. Top row-view of a single layer of the clays structure looking down upon the basal plane. Lower row-side view of the clays structure orthogonal to the basal plane.



Supplementary Figure 2. High resolution AFM images of defects of various clay particles. (a) topography image of gibbsite particle (left side, brighter) completely coated with organic adsorbate on mica substrate (right side, darker) displaying clean crystalline structure. Scale bar, 2 nm. (b) kaolinite particle with many steps, defects and screw dislocation defects. Scale bar, 15 nm. (c) atomic resolution phase image of Na-montmorillonite particle close to edge that display the defect free crystalline structure. Scale bar, 5 nm. (d) topography images of kaolinite (Kg-2) particle close to edge that display atomic steps, defects and adsorbents. Scale

bar, 15 nm. (e,e') Na-montmorillonite atomic resolution topography and phase picture showing the hexagonal periodic structure with a number of vacancy defects. Scale bars, 2 nm. (f) high resolution topography picture of kaolinite particle close to the edge that display monatomic and lattice steps. Scale bars, 15 nm. (g,g') atomic resolution topography and phase images of basal plane of Na-montmorillonite displaying a region with well-ordered hexagonal structure *and a region with disordered non-periodic structure.* Scale bars, 2 nm.