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From ions to islands: an atomic scale study of gibbsite nucleation on muscovite.

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Classical nucleation theory predicts precursors to nucleation are small molecular clusters that form spontaneously *via* statistical-mechanical fluctuations. However, recent studies suggest pathways to mineral nucleation are more complex, involving aggregation of clusters or the formation of liquid-like precursors. Numerous efforts have been mounted to directly detect the precursors to nucleation, because such an achievement would provide great insight into nucleation pathways. Unfortunately, the species present prior to nucleation are either too small or too scarce to be unambiguously detected using techniques such as *in situ* electron microscopy or X-ray scattering. However, recent advances with *in situ* AFM now allow for solution imaging of surfaces with atomic resolution. In this study we utilize *in situ* AFM to investigate epitaxial nucleation and growth of gibbsite (aluminum hydroxide) on muscovite mica. The results show that, at low pH (2.5), 30°C, and moderate $AlCl_3$ concentration (1 mM), the mica surface supports a dynamic population of atomic species, which appear to reside at the cation sites. As pH is increased to 4.0, a dynamic population of clusters about 1 nm in diameter develops, which is approximately monodisperse. As temperature is increased to 46°C, the surface becomes covered with irregular-shaped clusters ranging from 1 to 20 nm in diameter, as well as highly-transient species of unknown dimension. At 60°C the clusters evolve into crystalline islands comprised of 4 to 5 Å thick monolayers of gibbsite, which coalesce and coarsen with time. The dependence of cluster number density on concentration indicates the initial species are $Al(OH)_2^{1-}$. These observations point to a reaction pathway that differs from traditional assumptions of nucleation *via* rare, dispersed clusters built from monomeric species, but instead suggests the involvement of ion coordination complexes, the existence of special cluster sizes, and crystallization occurring by condensation of a relatively dense adsorbate layer.