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Mesocrystal growth through oriented sliding and attachment of nanoplates

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Oriented attachment (OA) is a unique crystal growth pathway through which nanocrystals self-assemble into larger structures via crystallographically controlled particle coalescence. While previous studies emphasized the intragap forces driving crystallographic alignment, the mechanisms enabling uniform stacking and superlattice formation remain poorly understood. Here, combining advanced experimental characterization with atomistic simulation, we demonstrate that macroscopic gibbsite [$Al(OH)_3$] mesocrystals form through directional sliding of hexagonal nanoplates into staggered positions.

Synchrotron wide- and small-angle X-ray scattering (WAXS/SAXS) of single mesocrystals reveals a monoclinic superlattice structure with nanoplates stacked at a uniform ~50° stagger along the gibbsite [010] direction. Selected area electron diffraction (SAED) confirms three-dimensional orientational order extending over hundreds of micrometers. Critically, in situ liquid-cell transmission electron microscopy (LCTEM) captures two-stage OA dynamics in real-time: (i) rotation and jump-to-contact achieving partial overlap within ~1 second, and (ii) preferential sliding along [010] that progressively decelerates with increasing overlap—kinetic behavior inaccessible without direct dynamic imaging.

To elucidate atomic-scale origins of this directional behavior, we performed molecular dynamics (MD) simulations using umbrella sampling to calculate the potential of mean force (PMF) for nanoplate sliding. These computationally intensive calculations reveal that the staggered arrangement corresponds to a global free-energy minimum rather than complete alignment. The simulations confirm that sliding along [010] is energetically favored over [100], with lower energy barriers per overlapping unit (0.17 vs. 0.41 kcal mol^{-1} $hexagon^{-1}$) and smaller initial alignment barriers (0.89 vs. 2.49 kcal mol^{-1}). By systematically varying particle separation and edge charge, simulations demonstrate that staggering arises from balancing electrostatic edge-edge repulsion, van der Waals attraction, hydration forces, and entropic contributions—quantitative insights unattainable through experiment alone. The predicted offset (~0.7 nm for model particles, scaling to 15.5 nm for experimental-sized platelets) matches the measured distribution (11.5 ± 6 nm).

This integration of synchrotron characterization, real-time liquid-cell imaging, and atomistic simulation provides unprecedented mechanistic insight into OA, linking macroscopic mesocrystal structure to the atomic-scale energy landscape of interparticle interactions. The findings have implications for rational design of hierarchical materials and interpreting natural mineral structures formed via OA in geochemical and biomineralization contexts.