

# Directional energy-structure relationship during oriented attachment of gibbsite nanoparticles

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Oriented attachment (OA) is a nonclassical crystal growth pathway, in which lattice-matched particles assemble into a larger crystal structure. The process can be divided into two steps: in the first step two particles separated by a thin layer of solvent rotate and translate to align with one another. In the second step the intervening solvent layer is removed, and particles jump to contact. Unraveling the directional energy-structure relationship during rotating, translating, and approaching of particles is key to understand and control the OA process. Here we demonstrate such relationship for gibbsite nanoparticles system. We first illustrate experimentally the oriented attachment of gibbsite plates. The suspension of uniform hexagonal gibbsite plates of 100 nm in diameter was evaporated at room temperature for several months. Scanning electron microscope image suggests that the plates stack on basal (0 0 1) surfaces along the (0 1 0) directions. We then use molecular dynamics simulations to calculate the potential of mean force for gibbsite particles relative motions. Multiple translational directions (e.g., along *a*- and *b*-axis), crystal facets (basal and edge), and intervening water layers (monolayer and bilayers) are considered in our work. Our results reveal that (1) basal-basal gibbsite surface attachment is more favorable than basal-edge, and edge-edge attachments, (2) translation toward alignment along the *b*-axis [or (0 1 0) direction] can be more energetic favorable than translation along the *a*-axis [or (1 0 0) direction], (3) particle-particle interactions and hydrogen bond network strongly relate to the features (e.g., energy minima, maxima, and barriers) observed on the potential of mean force profiles. This work provides the energy–structure relationships during particles relative motions to understand the crystal face selectivity and alignment pathway selectivity of OA.

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