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Effect of Particle Misalignment on Energetic Pathways towards Oriented Attachment of Gibbsite Nanoparticles

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The aggregation of (nano)particles in solutions is fundamentally important to colloidal science and its applications including nuclear waste management of Hanford and Savannah River tanks. Oriented attachment is a special case of particle aggregation in which crystalline particles assemble into a larger particle by attaching on specific crystal faces that are lattice-matched. If the two particles or crystals are not fully aligned, then the atom-by-atom mismatch between the two crystals creates forces that drive the particles towards oriented attachment. However, these forces that drive the particles to be perfectly aligned are not well understood. Here we use classical molecular simulations to calculate the potential of mean force (PMF) for three different particle-particle interactions: (1) the neighboring basal surfaces of two gibbsite particles slide over each other (sliding motion), (2) the two particles rotate with respect to each other on their basal surfaces (rotating motion), and (3) the two particles approach each other from a distance with basal surfaces properly aligned (approaching motion). For the sliding motion, as the particles overlap more, the system energy oscillates between minima and maxima with the minima becomes deeper and maxima become higher. The features in the PMF profile are determined by the misalignment or alignment of the two particles and the structure of water between the particles. Consequently, the two particles can slide into either completely overlapped/coaligned or partially overlapped/coaligned, or partially overlapped/misaligned configurations. Upon rotation, misaligned basal surfaces can easily become aligned without encountering energy barriers. During rotation, the particles reach a minimum energy every 60^o due to the hexagonal crystal structure of gibbsite which is also reflected in gibbsite particle morphology. The highest free energy barrier encountered for oriented attachment is for the removal of the last water layer between the particles. This research provides a molecular picture of the direction-specific energy-structure relationships during oriented attachment of gibbsite nanoparticles.

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