Voronoi Tessellation Analysis of Clathrate Hydrate Nucleation and Growth

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We have obtained a large number of molecular dynamics simulation trajectories that capture the spontaneous nucleation of methane clathrate hydrates. One of the challenges in analyzing these trajectories is to quantify the molecular events before, during and after hydrate nucleation. Since hydrate structures are defined from the formation of water cages enclosing a guest molecule, a geometric order parameter for identifying the cages formed could serve as a suitable descriptor for the nucleation and growth of hydrates. We will present analysis of the hydrate nucleation and growth trajectories using Voronoi tessellation. We will show that for perfectly crystalline sI and sII hydrates, tessellation of the volume occupied by methane molecules generates vertices that closely overlap with the oxygen atoms positions of the actual water molecules forming the cages. In applying the tessellation to the nucleating trajectories, we observe that several of the Voronoi properties (polyhedra volume, total surface area, asphericity parameter) show signatures of change when hydrates start forming. The results suggest that the Voronoi tessellation analysis is a powerful and efficient tool that can be applied to obtain further insight into the mechanisms associated with the ordering of methane and water molecules in the nucleation and growth processes of hydrates.