Novel States of Gas in Water

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Abstract

Dissolution of gases in liquid water is a general and fundamental phenomenon across living and nonliving things. Conventionally it has been assumed that dissolved gas molecules are well dispersed as monomers. However, many mysteries about gas dissolved in water, such as the nucleation mechanism of gas bubbles in water, whether nanobubbles exist in water, and the abnormal thermodynamic properties of gas-saturated water, persist up to now. We encapsulated water between two laminated graphene layers and used transmission electron microscopy (TEM) to examine whether dissolved gas forms any microstructures in water. Degassed water, deionized water, and water supersaturated with pure gas (N_2, O_2, Ar, Xe, CO_2, and SF_6) were investigated at room temperature. While neither degassed water nor deionized water yielded specific features, two major microscopic structures were evident in gas-supersaturated water: (1) individual polycrystalline nanoparticles in liquid water, and (2) novel mesoscopic clathrate structures. In the clathrate structures, water molecules form a crystalline matrix hosting a high density of gas-containing cells (cell separation of 4-8 nm). The observation of the novel clathrate state may help resolve several long-standing puzzles. Almost identical clathrate structures were also observed in ethanol-water mixtures. As many research groups reported the presence of bulk nanobubbles in the mixtures, raising the possibility that bulk nanobubbes might be the clathrate structures. Taken together, our studies suggest that the mesoscopic clathrate structures may play a wide role in aqueous solutions.