Discovering Hidden Structures of Clathrate Hydrates

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Clathrate hydrate has diverse crystal structures, among them, the three most prevalent ones covered nearly almost all hydrates, other structures are taken into account only when specific guest molecules are present. Here we report the observation of a hidden clathrate structure - the tetragonal structure (TS-I) in commonly formed gas hydrates, as evidenced from molecular dynamics simulations. We show that in a system supporting the formation of sI crystals, when two (or more) sI crystal grains with different growth directions come into contact or when the growth of sI crystal encounters geometrical frustration, the TS-I results as a co-crystal. And we give evidence that the TS-I may also play an important role in the combination and/or transition between sI and sII. These results hint that this previously neglected structure may be commonly present whenever sI and/or sII are formed. In a broader context, we suggest that confining the possible structures for common gas hydrates within three dominant structures may hinder an in-depth understanding of clathrate hydrates.