Cage Occupancy of Hydrogen Molecules in the Structure I, II, H, and VI Hydrates

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Hydrogen (H2) is an interesting target as a guest molecule of clathrate hydrates, because H2 is enclathrated in the hydrate cages and simultaneously migrates relatively-freely through hydrate cages. In the structure II mixed hydrates with thermodynamic promoters, large-cage occupancy of H2 (so-called “tuning effect”) is possible by using a specific preparation method. For the tuning effect occurring, the competitive condition of H2 and promoter molecules toward the large-cage occupancy is required. There is no report on the tuning effect in the structures H and VI. In the structure I mixed hydrates with methane (CH4) or carbon dioxide (CO2) occupying both the small and large cages, Grim et al reported the competitive cage occupancy of H2 and CH4 (or CO2) in the small and large cages dependent on the compositions. In the case of a large structure I former not occupying the small cages, that is, under less competitive conditions, how is the large-cage occupancy of H2 in the structure I mixed hydrate? Is a specific method necessary? We will report the experimental results on the tuning effect in the structure I mixed hydrates. In addition, the cage occupancy of hydrogen molecules in the structure I, II, H, and VI hydrates will be summarized.