Molecular Simulation of Natural Mixed Clathrate Hydrates

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Recently, natural clathrate hydrates are at the heart of important environmental problems and are also a subject of study for astrophysicists. The geological and astrophysical, *in situ* conditions, generally imply the presence of gas mixtures in the hydrate forming system. The question of the competition between the different molecular species in the process of clathration is then raised. Namely, the selectivity is of interest to determine the most stable hydrates formed for a given composition of gas mixtures. Grand Canonical Monte Carlo (GCMC) simulations constitute a useful tool to access this information. In continuity with the adsorption analogy used to model the equilibria of clathrate hydrate in the van der Waals – Platteeuw theory, GCMC simulations can give the quantity of trapped, or adsorbed-like, molecules in a clathrate hydrate. In the case of a gas mixture. Then, the structural and thermodynamic properties of the formed clathrate hydrate phase can be derived. Furthermore, natural clathrate hydrates are often in contact with mineral surfaces. Thus, the impact of different types of surfaces on gas trapping and the associated selectivity within clathrate hydrates are investigated.