Computer simulation of the carbon dioxide hydrate – water interfacial free energy

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Carbon dioxide hydrates are ice-like nonstoichiometric inclusion solid compounds with importance to global climate change, and gas transportation and storage [1]. The thermodynamic and kinetic mechanisms that control carbon dioxide nucleation critically depend on hydrate-water interfacial free energy. Only two independent indirect experiments are available in the literature [2,3]. Interfacial energies show large uncertainties and overestimated values due to the conditions at which experiments are performed. Under these circumstances, computer simulation tools over a way to estimate interfacial energies at coexistence conditions from a molecular perspective. We propose a novel implementation of a computational methodology based on the definition of interfacial energy [4] and the use of realistic and reliable models of water (TIP4P/Ice) [5] and carbon dioxide (TraPPE) [6] that have proven to predict accurately the ice-water interfacial energy and the dissociation line of carbon dioxide hydrates [7]. We found that simulations provide an interfacial energy value, at coexistence conditions, consistent with the experiments [2,3] from its thermodynamic definition [8]. This is the first calculation of the CO_2 hydrate interfacial energy using molecular dynamics simulations at coexistence conditions from fundamental principles, including the definition of interfacial free energy, Thermodynamics, and Statistical Mechanics. This pioneering work opens a door to estimate accurate interfacial energies of hydrates from a molecular perspective [8].

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