Molecular basis of tetrahydrofuran-induced enclathration

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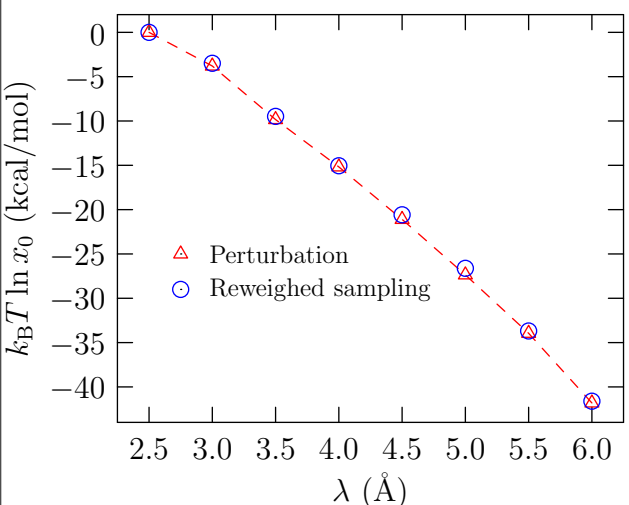
Clathrate hydrates are an important potential source of energy. They are also promising materials to sequester CO_2 or store H_2. In nature, hydrates are typically formed under high pressure and/or very low temperatures. We are investigating the molecular mechanism of how tetrahydrofuran promotes the enclathration of methane even at modest pressures and temperatures. Towards this goal, we are developing theory and simulation tools to understand the role of local chemical, packing, and long-range interactions in hydration thermodynamics. Two key developments are noted below.



We have developed a reweighed sampling approach to study the formation of large cavities around a target solute. The figure shows results for cavities around a water molecule itself. The results are in excellent agreement with the more computationally expensive perturbation approach.

We implemented an *ab initio* hybrid Monte Carlo method, and using this technique, we investigated several electron density functionals to see which functional/temperature combination best describes water. The figure to the right shows the balance of chemistry and packing for various coordination volumes.