Master Physics & Computational Physics
1st year project (2020-2021)

Title: Diffusion of diatomic molecules in gas hydrate

Key words: clathrate hydrates, DFT/AIMD calculations, diffusion coefficients

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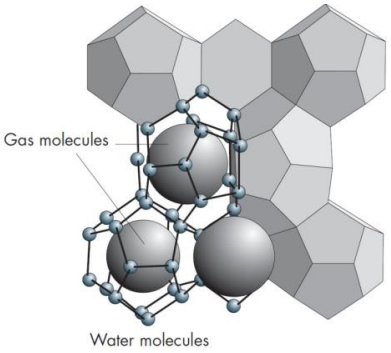
Subject :

Under specific conditions of pressure and temperature, the mixing of water and gas molecules can lead to the formation of nanoporous crystalline solids called clathrate hydrates (also called gas hydrates). These compounds are made by a tetrahedral network of water molecules (“host”) forming cages inside of which gas molecules (“guest”) are encapsulated (see figure).



Naturally abundant on Earth, gas hydrates can be observed on permafrost regions (Alaska, Siberia), on ocean floors and are generally expected to exist on icy celestial bodies (comets and planets of the solar system). This natural occurrence makes them relevant for many geophysical and astrophysical applications. Therefore, research studies on gas hydrates have steadily expanded nowadays toward a broad area ranging from molecular sciences to geosciences.

After a bibliographic research in the gas hydrates field, the student will study the **diffusion of diatomic molecules in gas hydrates**, specifically CO and N_2 molecules for which clathrate hydrate properties are quite similar. Indeed, experimental results show that both CO and N_2 gas hydrates are initially formed in the cubic sI structure and after some time (days for N_2 and weeks for CO) both crystals transform into the cubic sII structure. The sI structure would be kinetically promoted at the early synthesis stage whereas the sII structure would be thermodynamically favored. Previous studies showing the structural stability of sII hydrate were already performed. Nevertheless, the kinetic part still remain to be studied. One of the key aspects for the kinetic interpretation could be explained by the diffusion of gas molecules through the gas hydrate cages. Calculations will be performed on the CO/N_2 clathrate hydrate using nudge elastic band (NEB) method within a density functional theory (DFT) code. Potential energy barriers of gas molecules diffusing through the various cages will be extracted for both sI and sII structures. *Ab-initio* molecular dynamics calculations (AIMD) will then be done in order to complete static DFT calculations allowing to extract diffusion coefficients for gas molecules.

***Numerical tools*:** VASP software, fortran/python/bash scripts
Equipment: PC, computer cluster (mésocentre UFC)