UNDERSTANDING HYDROGEN STORAGE IN METAL – ORGANIC FRAMEWORKS USING MASSIVELY PARALLEL ELECTRONIC STRUCTURE CALCULATIONS

Research Challenge

Hydrogen, as a fuel, requires efficient storage materials for retaining and releasing hydrogen in large quantities. Metal–organic frameworks (MOFs) are a possible material, and their hydrogen storage potential and microscopic properties need investigation. We performed first-principles quantum-mechanical molecular-dynamics calculations to understand the behavior of hydrogen inside MOFs. The knowledge gained will inform the materials research community on underlying the properties of hydrogen in MOFs, and how we may improve the chemical composition for hydrogen storage.

Methods & Codes

This requires an accurate quantum-mechanical simulation at finite temperature. We employed the Car-Parrinello Molecular Dynamics simulation technique (CPAIMD), which allows the nuclei to move on the Born-Oppenheimer energy surface provided by plane wave-based DFT. By including the electronic degrees of freedom (valence) explicitly, we bypassed force field difficulties.

Results & Impact

The full MOF simulation has a large amount of "vacuum" (voids) in the structure to provide room for hydrogen diffusion, and a large internal surface area for hydrogen adsorption. The smaller size of the mini-MOF allows us to run very long molecular dynamics simulations and benchmark the results carefully. We performed our simulations at 77K and 300K for both systems to understand the effect of temperature on hydrogen diffusion. The simulations show significant diffusion of the H₂ molecules and are statistically meaningful.

Why Blue Waters

Massively parallel electronic structure calculations require tightly coupled computing nodes due to intense communication loads: electron waves are delocalized over the entire system so all parts of the system end up interacting with each other. For the MOF system of interest, the CPAIDM simulations require a massively parallel calculation with many hundreds of nodes.