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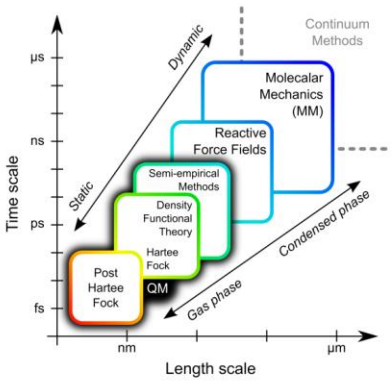
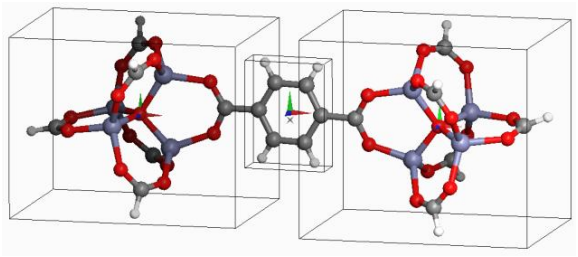
CHARGE EQUILIBRATION MODELS FOR METAL-ORGANIC FRAMEWORKS

Keywords: model development, density functional theory, force fields

Density Functional Theory (DFT) plays a major role in the field of computational chemistry and material science. It is a quantum mechanical model for the electronic many-body system and makes it practically feasible to study chemical reactions, material properties, the structure of liquids, etc. at the atomic scale on high-performance clusters (HPCs). Within DFT, there are several levels of approximation, and next to DFT there are also other theories to study molecular systems. The two main criteria to select one of these models for a given computational problem are (i) accuracy and (ii) computational efficiency. In many cases, DFT offers the best compromise. DFT is also used as a theoretical foundation to derive empirical models for the molecular charge distribution. The derivation of the Electronegativity Equalization Method (EEM) from DFT by Mortier et al. started an entire new field of charge equilibration (QEq) models. Qeq models were later combined with empirical molecular mechanics models and reactive force fields to create computationally much more efficient alternatives for the quantum mechanical approaches, at the expense of some accuracy.

An ongoing collaboration between the CMM and the LASIR group in Lille (France) resulted in a new scheme to calibrate the parameters in Qeq models that are transferable from small molecules (for which high-level QM reference data can be computed) towards periodic systems. So far, this promising scheme is only tested on Zeolite systems. The extension of this work to more challenging systems, e.g. Metal-Organic Frameworks (MOFs) is of great scientific value. MOFs are a recently invented and form a promising class of new microporous materials with diverse potential applications in the chemical industry. As shown in the figure below, they consist of a regular framework of alternating oxide clusters and organic linker molecules.





Goal The student will develop charge equilibration models for Metal-Organic Frameworks that are currently studied at the CMM. The Qeq model will be used as a component in the MOF force fields that are developed at the CMM. A second application is the effect of the substitution of functional groups in the organic linkers on the charge distribution in the metal clusters of the framework.

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