High-throughput screening of MOFs

Computational discovery of metal organic frameworks for energy conversion and storage

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In Short

- Development of metal-organic frameworks for energy storage and conversion.
- High-throughput screening from density functional theory.
- Energetic stability and gas-adsorption properties.

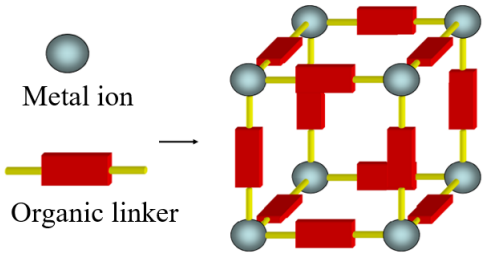

Figure 1: *Schematic MOF structure with its building blocks.*

The increasing demand for sustainable energy sources has become a pressing issue on the agenda of world leaders. Ambitious and yet necessary objectives such as the reduction of fossil fuel emissions, access to renewable sources, and sustainable usage and distribution of the available resources cannot be realized without scientific breakthroughs. The development of new materials that are able to respond to the above-mentioned demands for energy conversion and storage is one of the cornerstones to hit the target. In view of the current rise of geopolitical conflicts and to ensure that these technologies are both sustainable and cost-effective, it is essential to employ non-toxic and earth-abundant materials. Metal-organic frameworks (MOFs) are particularly promising materials in this regard as they can be manufactured from cheap and sustainable sources [1]. Accordingly, research on MOFs has gained a lot of interest in the last few years.

MOFs consist of metals or metal clusters and organic molecules as nodes and linkers, respectively, which build up a crystalline network as shown in Figure 1. Their flexible structure promotes mechanical and chemical tuning as efficient tools to optimize selected material properties for the targeted functionality and their porous nature is ideal to host gaseous molecules. Last but not least, the composition of MOFs can be flexibly modulated to maximize the presence of environmentally friendly elements and reduce or even exclude polluting and toxic building blocks.

In this project, we focus on two applications of MOFs in the field of sustainable energy, namely the storage of hydrogen to be used in combination with fuel cells and the capture and storage of carbon dioxide. The latter process is also known as carbon sequestration and it is considered a promising route to reduce the concentration of CO_2 that is present in the atmosphere. By embedding first-principles simulations in automatized high-throughput workflows we aim to explore a manifold of possible MOF structures composed of sustainable elements to identify the most suitable candidates for the application.

The technical implementation foots on our previous work in which we have integrated in AiiDA [2], one of the most established platforms for this type of calculations, routines to perform high-throughput screening of materials for targeted applications, such as photocathodes [3]. With the underlying quantum mechanical calculations performed within the framework of density functional theory (DFT) we have access to quantitative estimates of the electronic structure and adsorption properties of the guest molecules for hundreds of MOF structures giving us valuable insights on the fundamental mechanisms that govern the material's performance.

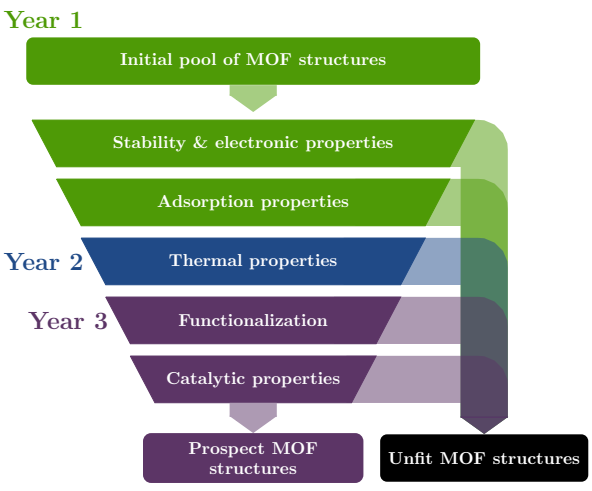

Figure 2: *Computational workflow representing the tasks of the project over the planned 3-year period.*

The high-throughput workflow itself is based on a funnel principle and divided into subsequent stages, as illustrated in Figure 2. Starting from an initial pool of candidates extracted from existing databases such as CoRE-MOF [4] and qMOF [5], the input structures for the actual calculations are selected to fulfill the sustainability criteria defined above. Metal nodes and organic linkers are chosen among abundant and non-toxic elements. For the latter, biological molecules will be preferentially selected, such as lignin, caffeine, etc. At each stage of the workflow, a key property for the MOFs is calculated, starting from the energetic stability and proceeding with the electronic properties, the ability to adsorb/desorb guest molecules, vibrational analysis, etc. Unfit structures are discarded at each step, progressively narrowing down the pool of candidates.

The project is planned to run for a total duration of 3 years. In the first year, we will focus on the high-throughput screening part of the project, aimed at identifying suitable MOF structures and compositions for hydrogen storage and carbon sequestration. In the second year, we plan to focus specifically on the evaluation of the thermal properties, which are crucial given the porosity and hence the flexibility of MOF structures and also the room temperature conditions in which the considered processes occur. In the third year, we will take the most promising candidate structures and explore different functionalizations to enhance the adsorption properties of the material. At that stage, we will also explore the suitability of the MOFs to split the carbon dioxide molecule by reconstructing the reaction pathway theoretically.

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More Information

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DFG Subject Area

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