Automated Workflow and Multiscale Modeling of MOFs

A workflow for creation, optimization and analysis of PCU-MOFs

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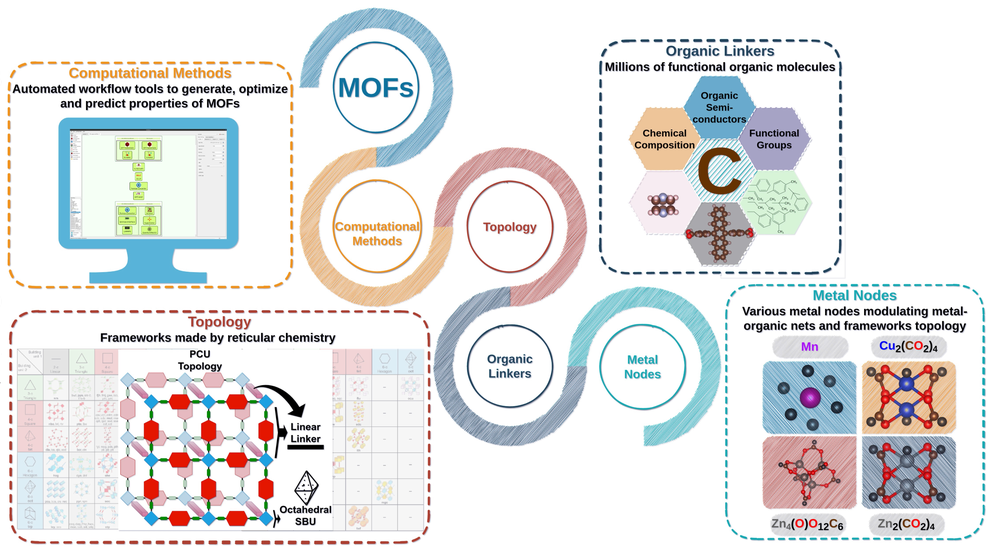
Introduction

- Metal-Organic Frameworks(MOFs) consist of organic linkers and inorganic clusters (SBUs) and are vital in material science for applications like sensors and gas storage.
- Lab-based MOF synthesis is expensive and time-consuming, relying on trial and error.
- Electronic structure modeling of MOFs offers a cost-effective approach to accelerate material development.
- Employed the SimStack workflow environment (`https://www.simstack.de/`) to develop and utilize modular components (WANO) for the creation, optimization, and analysis of Metal-Organic Frameworks (MOFs) following a primitive cubic topology (PCU).
- These WANO modules are adaptable for integration into diverse workflows



Materials & Methods

- MOFs enable semiconductor fabrication for regulating organic electronic properties through networked ordering.
- Pentacene (Pn), a highly mobile organic molecule, serves as a promising linker, forming ordered π-stacks.

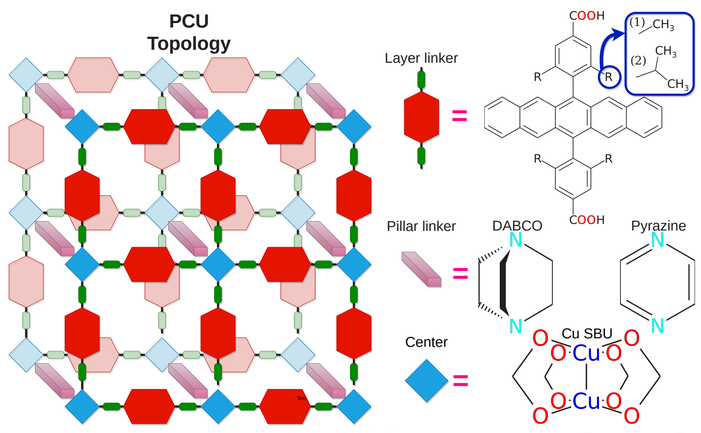


Fig. 1: Pentacene and modifications in PCU-MOF creation

- Zn-Pn SURMOF shows hopping-like transport with an activation energy of 64 meV.
- In silico crystal engineering reduces Pn fluctuations within MOFs, substantially enhancing charge carrier mobilities.
- These effects are explored in Cu-Pn MOFs and various pentacene modifications.[1]

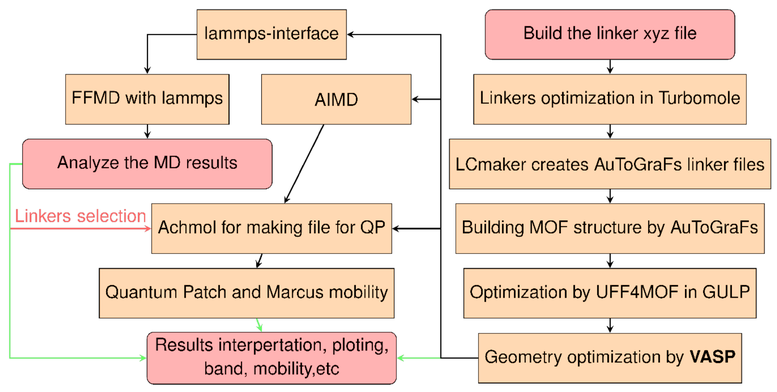


Fig. 2: Conceptual diagram for possible MOF workflows using our WANOs

- The workflow employs DABCO and pyrazine as pillar linkers and pentacene as the layer linker.
- The PCU topology workflow comprises three parts, each with specific WANOs functioning as GUIs for backend codes.

Fig. 3: An example of the Simstack workflow for building MOFs in PCU topology

Results

- Our result confirms the previous electronic coupling and shows a stable structure for DABCO and Pyrazine as pillar linker. [2, 3]

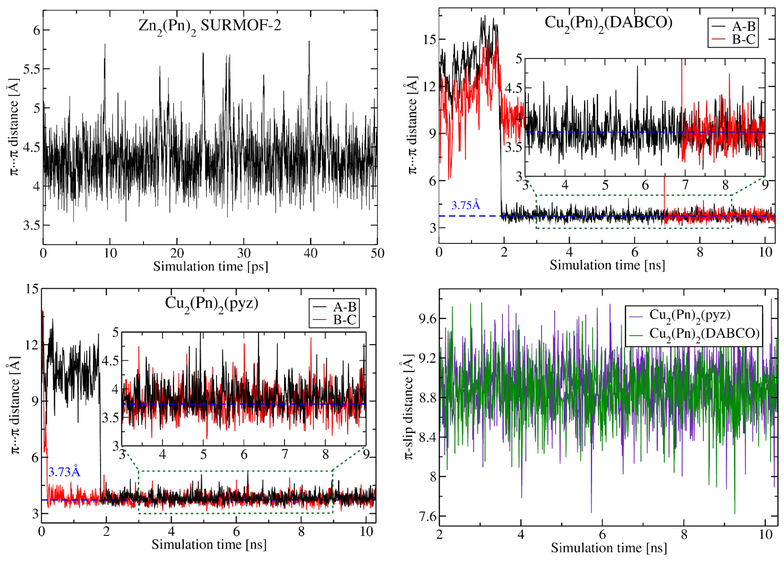


Fig. 4: Dynamical change of π-π interaction and π-slip distances between pentacene cores and center of mass distance obtained from MD simulations at room temperature showed the coupling.

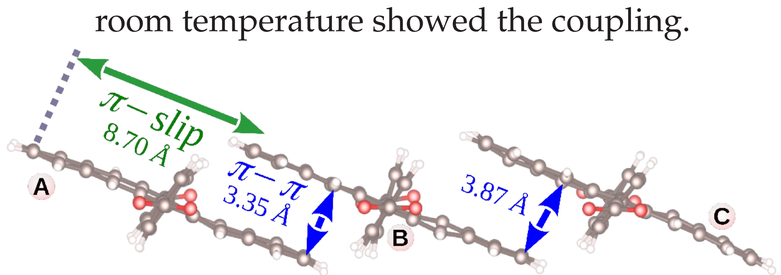


Fig. 5: $\pi - \pi$ stacking between the Pn linkers

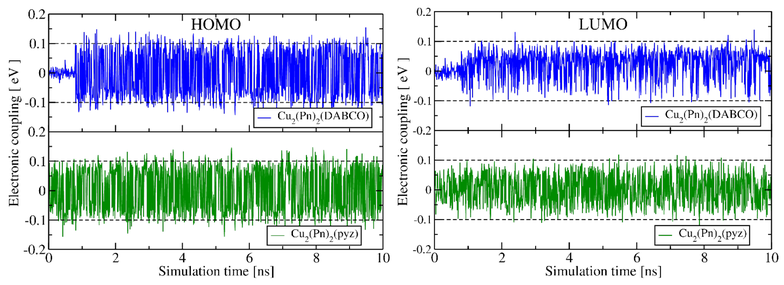


Fig. 6: Electronic coupling is calculated by our workflow for Pn-DABCO with Pyrazine as pillar linkers

Table 1: Coupling from Quantum Patch (B3LYP/def2-SVP) in meV

	DABCO (<9.224 Å>)	Pyrazine (<9.228 Å>)
<JHOMO>	75.7 (-73.2)	77.3 (-77.3)
<JLUMO>	48.2 (-42.1)	47.1 (-47.4)

- Changing of Pn with modified Pn as linker destabilizes stacking.

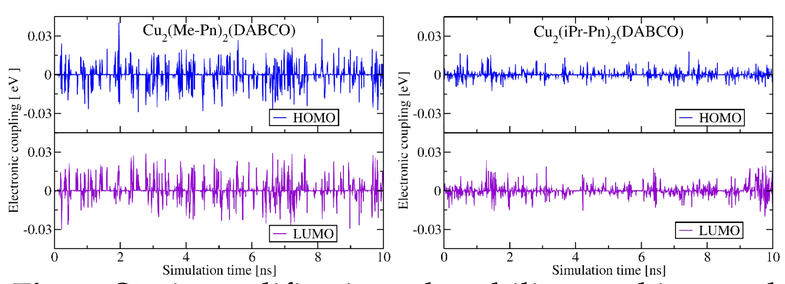


Fig. 7: Steric modifications destabilize stacking, and the coupling is significantly lower

Conclusion

- Our workflow accelerates MOF synthesis, optimization, and analysis in PCU topology.
- Improved Pn-DABCO stacking enhances electronic coupling.
- Electronic coupling in $Cu_2(Pn)_2$(DABCO) and $Cu_2(Pn)_2$(pyz) OSCs averages 78-82 meV and 46-52 meV for HOMO and LUMO orbitals, respectively.
- Steric modifications disrupt stacking and reduce coupling[4].

References

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