Data-Driven Exploration of Thermal and Elastic Properties in Covalent Organic Frameworks

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Covalent Organic Frameworks (COFs) are porous, highly tunable materials composed of organic cores and linkers that assemble into regular two- or three-dimensional networks, offering high surface area, adjustable chemistry, and structural versatility suited for applications such as gas storage, flexible electronics, membranes, and catalysis. [1] Experimentally synthesized COFs often exhibit low crystallinity, including features such as missing cores or linkers, point defects, grain boundaries, and stacking irregularities [2,3] that affect their thermal [4] and elastic [5-7] properties. Therefore, accurately predicting their properties requires computational methods capable of simulating systems containing tens of thousands of atoms. Traditional approaches such as density functional theory offer high accuracy but scale poorly, limiting their applicability to small unit cells, while classical force fields scale well but lack the flexibility to capture complex interatomic interactions. Machine Learning Interatomic Potentials (MLIPs) provide a promising alternative by interpolating high-dimensional potential energy surfaces learned from *ab initio* data, achieving near-DFT accuracy at a fraction of the computational cost. Recent advancements in equivariant graph neural network architectures, particularly the MACE [8] family of models, offer improved transferability and scalability for chemically diverse systems. In this work, we develop an MLIP based on the MACE architecture to study monolayer 2D COFs with a focus on their mechanical and thermal behavior in the presence of defects. Our model is trained on a diverse set of *ab initio* data [9], which includes highly strained conformations, and evaluated for its ability to generalize to COFs containing a variety of structural imperfections. Using this MLIP, we analyze the mechanical response of defective COF monolayers under tensile strain and show that the Young's modulus remains practically unaffected by the presence of defects. Next, we compare the effect of defects on the thermal conductivity of two 2D COFs of different stiffness. Overall, this study demonstrates the effectiveness of MACE-based MLIPs for probing large defective COF systems and provides new insights into how structural imperfections modulate their mechanical and thermal properties, offering a path toward more predictive design of COF materials for real-world applications.

References

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Figures

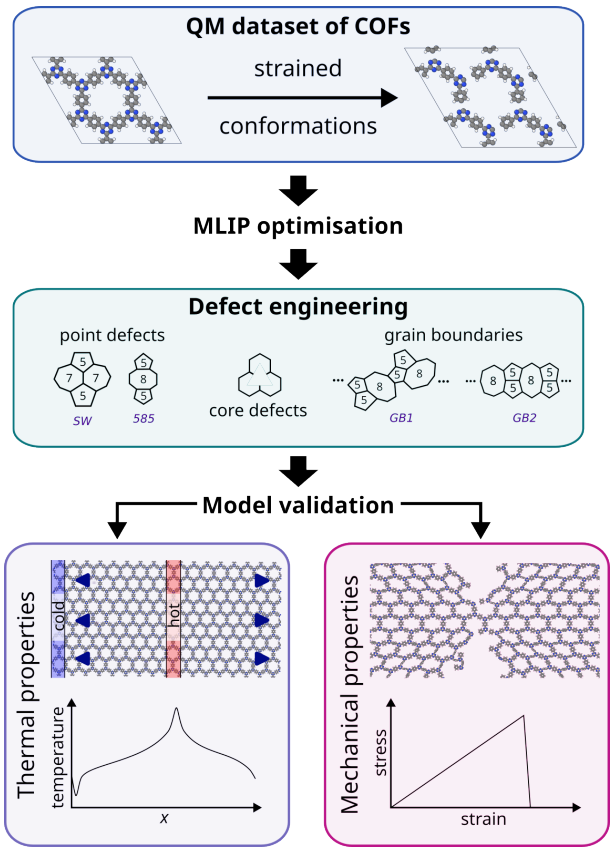

Figure 1. Schematic workflow from database via model training to property prediction. The MLIP is trained on a database of over 36 thousands monolayer structures, including highly strained structures. The model is validated on a range of defective structures, showing excellent transferability. Finally, it is used for elastic and thermal property prediction.