

# Predicting structures and properties of magnetic metal-organic frameworks (MOFs) by Crystal Structure Prediction

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Magnetic materials play a key role in modern technologies such as electric motors and data storage. However, many inorganic magnets, especially those that include rare-earth elements, often lack flexibility in their magnetic properties [1]. Metal-organic frameworks (MOFs), which are made from transition metal ions connected by organic linkers, have emerged as a potential solution. They allow for tunable magnetic behavior because the metal ions contribute magnetic moments, while the linkers help control how these moments are spaced and aligned. Still, figuring out the best combinations of metals and linkers is a major challenge due to the huge number of possible structures.

In our research, we use a theoretical method to explore magnetic MOFs by applying crystal structure prediction (CSP) through ab initio random structure searching (AIRSS) [2], along with Wyckoff Alignment of Molecules (WAM) [3]. AIRSS works by randomly placing metal nodes and linkers into trial unit cells, while WAM checks their symmetry and matches them with suitable crystal space groups. This combined method improves structure generation by making use of both molecular and crystallographic symmetry.

To determine which structures are most likely to form, we rank them by energy, which is usually done with periodic density functional theory (DFT). However, DFT can be very computationally expensive for large MOFs. To handle this, we use a machine-learned potential (MLP), specifically the MACE model. MACE is a machine learning software that combines equivariant message passing with efficient many-body interactions to predict atomic interactions and generate force fields [4].

We tested MACE potential's [5] accuracy on our study subject, which is Copper-based MOF74 (Cu-MOF74), by comparing its predicted energies to those from DFT calculations. The results showed a good match, with a low root mean square error (RMSE) of 0.032 eV/atom and a mean absolute error (MAE) of 0.0251 eV/atom. The coefficient of determination ( $R^2$ ) was 0.91, which indicates that MACE provides a reliable estimation of DFT energies. These results suggest that MACE can be a useful alternative for optimizing geometry and calculating energy in CSP workflows. Based on the MACE energy rankings, the low-energy structures can be further examined for their magnetic characteristics using periodic DFT calculations. This enables the investigation of magnetic ordering types and the strength of magnetic coupling, offering valuable insights for the targeted experimental synthesis of MOFs with potentially desirable magnetic behaviors.

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