

Exploring Phosphine Ligation States through QM/MM Simulations with Equivariant Graph Neural Networks

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For validation of a previously developed graph neural network (GNN) featuring anisotropic message passing investigation of ligation states of phosphine ligands within transition metal complexes was chosen. These ligands play a pivotal role in cross-coupling reactions. By integrating Cartesian multipoles as an anisotropic state into the neural network, the network captures directional information, enhancing the model's stability and transferability. The neural network was successfully used to replace the quantum mechanical calculations in a quantum mechanics/molecular mechanics (QM/MM) molecular dynamics (MD) simulations across 20 distinct phosphine ligands attached to a nickel benzaldehyde complex.

The model achieved a mean absolute error (MAE) for energies below 2.3 kJ/mol when it was trained on 43,000 data points that included energies, coordinates, and molecular multipole information. The model demonstrated good transferability for four ligands, not present in the training dataset, maintaining a mean absolute error for energies between 2.3 and 6.2 kJ/mol. Notably, it achieved chemical accuracy for three of these ligands. All complexes underwent stable simulations for extensive periods, spanning several hundred picoseconds. Remarkably, the simulation of the (PteroPhos)₂Ni(benzaldehyde) complex, encompassing 385 atoms, was executed efficiently. This was achieved by substituting the quantum mechanics calculation, which takes several hours, with the machine learning model that operates in just milliseconds.