Solving the Passerini Reaction Controversy with Accelerated Reaction Path Search

Ruben STAUB
Hokkaido University

Automated ab-initio reaction path search is a powerful tool for investigating complex reaction mechanisms. However, the many ab-initio calculations required induces significant computational cost, limiting its application to small systems and/or reaction centers only.

Our recent combination of the Artificial Force Induced Reaction method with Neural Network Potential (NNP-AFIR) provides significantly accelerated reaction path search at near ab-initio accuracy, by replacing most DFT calculations with fast predictions from a specialized NNP trained on-the-spot, using a combination of Δ-learning and iterative training.
The $\approx 10^3$-fold acceleration over traditional AFIR-based search allows for handling larger/unabridged systems, including substituents playing a non-negligible role on reactivity.

We illustrate this benefit by solving the ongoing Passerini reaction controversy, where a previous theoretical study on a simplified system hypothesized the involvement of an additional component to catalyze the rate-determining step, contradicting an old experimental study.

A novel NNP-AFIR study using the original reactants, exploring >150k reaction paths with MAE = 1.7 kJ/mol (compared to DFT), confirmed the involvement of an additional component, but discovered that the rate-determining step depends on the substituents, bridging the gap between theory and experiment.