Predicting hydrogen atom transfer energy barriers using Gaussian process regression

Predicting reaction barriers for arbitrary atomic configurations based on only a limited set of density functional theory (DFT) calculations would render the simulation of reactions within complex materials highly efficient. We propose Gaussian process regression (GPR) as a method of choice if DFT calculations are limited to hundreds or thousands of barrier calculations. For the case of hydrogen atom transfer (HAT), we obtain a mean absolute error of 3.23 kcal/mol using SOAP descriptors. We assess the uncertainty of HAT barrier predictions using the predictive distributions obtained directly from GPR as well as from an ensemble of a graph neural network-based model. Especially in the low-data regime, we find that GPR outperforms the latter with respect to various proper scoring rules. We suggest GPR as a valuable tool for an approximate but data-efficient model of chemical reactivity in a complex and highly variable environment.

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