Machine learning models to predict reactivity and products of metal-zeolites

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ABSTRACT: Developing easily accessible activity descriptors to predict possible intermediates and products of NRR [1] and CO_2RR [2] is of great importance. The free energy change (ΔG) for all possible reaction intermediate and product probability (P) of CO_2 reduction to methanol, methane, and formaldehyde on 26 single-atom catalysts (SACs) in zeolites were predicted by density functional theory calculations (DFT) and machine learning (ML) models [2]. The adsorption free energies of ΔG_{*OH} and ΔG_{*O*CH2} were highly correlated to catalytic activity. Producing methanol was favorable for metal-zeolites with early transition metal and main group elements. Methane production is more feasible for some systems such as Co-zeolite, due to the low free energy, and high selectivity against the hydrogen evolution reaction (HER). Both XGBoost and ExtraTrees algorithms could give satisfactory predictions of ΔG and P in CO_2RR using descriptors of reaction pathways, metal, charge transfer (CT) between metal and reaction intermediate, hydrogen bond interaction between intermediate and zeolite framework, and geometry. The CT feature could be replaced by some additional descriptors such as band gap (E_g) or coordination number of metal to intermediate in training ML models for free energy prediction. ML models on external test such as MOFs, 2D materials, and molecular complexes materials indicate that the proposed descriptors are general for the reaction free energy change and product prediction of SACs in CO_2RR. For NRR, we applied deep learning model with attention mechanism to gain an insight into the important role of hydrogen bonding between reaction intermediates and zeolites in increasing the energy costs of the desorption stage.[1]

Reference

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2. Qin Zhu, Yuming Gu, Xinyi Liang, Xinzhu Wang, Jing Ma, A Machine Learning Model To Predict CO2 Reduction Reactivity and Products Transferred from Metal-Zeolites. ACS Catal., 2022, 12, 12336−12348.