Alchemical Predictions for Computational Catalysis: Potential and Limitations

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Kohn–Sham density functional theory (DFT) is the workhorse method for calculating adsorbate binding energies relevant for catalysis. Unfortunately, this method is too computationally expensive to methodically and broadly search through catalyst candidate space. Here, we assess the promise of computational alchemy, a perturbation theory approach that allows for predictions of binding energies thousands of times faster than DFT. We first benchmark the binding energy predictions of oxygen reduction reaction intermediates on alloys of Pt, Pd, and Ni using alchemy against predictions from DFT. Far faster alchemical estimates yield binding energies within 0.1 eV of DFT values in many cases. We also identify distinct cases where alchemy performs significantly worse, indicating areas where modeling improvements are needed. Our results suggest that computational alchemy is a very promising tool that warrants further consideration for high-throughput screening of heterogeneous catalysts.
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