How to choose the right descriptor for electrochemical reactions?

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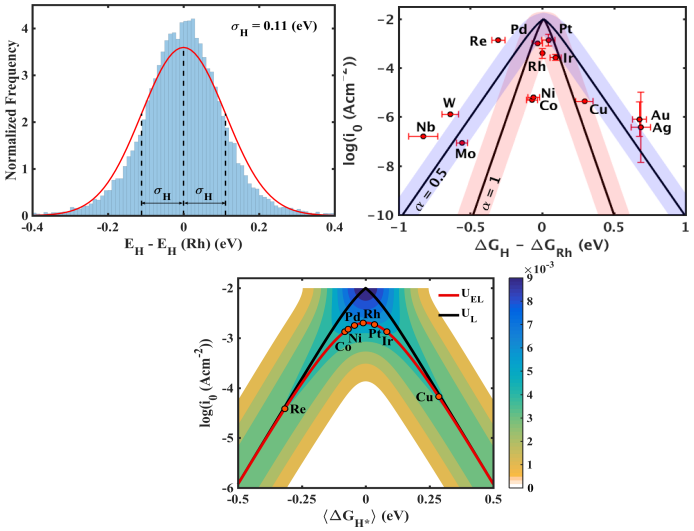
Introduction

- Successful theory-guided material design through descriptor-based search
- Robust theoretical framework – descriptor selection for various reactions.
- XC: BEEF-vdW with uncertainty estimation capability.
- Choice of descriptor – maximization of differentiability
- Prediction Efficiency – quantitative measure for distinguishability

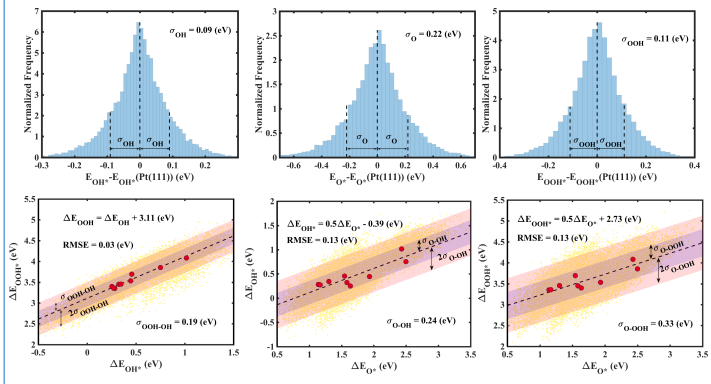
Hydrogen Evolution

- Volmer-Heyrovsky reaction mechanism
- Recent interests: Solar water splitting for hydrogen production
- Single Descriptor for activity: ΔG_{H^*}



Oxygen Reduction Reaction

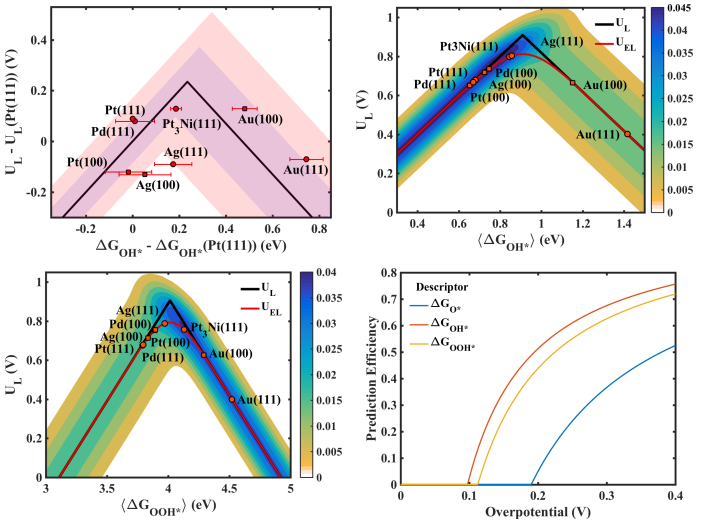
- Studied extensively both experimentally and through first principles DFT.
- Multiple activity descriptors: ΔG_{OH^*}, ΔG_{O^*}, ΔG_{OOH^*} - scaling relation
- $4e^-$ process - associative mechanism where oxygen is reduced to water
- $2e^-$ process – single intermediate OOH* - oxygen reduced to H_2O_2
- Quantitative basis for choosing descriptor – prediction efficiency



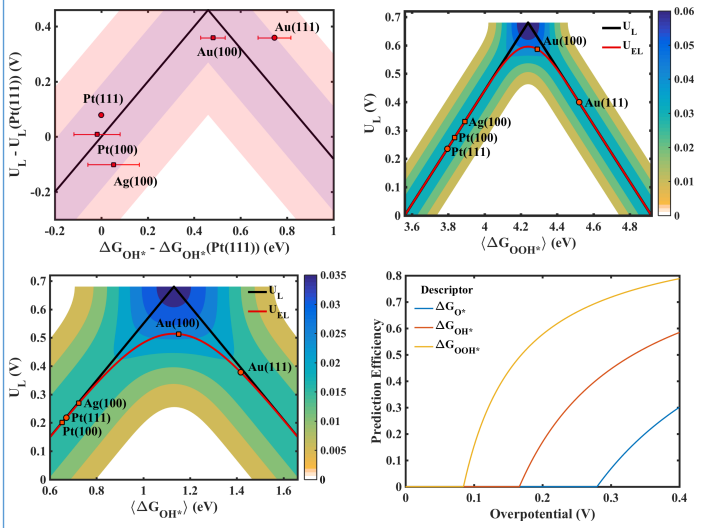
$$\sigma_{O*} > \sigma_{OOH^*} > \sigma_{OH^*}$$

$$\sigma_{O-OOH} > \sigma_{O-OH} > \sigma_{OH-OOH}$$

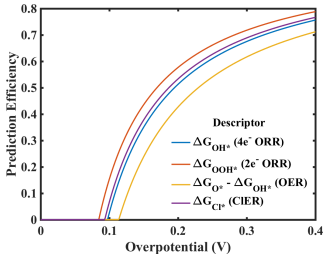
$4e^-$ ORR Process



$2e^-$ ORR Process



Comparing Predictability for Different Reaction Schemes



References

1. Nørskov, Jens Kehlet, et al., *J. Electrochem. Soc.* 152.3 (2005): J23-J26.
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