Combining DFT and Machine Learning to Enhance the Screening of Oxygen Evolution Reaction Catalysts

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The development and improvement of sustainable energy technologies is an urgent need to mitigate climate change, reduce environmental pollution, and ensure energy security for future generations. In this context, green hydrogen production can contribute to cut down carbon emissions by offering a clean, renewable source of energy. However, the current efficiency and cost of oxygen evolution reaction (OER) catalysts, particularly those relying on iridium (Ir), impose a major challenge that requires the discovery of new materials to envision the widespread adoption of green hydrogen production. The creation of mixed metal oxides presents a promising solution to reducing the dependence on Ir while maintaining or enhancing the catalytic efficiency, making the production of green hydrogen more economically viable and sustainable. Computational catalysis studies have facilitated the screening of mixed oxides as potential OER catalysts. Figure 1 illustrates the typical workflow for computational high-throughput screening for OER catalysts, which is composed of three tasks that have been performed solely based on expensive density functional theory (DFT) calculations [1]: i) creation ordered mixed metal oxide bulk systems and filter systems based on stability descriptors from bulk calculations; ii) generation of unique surface terminations with low Miller indices and calculation of simplified surface Pourbaix diagrams to determine likely intermediate coverages; iii) calculation of adsorbate interactions with the selected models to obtain theoretical overpotentials. At the end of the workflow, one can select systems that are possibly active based on the theoretical overpotentials, and stable based on stability arguments from bulk Pourbaix diagrams. The typical workflow is based on chemically ordered structures, which significantly reduce the total number of DFT calculations. Despite the advances that such a workflow brought to the field, it could also hinder the discovery of alternative materials with active sites based on different local chemical order. Here, we discuss how pre-trained machine learning force fields (MLFFs) [2] can be included in the catalyst discovery workflow to make the whole process computationally cheaper and allow for the exploration of a higher chemical space or the improvement of the workflow by allowing the exploration of distinct atomic ensembles for each system as potential active sites during the screening process. We show a workflow that includes DFT calculations, MLFFs, and data science tools to model mixed metal oxides. We discuss how our approach could be used to accelerate the screening of different materials or increase the insights offered to the community without significantly growing the associated computational cost.

References

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Figures

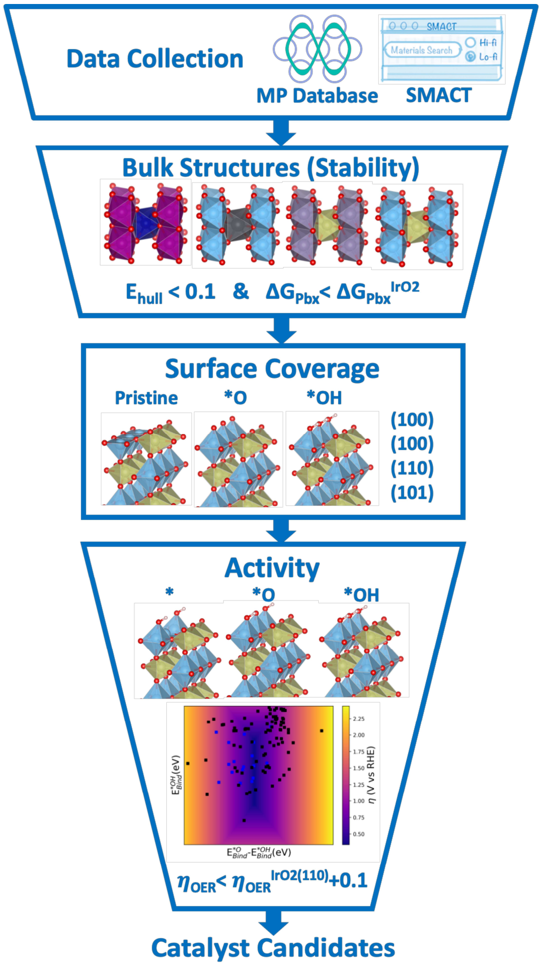

Figure 1. Workflow for computational high-throughput screening of oxygen evolution reaction catalyst candidates.