Machine Learning Assisted Discovery of Materials for Hydrogen Energy

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Abstract

Advancing hydrogen as a sustainable energy carrier requires both efficient catalysts for the green hydrogen evolution reaction (HER) and materials with optimal proton transport properties for proton exchange membrane (PEM) electrolyzer or fuel cells, enabling a comprehensive approach to clean hydrogen production and utilization.

For HER, electrochemical[1] and photochemical[2] technologies show advantages for high efficiency, selectivity, and scalability, making them the most promising approaches for producing hydrogen from H_2O or H_2S splitting. Metal sulfides have gained attention as promising catalysts for HER due to their unique electronic and optical properties, such as high charge transfer efficiency, narrower band gaps and abundant active sites, leading to enhanced catalytic activity[3]. To achieve efficient exploration of electro- and photo- catalysts for HER, supervised machine learning (ML) algorithms were employed to assist a large-scale screening of metal sulfide with optimal structural stability and reactive activity.To select the most suitable catalysts for HER, simple but thermochemical reasonable descriptors enabling the representation of catalyst properties and catalytic activities are essential for discovering or designing new catalysts via computational screening, including stability, band gap, and intermediate adsorption energy analysis. However, with the large number of possible metal sulfide compositions, facets and active sites, it is challenging to access all these descriptor values through experimental or DFT screening alone. Especially, intermediate adsorption energies on the metal sulfide surfaces are not readily available in online databases. Hence, we implemented a machine learning-aided screening approach to explore optimal metal sulfide catalysts over 881 MxSy lattices for HER. DFT was introduced as a computational tool to obtain the convergence capacity of 3680 adsorption structures and the H adsorption energies on the surfaces (around 2000), serving as targets for supervised ML models as shown in Figure 1. Four different supervised ML algorithms are selected, Artificial neural network (ANN), random forest (RF), support vector machine (SVM), and Gaussian process (GP). Overall, the best-trained random forest regression model can rapidly predict the energies over 10,000 unique adsorption structures and finally identified 10 and 37 potential metal sulfide lattices with optimal stability, band gap, and catalytic activity for HER. ML accelerates novel materials discovery by reproducing DFT results at lower research cost and interprets physically meaningful rules to bridge theory and experiments, facilitating efficient materials exploration and catalyst design.

PEM[4] are specialized materials designed to facilitate the flow of protons (H+) while blocking the passage of electrons and other gases, and they are important in hydrogen elecytrolyzer or fuel cells. The two-dimensional (2D) crystals hold the potential to significantly advance the field of PEM technology, characterized by exceptional molecular permeability and selectivity. Previous research has proved that understanding the energy barriers involved in proton permeation on 2D materials is essential for designing efficient proton-conductive materials[5]. ab-initio molecular dynamics (AIMD) simulations and machine learning (ML) techniques were used to predict and analyse proton permeation barriers in non-metal two-dimensional (2D) materials as shown in Figure 1. We calculated permeation barriers on around 500 2D materials through AIMD, thereby establishing a dataset that correlates 9 simple structural and electronic properties with proton permeation capacity, shedding light on the key determinants of proton permeation, which includes pore diameter, pore size and atomic electron affinity in modulating proton transport, offering insights into the design of advanced 2D materials for proton exchange membranes and fuel cells. Further AIMD simulations studied the selectivity for H_2/H^+ with low proton permeation barrier, which screened out 18 promising candidates including widely studied graphene, silicene, and h-BN, supporting the robustness and credibility of predictions. Notably, experimentally synthesized but underexplored materials for PEMs like germanene, cubic silicone, TeC, TeCl, GeSe and CSe, hold significant potential for investigation, while other candidates currently exist only as theoretically stable structures. Therefore, the integration of first-principles computational methods(DFT and AIMD) and ML accelerates the discovery of high-performance proton-conducting membranes and provides a deeper understanding of the fundamental mechanisms underlying proton permeation in non-metal 2D materials.

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Figures

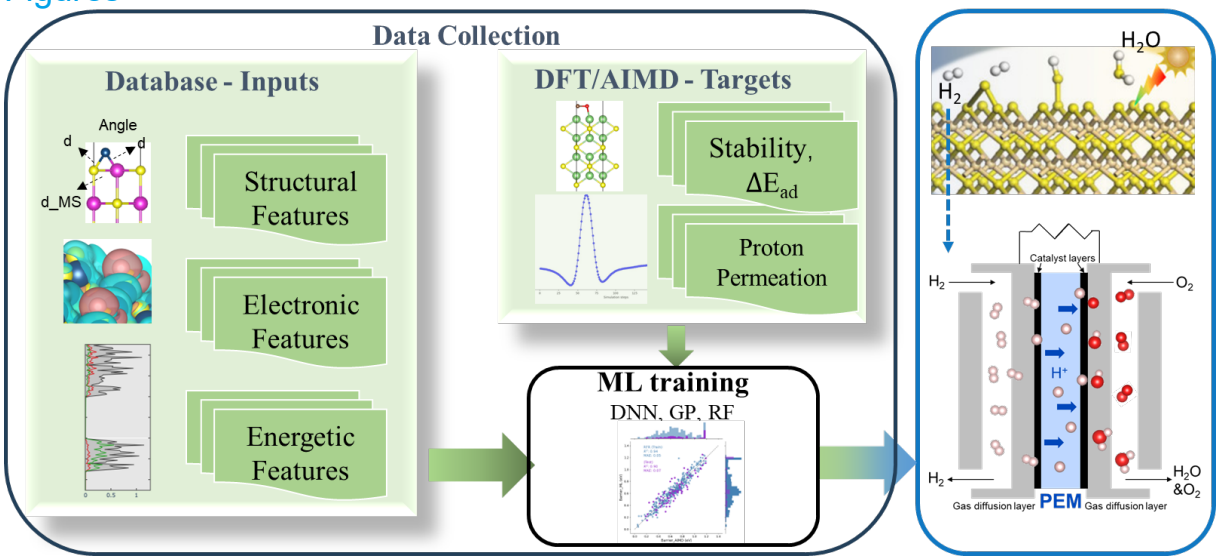

Figure 1. Schematic illustration of discovery materials for HER catalysts and PEMs.