Machine learning analysis of halide perovskite for photocatalytic CO_2 reduction and water splitting

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Significance and Relevance

Halide perovskites, well-known for their exceptional optical and electrical characteristics, have proven effective as light absorbers in solar cells. Similarly, these features such as strong light absorption, flexible bandgap tuning, and excellent charge transport are also crucial for photocatalytic applications. This research applies machine learning tools to explore how these properties can be leveraged to enhance the efficiency of CO_2 reduction and hydrogen evolution through photocatalysis. Random forest models were developed to predict the bandgap of perovskites and product formation rates considering the structure and properties halide perovskite as well as reaction conditions as the descriptors (input variables).

Preferred and 2nd choice for the topic: Preferred: Photocatalysis and Photoelectrocatalytic approaches, Solar Energy Utilization; 2nd: CO_2 utilization and recycling
Preferred presentation: Oral

Introduction and Motivations

Increasing environmental concerns have significantly boosted the demand for renewable and sustainable energy sources in recent years.[1,2] Harnessing solar energy and transforming it into alternative forms, such as chemical energy by producing solar fuels like hydrogen, methane or methanol, offers a promising strategy to address energy needs.[1] For instance, photocatalytic H_2 evolution is considered as sustainable and environmentally friendly process for generation of hydrogen fuel.[3,4] Similarly, photocatalytic CO_2 reduction (ie. artificial photosynthesis) is a novel way to produce valuable chemicals like hydrogen, carbon monoxide, methane, and methanol utilizing water and undesired CO_2.[1,5] These photocatalytic reactions require effective photocatalysts that can be activated by solar illumination and are stable within reaction conditions during processes.[5]

Halide perovskites, represented by the formula ABX_3 (where A and B are cations and X is a halide anion) have gained great attention as semiconductors due to their revolutionary performances in photovoltaics in last two decades. Although they could not be initially used for photocatalytic applications due to their inherent instability under humid conditions, this has changed with recent developments to improve their stability while their unique chemical structures allow for the easy tuning of properties like bandgap, light absorption efficiency, charge carrier mobility, and diffusion length to optimize photocatalytic performance.[1,6] Various halide perovskite materials with tailored properties have been developed experimentally for CO_2 reduction and H_2 evolution reactions, resulting in a significant body of research in the literature.

This study aims to gather insights on photocatalytic CO_2 reduction and H_2 evolution over halide perovskites from existing literature, compile a comprehensive dataset, and evaluate it via machine learning techniques. Key trends and influential factors were identified to create predictive models for the bandgap of halide perovskites, CO_2 reduction and H_2 evolution rates.

Materials and Methods

Photocatalysis over halide perovskites has been investigated in the literature for years between 2017 and 2024. The dataset was established using 115 articles on CO_2 reduction and 62 papers on H_2 evolution using the details of experimental conditions and recorded performance. Information on the

structure of halide perovskites, their synthesis methods, cocatalysts, cocatalyst deposition methods, reaction conditions, illumination details were used as descriptors while the total product formation rate was taken as the output variable. Photocatalytic CO_2 reduction and H_2 evolution reactions handled separately; however, all the perovskites used as photocatalysts in the dataset were examined together. The dataset was analyzed using descriptive statistics and machine learning methods to identify important variables and patterns, which were subsequently used to predict the perovskite bandgap, CO_2 reduction and H_2 evolution rates (expressed in terms of total product formation rate). The power of predictive models was assessed according to their root mean squared error and r-squared values. The relative importance of descriptors was also analyzed using The SHAP (SHapley Additive exPlanations) analysis.

Results and Discussion

The dataset was first analyzed using simple descriptive statistic to understand the major trends and identify the important descriptors for the predictive models. Then, a predictive model for the bandgap of perovskites were constructed first to compute its missing values in the dataset and computed values, together with the experimental bandgaps, were used (as one of the main descriptors) for the predictive models for CO_2 reduction and H_2 evolution processes. Figure 1 shows the prediction results of random forest models developed for the bandgap of halide perovskites and total product formation rate for CO_2 reduction; the models successfully predicted both bandgap of perovskites with RMSE of 0.12 (R^2=0.92) and the CO_2 reduction with RMSE of 0.33 (R^2=0.91). Analysis showed that the chemical components of perovskites were highly influential for photocatalytic performance, and hot injection was the most common perovskite synthesis method.

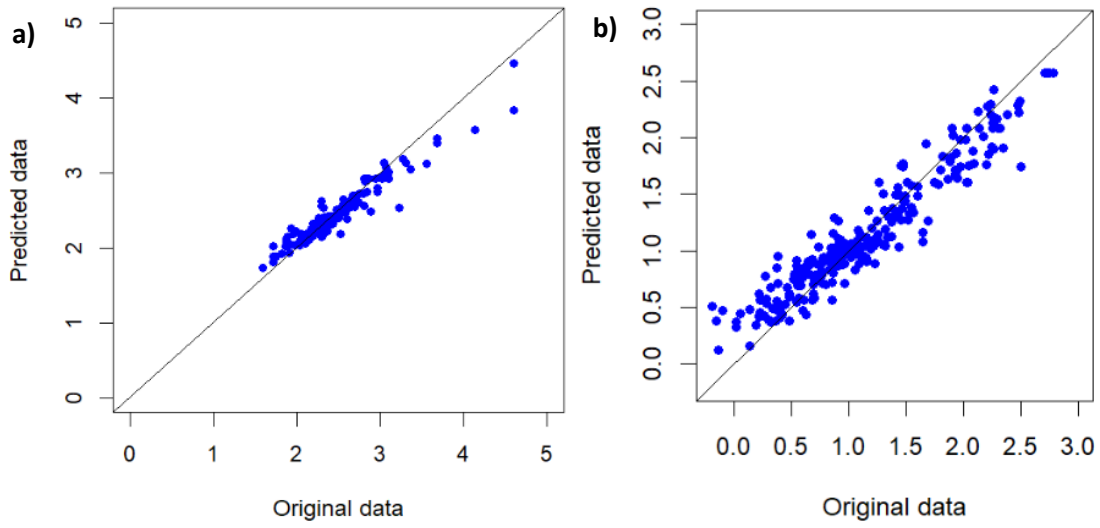

Figure 1 Results of random forest prediction models for a) bandgap b) total gas production rate.

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