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Short Communication

High-throughput screening of MXenes for hydrogen storage via graph neural network

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ABSTRACT

To better understand the recent experiment on hydrogen storage in MXene multilayers [Nature Nanotechnol. 2021, 16, 331], we propose a multiscale workflow to computationally screen 23,857 compounds of MXene for hydrogen storage in near ambient condition. By using density functional theory simulation to produce the dataset, we trained physics-informed atomistic line graph neural networks to predict hydrogen's adsorption performance on MXenes, which is further validated through grand canonical Monte Carlo simulation. As a result, ScTC is identified to exhibit a hydrogen storage capacity of 5.7 wt% at 230 K and 100 bar, showing the promise for hydrogen storage.

1. Introduction

The increasing concentration of carbon dioxide in the atmosphere has resulted in global warming, climate change, and a host of other environmental issues. One of the most promising solutions to achieving carbon neutrality is to use hydrogen energy, which has high gravimetric energy density, renewability and zero CO_2 emission, showing the potential to replace fossil fuels in the future's sustainable energy system [1–4]. However, one main obstacle that hinders the vast commercial application of hydrogen energy is lack of hydrogen storage materials with both high gravimetric and volumetric density under near ambient conditions [4,5]. The search target set by US Department of Energy for 2025 requires 5.5 wt% and 40 g/L hydrogen with an operating temperature range of −40 °C to 60 °C [6]. To balance the energetics of storage and kinetics of release, the desirable adsorption energy window is 20 ~ 80 kJ/mol for fast reversible hydrogen storage under near ambient temperature and moderate pressure (less than 100 bar).

In a recent experimental study, S. Liu et al. [7] applied incompletely etching strategy to prepare multilayer Ti_3CT_x (T: functional group) that has an unprecedented 8.8 wt% hydrogen uptake at room temperature and 60 bar pressure. The storage mechanism was attributed to a nano-pump-effect-assisted weak chemisorption due to the existence of fluorine functional group and narrow interlayer distance (~ 7 Å). From the computational aspect, several studies [8–12] discussed the adsorption mechanism and hydrogen storage performance of specific compounds of MXene. Nevertheless, the current studies have only covered a very small part of the MXene compounds as compared with the more than 23,000 MXenes as reported in MXenes database (aNANt) [13].

To explore the hydrogen storage materials more effectively and efficiently, it is necessary to apply high-throughput computational screening together with machine learning methods [14–18]. For physisorption type hydrogen storage materials, Ahmed and Siegel [18] selected 8,282 candidates for hydrogen storage by screening 918,734 metal organic frameworks through extremely randomized trees algorithm and GCMC simulation. For chemisorption type hydrogen storage materials, Rahnama et al. [15,16] predicted the hydrogen weight percentage and optimal material composition of metal hydrides with several supervised machine learning models based on Hydrogen Storage Materials Database. For quasi-molecular adsorption governed by Kubas interaction, however, no studies are reported to utilize high-throughput computational screening with machine learning. The main underlying reason lies in fact that the interactions with hydrogen are more complicated due to electron donation and back-donation between transition metal's unfilled d orbitals and hydrogen's σ orbital [19], so it is expensive and time-consuming to precisely calculate the adsorption energy of hydrogen on adsorbent, and high level computational methods are required [20,21] which is different from physisorption [22] where the weak Van de Waals interaction can be reliably described by non-

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