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AI Guided Membranes Materials Development for Hydrogen Evolution and Separation

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ABSTRACT

Hydrogen energy is one of the potential clean energy sources that could be used on a large scale. Membrane-based hydrogen evolution and separation is one of the most promising approaches to low-cost hydrogen production. However, suitable membrane technologies are lacking and the development of advanced materials needs to be accelerated. In this paper, we provide a mini review of artificial intelligence (AI) applications to hydrogen separation membrane and hydrogen evolution membrane reactor discovery. By referring to the AI-guided development cases, readers will obtain a concise perception of popular machine learning (ML) methods and how they work to realize targets in specific application. We aim to assemble ML methods with the membrane materials development process. Current limitations to be addressed and prospect of AI applications in membrane discovery are also highlighted in the conclusion.

Keywords: Hydrogen energy, Machine learning, Separation membranes, Membrane reactors

NONMENCLATURE

Abbreviations	
AI	Artificial Intelligence
DFT	Density Functional Theory
GHG	Greenhouse Gases
MC	Monte Carlo
MD	Molecular Dynamics
ML	Machine Learning

1. INTRODUCTION

With cumulative releases of greenhouse gases (GHG) by human activities, mainly CO_2, leading to global climate change, net emissions of anthropogenic CO_2 from not only agricultural and industrial production but also land use and energy services—must approach zero, which is also known as Carbon Neutralization or Net Zero. Hydrogen energy or Hydrogen gas (H_2) is regarded as one of the most potential alternatives to fossil energy. However, large-scale hydrogen application is facing many challenges, and low-cost hydrogen production is one of the bottlenecks. Membrane science and engineering is a promising approach to low-cost hydrogen evolution and separation, via membrane reactors and separation membranes. However, the challenge of predicting a precise structure-process-property relationship to effectively design advanced and satisfactory membranes has blocked the application of membrane technologies. The challenge can be attributed to three reasons: features of high dimensionality in membrane design such as membranes' intrinsic nature (e.g. constitution, structure, pore size , and surface properties) and extrinsic conditions (e.g. reaction temperature, concentration and pH); vast design space of potential membranes that is almost impossible to exhaust (e.g. organic, inorganic, metal-organic framework (MOF) and mixed matrix membranes); Incomplete understanding of physical and chemical properties of complicated membrane systems (e.g., selective layers' thermal and chemical stability, catalyst layer/membrane interface). The complexity of experimental data exceeds human's analytical capability, which slows down advanced membrane development.

Apart from experimental methods, computational simulation methods such as density functional theory

(DFT) and molecular dynamics simulation (MD) can simulate membranes' performance without real membrane synthesis. The computational simulation methods access the answer through another approach which is comparatively low-cost. Computational chemistry originated from structure-property calculation, then progressed to crystal structure prediction and grew into statistically driven design in current use. In membrane research, the first two stages have been already achieved in gas, liquid and fuel cell separation membranes. However, the expense of computation surges with the complexity of systems, which greatly limited simulations in low time scale and small space scale.

With the rapid development of artificial intelligence (AI), especially machine learning (ML), data-driven techniques have been successfully applied to interdisciplinary area. By setting proper metrics, giving well-modulated datasets and using suitable ML algorithms, a precise and accurate model for certain tasks is obtained. With the challenges in analysis of high dimensional information and efficient search of the huge material space emerging, AI shows potential in accelerating membrane development. In fact, its irreplaceable advantages over human are its inherent capacity in handling massive and high-dimensional data, which is the key of AI applications for membrane research.

In this review, we aim to show the promising potential of ML in membrane technology through each step of membrane development. A general introduction to practical ML algorithms in membrane science is firstly proposed. Various applications of AI in different material systems are then discussed with representative works in recent years. In the end, we show our perspective toward current limitations and future directions.

2. AI ACCELERATED DISCOVERY OF MATERIALS FOR HYDROGEN EVOLUTION AND SEPARATION

2.1 Introduction

A typical membrane design process can be divided into following stages: material, membrane, separation, and operation. The initial stage is searching for proper materials, mainly depends on the membrane's separation targets. Synthesis protocol designing is followed, which needs to take extrinsic and intrinsic factors into account. After the membrane is fabricated, researchers use a series of index to characterize it. Then separation performance is test and discussed, including permeability, solubility, and selectivity. By analyzing the experiment data, some mechanism or predictive mode may be proposed. Finally, enhancing membrane's resistance to fouling, degradation and plasticization is the key of operation. At laboratory and industry scale,

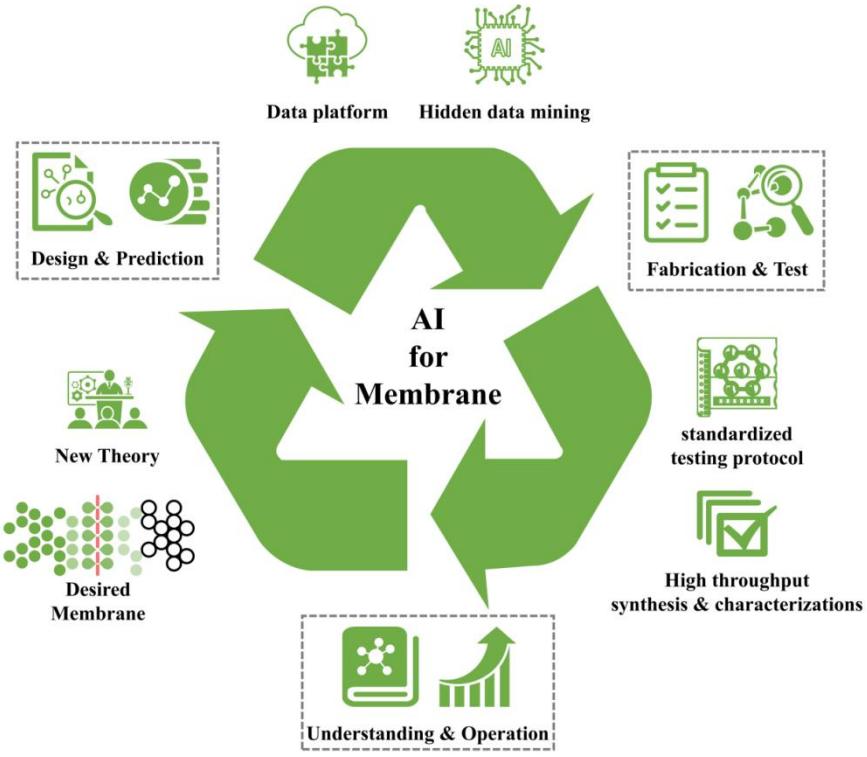

Fig. 1. Ideal AI-aided membrane discovery and development process

various functional membranes are constantly discovered and manufactured during numerous iterations. However, it's becoming more and more difficult to exhaust the vast membrane design space via experimental methods thus a new mode should be employed.

In Figure 1, we illustrate an ideal membrane discovery process, which contains three stages. AI-aided membrane development starts from design and prediction based on existing data. Standardized testing protocol is the basis of AI-aided membrane test and fabrication, which severely lacks data up to now. With AI's inherent capacity of analyzing high-dimensional data and recognizing particular pattern which is far more powerful than human, high throughput synthesis and characterization is no more impossible. Based on newly attained data, another round of iteration can be implemented. The iterations end till desired membrane is discovered, and new theory may be extracted from database that is enlarged during iterations. The article focused on the initial stage, searching proper materials, which mainly depends on the membrane's separation targets, via two cases in two material systems.

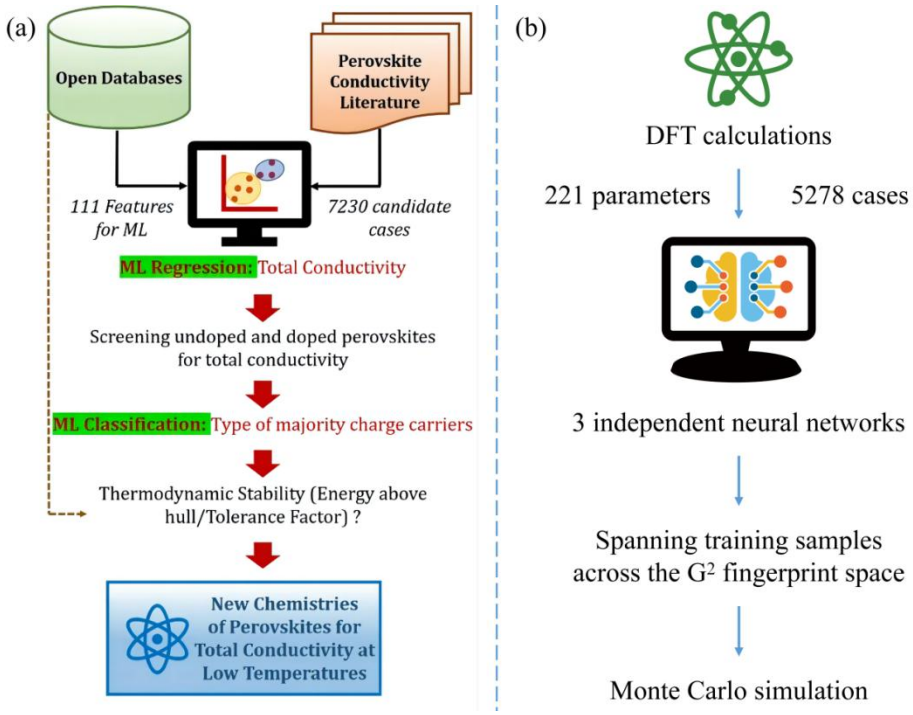

Fig. 2. AI accelerated discovery of materials for hydrogen evolution and separation. (a) Schematic of design and discovery of perovskites through machine learning [5]; (b) An illustration of [11]'s framework.

2.2 AI-guided Membrane Materials Discovery Cases

ABO_3 is chemical formula for a large group of chemicals. In material science, $CaTiO_3$ or perovskite oxides and its material system are a hotspot. ABO_3-type perovskite oxides are a mature material system, which is widely used in energy field. A critical part of gaseous hydrocarbons dehydrogenation reactors is dense proton-conducting membrane [1, 2]. Perovskite oxides are widely used in electrocatalytic hydrogen evolution [3, 4]. To achieve high-throughput screening of perovskite oxides, Priya and Aluru [5] used machine learning tools including Bayesian ridge regression, linear regression, support vector machines, k-nearest neighbor regression, random forest (RF) regression and neural networks, as Figure 2 (a) shows. They collected more than 7000 data points from open database and published literature to build up training and testing set. A comparison of different methods' output showed XG-boosted RF algorithm provided the best output. To report good-conductive perovskites, the model screened 1793 undoped and 95,832 A-site and B-site doped perovskites. A list of perovskites with predicted conductivities is given, which contains not only many perovskites studied by previous researchers but also several new candidates.

PdCuAu ternary alloys are an ideal material system for hydrogen separation. Compared with pure Pd membrane, PdCuAu ternary alloy membrane mitigates hydrogen embrittlement and H_2S poisoning [6-9]. The Pd-membrane based hydrogen separation starts at dissociative adsorption, then diffusion, and end up with desorption [10]. Concentration in surface and bulk are

main factors of the three steps. Yang et al. [11] built a neural network model to calculate the slab configurations' potential energies with accuracy comparable to DFT's while making computing expense affordable, and Monte Carlo simulation (MC) of a system containing enormous atoms which was impossible previously was achieved. Samples in training set were tested in the G^2 fingerprints space, which showed that the selected 3*3*5 units are large enough to span almost all atomic environment, thus the neural network's capacity of predicting the potential energy for larger units in MC simulation was guaranteed. With a given bulk concentration, MC process could calculate the average surface concentration via 15000 successful MC steps, and a representative MC process was illustrated by figure 2 (b). A simulation of segregation profile in PdCuAu were reported and it was important to investigate the separation performance. The simulation results are quite correct along PdAu and CuAu lines while inconsisitent with experimental data for PdCu.

2.3 Discussion and Conclusion

It is a common fact in artificial academia that machine learning algorithm usually requires massive data for training. In the membrane science field, however, data is much more difficult to acquire due to experimental limitations. Insufficient data may magnify the influence of experimental error and could lead to a series of issues, such as overfitting and underfitting. Apart from the lack of data, a rigorous scientific process of designing and evaluating AI model should be emphasized.

Future work may also focus on practical approaches on the augmentation of existing data set to compensate for the lack of data, such as hidden data mining. In order to obtain more data, we can also make minor changes to the existing data set. Taking image data for example. Flips, translations, rotations to the image will let the network consider it as a different image. Another perspective to improve the machine learning pipeline is to achieve a higher data sampling efficiency via active learning.

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