Research Highlight

Accelerating catalysts design by machine learning

Haishan Yu, Jun Jiang *

Hefei National Laboratory for Physical Sciences at the Microscale, CAS Key Laboratory of Mechanical Behavior and Design of Materials, School of Chemistry and Materials Science, University of Science and Technology of China, Hefei 230026, China

The climate crisis associated with rapid accumulation of carbon dioxide in atmosphere has posed increasing threat to human well-beings [1]. It is of great significance to capture and convert CO_2 to realize sustainable utilization of carbon resources [1]. The electro-catalytic CO_2 reduction reaction (CO_2RR) provides a sustainable, carbon-neutral method for solving this problem. Yet the difficulty of finding an efficient catalysts slows down the realization of the ambitious goal. It is mainly because it requires simultaneous optimization of selectivity for a specific product, energy conversion efficiency, current density and stability of the reaction system. Therefore, it is desirable to develop an efficient approach towards identification of electrocatalysts for CO_2RR.

Computational chemistry makes the discovery of new catalysts more predictive, yet the huge space for catalytic materials also makes the screening of catalysts rather inefficient using conventional computational chemistry based on quantum mechanics. Luckily, artificial intelligence techniques based on machine learning algorithms can reduce the barrier between materials design and modeling, which appreciably speed up the process. For example, O'Connor et al. [2] established a quantitative relationship between descriptors and adsorption energy through machine learning, which can predict the strength of interaction between individual metal atoms and oxide supports, and help to design sinter-resistant single-atom catalysts. Sun et al. [3] used a machine learning approach to conduct screening of graphdiyne-atomic catalysts for hydrogen evolution reaction (HER) emphasizing the relationship between adsorption energy and electronic states. Wang et al. [4] proposed electric dipole moment as a promising new type of descriptor for catalyst design because it can be used to quantitatively predict molecular adsorption energy and transferred charge, which are two key parameters of surface-molecular adsorbate interactions.

Back to CO_2 reduction, selective reduction to ethylene has been found to be related with the oxidation state, crystal surface and additives of Cu by experiments [5–7]. To date, the best reported catalyst still suffer from small local current density (7 mA cm^{-2}), low Faradaic efficiency (up to 60%), and low energy efficiency. In the latest issue of *Nature*, Sargent and co-workers [8] reported a breakthrough for the electrocatalyst that converts CO_2 to ethylene with substantially better efficiency than ever before, by means of density functional theory calculations and active machine learning. They investigated 244 different copper-containing intermetallic compounds, and enumerated 12,229 surfaces and 228,969 adsorption sites. Machine learning was used to screen catalysts based on the indicators of electronegativity on the surface of the material, the adsorption energy of CO and H, and the effective coordination number. Combined with the volcano scaling relationships, the excellent performance of Cu-Al electrocatalyst was predicted for the first time. The results show that the abundance of Cu-Al adsorption sites and site types is the highest, and the CO adsorption energy is close to the optimal value (ΔE_{CO} = −0.67 eV). This indicates that the Cu-Al alloy may be active in a relatively wide range of surface compositions and site types. The Cu-Al catalyst greatly reduces the reaction energy barrier of CO_2 activation hydrogenation, C–C coupling, and C–H bonding, thereby effectively promoting the formation of the multi-carbon product (C_2H_4). Subsequently, the authors verified this prediction through experiments. The C_2H_4 Faradaic efficiency of the Cu-Al catalyst is about 60% at a current density of 600 mA cm^{-2} in a 1 mol L^{-1} KOH electrolyte, which is higher than the C_2H_4 Faradaic efficiency of the pure Cu catalyst of 35%. Moreover, the CO Faradaic efficiency of the Cu-Al catalyst is suppressed to about 10%, which is one third of that of pure Cu.

In order to further optimize the Cu-Al catalyst, the authors developed a method of thermal evaporation and co-sputtering followed by chemical etching to synthesize the de-alloyed nanoporous Cu-Al catalyst, and realized the adjustment of the alloy surface component content. The de-alloyed nanoporous Cu-Al catalyst has catalytic active sites with high geometric density, which is conducive to the adsorption and electroreduction of CO_2. In a 1 mol L^{-1} KOH electrolyte, the Cu-Al alloy catalyst has a Faradaic efficiency of 80% for the conversion of CO_2 to C_2H_4 at an ultra-high current density of 600 mA cm^{-2}. The C_2H_4 Faradaic efficiency of 82% can be achieved at a current density of 400 mA cm^{-2}, and the Cu-Al catalyst has a stability exceeding 100 h. Furthermore, the authors also studied Cu-Al performance under different pH conditions and found that it can get good results in the 3 mol L^{-1} KOH and 3 mol L^{-1} KI electrolyte. As long as the current density of 150 mA cm^{-2} is maintained, the Faradaic efficiency of CO_2 to C_2H_4 is 73% ± 4% and the energy conversion efficiency of cathode electrical energy to chemical energy is 55% ± 2%.

Using *in situ* synchrotron X-ray absorption spectroscopy and theoretical calculations, the authors found that a high-performance

* Corresponding author.
  E-mail address: jiangj1@ustc.edu.cn (J. Jiang).
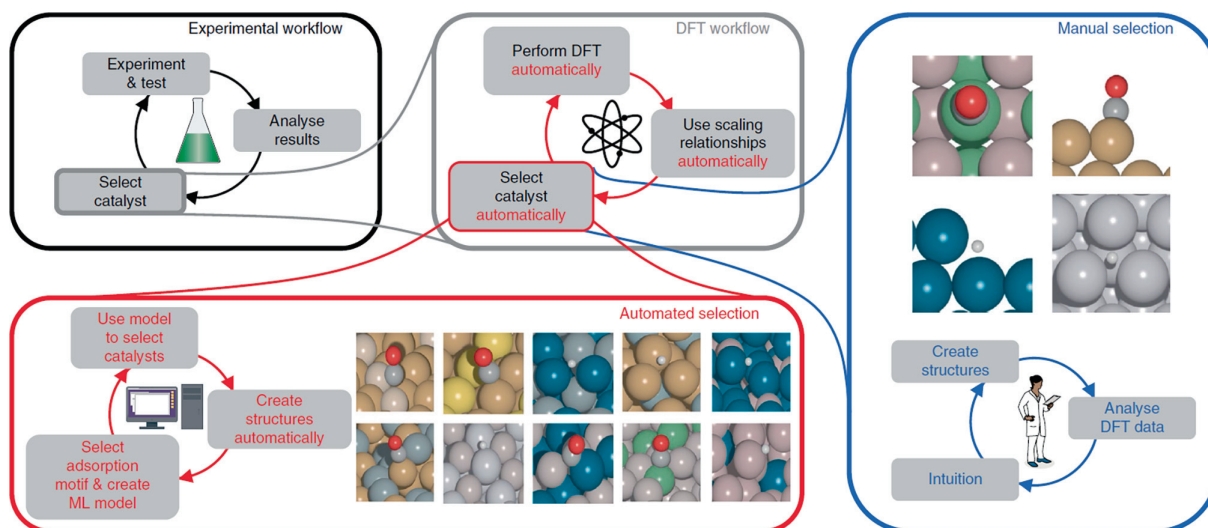
Fig. 1. Workflow for automating the discovery of theoretical catalysts with combining DFT calculations and machine learning. This figure was taken from Ref. [10]. Copyright © 2020 Springer Nature.

Cu-Al catalyst benefit from a favorable Cu coordination environment that enhances C–C dimerization. This work clearly demonstrate the critical role of theoretical calculations and machine learning in accelerating discovery of new electrocatalysts of CO_2RR in experiments.

With the high accuracy and convenience of DFT theoretical calculations, machine learning can conduct high-throughput screening of complex catalytic systems for saving enormous experimental time and costs [9]. Workflow for automating theoretical materials discovery (Fig. 1) was reported by Tran and Ulissi [10] who used machine learning to guide DFT calculations for screening CO_2 reduction and hydrogen-producing electrocatalysts. In the face of the diversity of selection combinations, machine learning can significantly accelerate the design of new catalytic materials and guide experimental discovery of new catalysts.

Conflict of interest

The authors declare that they have no conflict of interest.

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Haishan Yu received his bachelor degree in Materials Science in 2013 at Tiangong University. Presently, he is a Ph.D. student in Prof. Jiang's group at University of Science and Technology of China. His current research interests are the DFT calculations of two-dimensional materials and the application of machine learning in material science.

Jun Jiang received his B.S. degree in Physics in 2000 at Wuhan University, and Ph.D. degree in Theoretical Chemistry in 2007 at Royal Institute of Technology, Sweden. After three-year Post-doc study at University of California Irvine, USA, he joined the University of Science and Technology of China in 2011 as a Professor in theoretical chemistry. His research interests focus on the development and application of multi-scale modeling methods and machine learning techniques for electron kinetics in complex system, while targeting on a wide range of physics or chemistry applications.