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Recent Advances in The Data-Driven Development of Emerging Electrocatalysts

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Abstract

Data-driven strategies have proven efficient for the design of high-performance electrocatalysts from the vast material search space. In this review, we present an overview on data-driven approaches to emerging electrocatalyst design: high-throughput experiments, high-throughput calculations, and machine learning. High-throughput experiments facilitate rapid synthesis and characterization of electrocatalysts, leading to efficient exploration of various materials. High-throughput calculations predict and screen materials' properties, allowing for the identification of promising electrocatalysts. The integration of machine learning further augments these high-throughput approaches through critical insight extracted from the large dataset, fast prediction of materials' performance, and optimization of materials discovery. Employing these data-driven strategies synergistically could accelerate the development of electrocatalysts. Such advancements could promote green energy technologies and substantially contribute to mitigating grand challenges posed by global climate change.

1. Introduction

Green energy is of paramount significance to combating global warming and alleviating our heavy reliance on fossil fuels. However, green energy resources such as solar and wind power are inherently intermittent, which need to be circumvented in pursuit of reliable and stable energy supplies. In this context, (opto-)electrochemical reactions offer promising solutions to conversion, storage and utilization of intermittent green energies [1]. To date, many electrochemical reactions have been explored, as depicted in Figure 1. Driven by renewable electricity, for instance, hydrogen evolution reaction (HER) [2] and oxygen evolution reaction (OER) [3] can generate hydrogen from water, which has been widely considered a promising carbon-free energy carrier. Carbon dioxide, one of the main causes of global warming, can be reduced to value-added chemicals such as methane, ethylene, and ethanol via carbon dioxide reduction reaction (CO_2RR) [4]. Moreover, nitrogen reduction reaction (N_2RR) [5] combined with green energies is an emerging alternative to the energy- and carbon-intensive Haber-Bosch process for ammonia production.

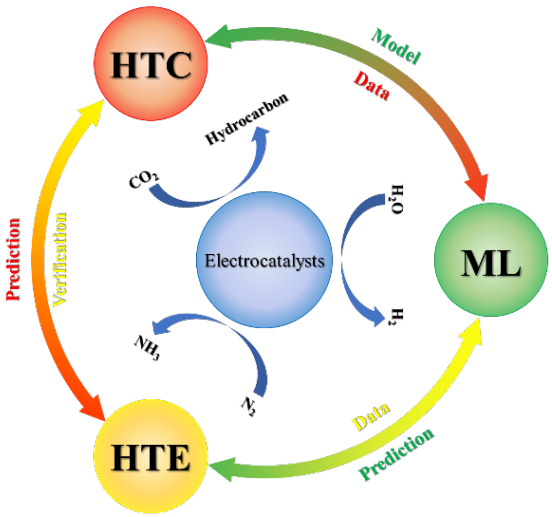

Figure 1. Schematic illustration of applications of high-throughput experiments, high-throughput calculations, and machine learning in electrocatalyst design (HTE: high-throughput experiments; HTC: high-throughput calculations; ML: machine learning).

The heart of these electrochemical reactions is high-performance electrocatalysts. However,

most current electrocatalysts are based on expensive and scarce noble metals, *i.e.*, Pt-group based materials for HER, IrO_2 and RuO_2 for OER, and Ru for N_2RR [6-12]. For large-scale applications, potential electrocatalysts should be not only highly performant but also cost-effective. Yet, it remains a grand challenge to efficiently identify such electrocatalysts from hundreds of thousands of synthesizable materials, especially when traditional case-by-case experiments or first-principles calculations are employed [*13]. Recently, data-driven approaches leveraging high-throughput techniques and machine learning have been demonstrated efficient and necessary for electrocatalyst explorations across the vast materials search space [14]. In this review, we delve into the recent progress in high-throughput experiments, high-throughput calculations, and machine learning in the realm of electrocatalyst development. By exploring the integration of these cutting-edge techniques, we highlight their transformative potential in revolutionizing electrocatalyst design and driving advancements in sustainable energy technologies.

2. High-Throughput Experiments

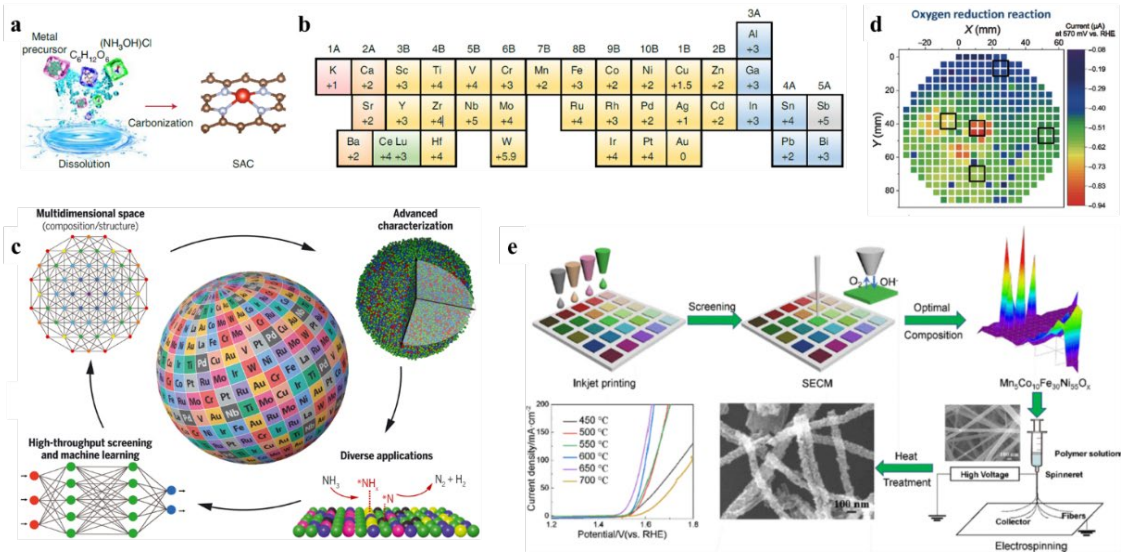

Figure 2. (a-b). An illustration of single atom catalyst (SAC) synthesis based on 37 metallic elements. [**15] (c). A workflow for developing HEA electrocatalysts. [**16] (d). A mapping of ORR activity at 0.57 V vs RHE in the (TiNi)-Cu-Hf-Pd-Zr alloy systems. [17] (e). The high-throughput experimental screening of the Mn-Co-Fe-Ni oxide library for OER. [18]

High-throughput experiments refer to a systematic approach, by which multiple experiments are conducted simultaneously or in rapid succession. Compared to traditional methods, high-throughput experiments allow more efficient explorations of broader materials space at reduced costs [19-24]. It should be noted that universal or general synthesis or characterization methods are required to achieve high-throughput experiments. For instance, Xin, et al. [**15] developed a dissolution-and-carbonization method to synthesize single atom catalysts (SACs) based on 37 different metallic elements, as depicted in Figures 2a-b. These synthesized SACs constitute one of the largest libraries of SACs reported to date, offering a wealth of potential candidates for various catalytic applications. Interestingly, the SACs containing 12 different single metallic atoms exhibit a much higher activity towards OER than the SACs containing only one type of the constituent single metallic atom. This manifests the subtle yet significant synergistic effects among SACs, which is worthy of future investigations.

High-entropy alloys (HEAs), which are near-equimolar alloys of five or more elements, have attracted tremendous attention in the catalysis community in recent years. HEAs feature abundant elemental combinations and composition ratios, and infinite surface configurations, as illustrated in Figure 2c. These two enable the binding strength of adsorbates or the catalytic activity to be fine-tuned in a near-continuous way. On the other hand, they also render high-throughput experiments inevitable to thoroughly explore the HEAs even with a fixed elemental combination. In this process, co-sputtering of multiple metal sources and shock-based syntheses are typically employed[**16]. For instance, Andronescu, et al. [17] developed a high-throughput process for co-sputtered material libraries and performed high-throughput characterizations, as illustrated in Figure 2d, leading to the drastically accelerated identification of previously unknown HEA compositions of (TiNi)-Cu-Hf-Pd-Zr with a superior catalytic activity towards either oxygen reduction reaction (ORR) or HER.

Regarding high-throughput characterizations, scanning electrochemical cell microscopy (SECCM) in combination with mobile droplet cells is a simple yet effective method to measure the catalytic performance of batches of electrocatalyst samples [25, 26]. SECCM employs a single/dual-barrel probe from which a thin layer of electrolyte protrudes. This results in a well-

defined meniscus contact with a substrate surface, thus defining an active surface area of an electrochemical cell that can precisely be visualized and measured at the micro/nanoscale [27]. This technique has been widely adopted in investigations of HEA-based electrocatalysts [26]. Recently, Li, et al. [18] also applied SECCM to screen a library of inkjet-printed quaternary Mn-Co-Fe-Ni oxides for OER and found that $Mn_5Co_{10}Fe_{30}Ni_{55}O_x$ has the highest activity, as illustrated in Figure 2e. With Pd nanocrystals as a model system, Zeng et al. [*28] deployed SECCM and observed that strained Pd icosahedra displays improved HER activity to unstrained Pd octahedra of similar size, offering direct experimental evidence of the surface strain dependence of catalytic activity. By means of SECCM, the quantitative correlation between local surface strain and catalytic performance may even be scrutinized as well in the near future. In addition to SECCM, other techniques such as rotating disk electrode (RDE) [29] and electrochemical impedance spectroscopy (EIS) [30] have also been adapted for high-throughput analysis of electrocatalysis.

3. High-Throughput Calculations

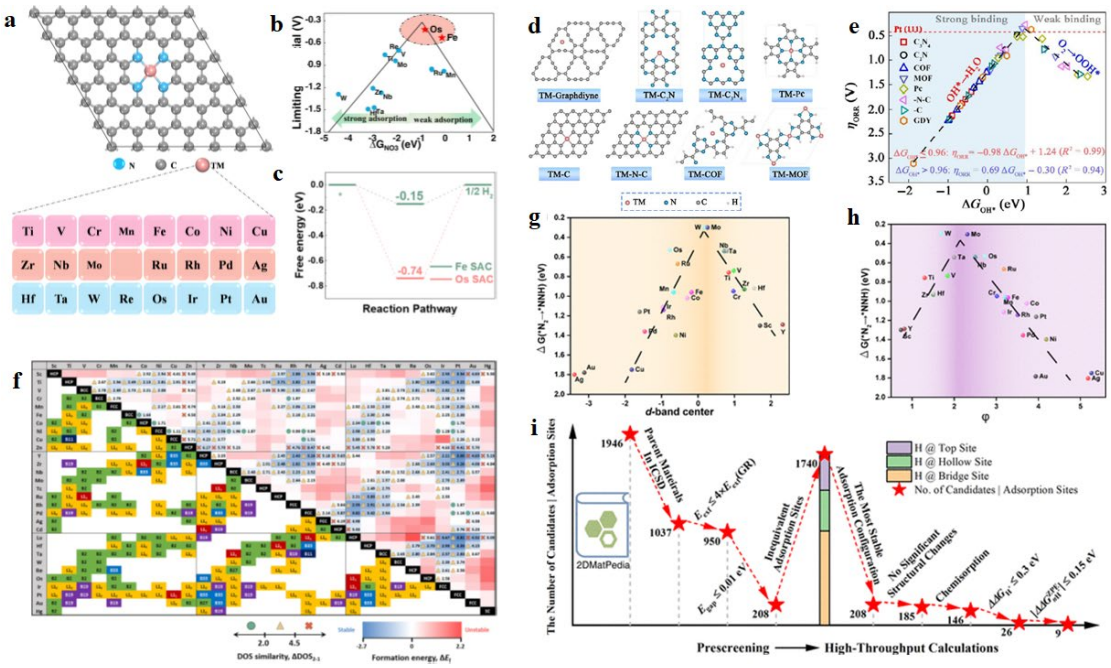

Figure 3. (a-c). The atomic structure of the transition metal SACs, (atom label: C = gray, N = blue, and transition metals = pink) and the corresponding (b) volcano plot for NO_3RR of the SACs with a descriptor of ΔG_{NO_3} (the adsorption energy of NO_3^-) and (c) Gibbs energy diagram for HER on Fe SAC and Os SAC. [31] (d-e) SACs supported on a variety of carbon-based

substrates and (e) their theoretical ORR overpotential as a function of ΔG_{OH*}. [32] (f). A mapping of the thermodynamic stabilities and DOS similarities of bimetallic alloy systems. The diagonal represents the most stable structure of pure metals. The components below the diagonal denote the most stable structure of bimetallic alloys and the components above the diagonal imply the formation energy (color) and the DOS similarities (number and graphics) of bimetallic alloys. [33] (g-h). Volcano relationships between $\Delta G_{(*N_2 \rightarrow *NNH)}$ and (g) the d-band center and (h) a new descriptor $\varphi = \frac{d}{\sqrt{E_m}}$ (d: the number of d-electrons of the doped metal; E_m: the electronegativity of the doped metal) in the Au-based single-atom alloy systems for N_2RR. [34] (i). High-throughput computational screening of potential 2D HER electrocatalysts with active basal planes from 2DMatPedia. [35]

First-principles calculations based on density functional theory provide a fundamental and accurate prediction of many materials' physical and chemical properties, which are more resource-efficient than experiments[36]. Similarly, the high-throughput techniques (semi-)automate first-principles calculations to concomitantly assess numerous candidate materials [37-39]. Due to the cost-performance benefit, high-throughput calculations have been widely used to screen/design electrocatalysts and offer experimentalists theoretical insights or guidelines. Over the past decade, SACs, which are well-known for high utilization, catalytic performance and distinct active sites, have been intensively investigated using (high-throughput) calculations. [40-42]. Hui et al. [31] conducted high-throughput calculations to screen potential SACs anchored on nitrogen-doped carbon substrates for nitrate reduction to ammonia (Figure 3a). By considering stability, activity and selectivity, osmium (Os) SAC was predicted as the best SAC candidate towards nitrate reduction (NO_3RR) (Figures 3b and 3c). In addition to the transition metal center, its coordination with neighboring atoms also plays a key role [43, 44]. Using high-throughput screening, Wang, et al. took into consideration both the transition metal center and the coordination environment. [44] Among over 500 systems, the single tungsten (W) atom coordinated with three carbon atoms in a graphene substrate exhibits the most outstanding performance towards N_2RR. Besides, Shui, et al. [32] studied SACs supported on various carbon substrates for ORR. The transition metal single atom (TMSA) embedded in

carbon substrates (X) is here labeled as TMSA-X. The resulting volcano plot (Figures 3d and 3e) indicates that Co-MOF, Ir-Pc, Co-N-C, Co-GDY, and Rh-Pc could be promising candidates.

Alloy systems are another type of appealing electrocatalyst candidates. High-throughput calculations for the alloy system are greatly demanded due to its large search space. Various attempts have been made to reduce the computational burden. One such approach is to use new descriptors which serve as a substitution for ΔG [33, 34, 45]. For instance, Han, et al. [33] introduced a descriptor as shown in equation (1) and (2), ΔDOS_{2-1} to develop a bimetallic electrocatalyst, $Ni_{61}Pt_{39}$, for H_2O_2 synthesis.

$$\Delta DOS_{2-1} = \left\{ \int [DOS_2(E) - DOS_1(E)]^2 g(E;\sigma) dE \right\}^{\frac{1}{2}} \quad (1)$$

$$g(E;\sigma) = \frac{1}{\sigma\sqrt{2\pi}} e^{-\frac{(E-E_f)^2}{2\sigma^2}} \quad (2)$$

This approach was applied to screen a material pool of 4350 structures, leading to the identification of various promising candidates, as illustrated in Figure 3f. Chen, et al. [34] have indentified the volcano relationship between ΔG and *d*-band center (Figure 3g) and φ ($\varphi = \frac{d}{\sqrt{E_m}}$, E_m means electronegativity, and d represents the number of d-electrons in the valence orbital) (Figure 3h) in N_2RR for Au-based single-atom alloys. Leveraging these two descriptors, they successfully predicted that Mo and W/Au (111) could have superior activity and selectivity towards N_2RR. These findings highlight the potential of using such descriptors in high-throughput calculations/screening process, which can be used in alloys and many other material systems to guide the development of highly efficient electrocatalysts for specific reactions.

High-throughput computational investigations based on elemental substitutions or likewise usually concentrate on a few specific categories of candidate materials, as discussed above. On the contrary, materials databases offer an excellent platform to explore a diverse range of potential materials [35, 46, 47]. Alongside experimental databases [48], computational materials databases have recently been established, such as the Materials Project (MP) [49], the Open Quantum Materials Database (OQMD) [50], AFLOW [51], and 2DMatPedia [52].

Although the DFT codes deployed to build these databases may differ from one to another, most codes/methods are able to converge toward the same result with errors comparable to those of experiment [53]. These computational databases provide a wealth of properties that may be challenging to measure in experiments at a large scale and can be leveraged to facilitate high-throughput computational screening of functional materials. As shown in Figure 3i, Yang, et al. [35] conducted a high-throughput screening on 2DMatPedia to search basal-plane-active two-dimensional (2D) HER electrocatalysts. Prior to high-throughput calculations, existing properties in 2DMatPedia, which include ICSD (Inorganic Crystal Structure Database) numbers of parent materials, exfoliation energies and band gap sizes, have been analyzed to prescreen 2D candidate materials. Such prescreening dramatically reduces the number of candidates to 208, for which high-throughput calculations were performed. Based on high-volume Gibbs energy calculations, nine potential 2D HER electrocatalysts with diverse compositions and structures were identified. Analogously, Huang et al. [46] explored the CoRE database and discovered that GAFRUD, CAJQEL and cg400449c are potential Metal-Organic Frameworks (MOFs) electrocatalysts for CO_2RR. From the Materials Cloud (2D materials database) [54], Andreussi, et al. [47] identified CoO_2 and FeS with the lowest overpotentials and excellent aqueous stability at the acidic condition for HER applications.

4. Machine Learning

Although high-throughput experiments or computations can greatly accelerate the discovery of potential electrocatalysts, it is practically unfeasible for them alone to efficiently search for potential candidates from hundreds of thousands of materials or even investigate a single complex material system (*e.g.*, HEAs). Recent reports have shown that integrating high-throughput experiments/computations with machine learning (ML) technologies can make an enormous difference to electrocatalyst explorations. [55-58] A typical workflow of machine learning-enabled electrocatalyst design is illustrated in Figure 4a, which usually consists of data generation using high-throughput experiments or computations, ML model training and test, and deployment of trained ML models. For example, Li, et al. [*55] implemented high-throughput calculations to generate a training dataset of double-atom catalysts (DACs), based on which topological information-based ML models were trained to predict the stability and

OER/ORR electrocatalytic performance. These ML models eventually enabled them to identify 511 DACs for OER, 855 DACs for ORR and 248 bifunctional DACs for both OER and ORR from 16767 candidates. The screening time was found to be 114000 times less than pure high-throughput computational screening. Furthermore, Rossmeisl, et al. [56] combined high-throughput calculations and ML to study the ORR catalytic performance of Ir-Pd-Pt-Rh-Ru HEAs. With 871 *OH and 998 *O DFT adsorption energies as the input, the trained ML models could reach high accuracies with the room-mean-square deviation (RMSD) of 0.063 and 0.076 eV for *OH and *O, respectively (Figure 4b-e). These ML models make it practically achievable to efficiently navigate the complex composition space of HEAs and maximize ORR active local surface structures. It is noteworthy that such combinatorial methods are general and can be applied to HEA-based electrocatalyst exploration for other electrochemical reactions. [59]

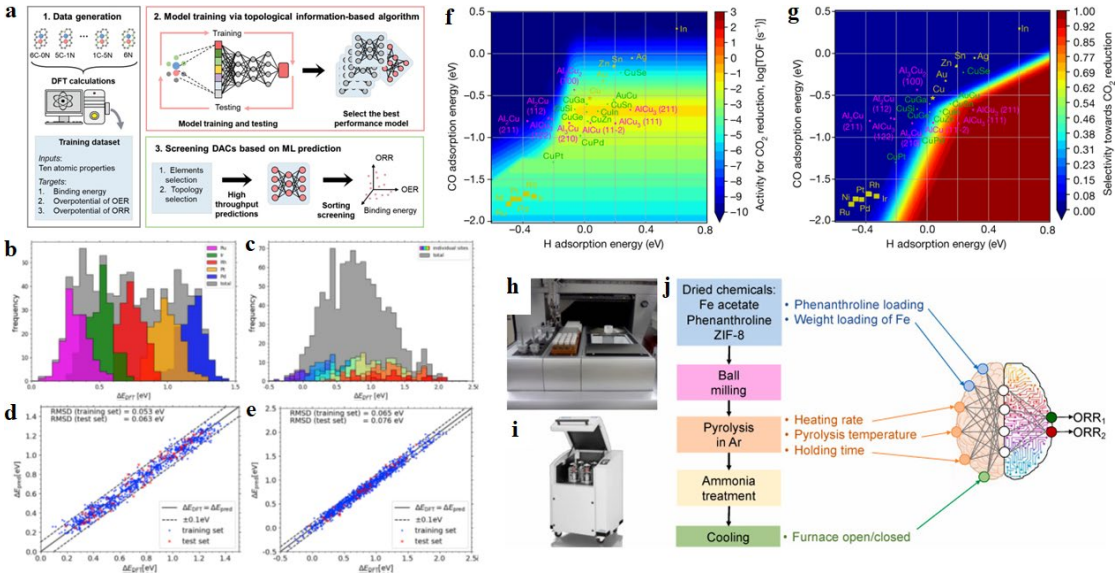

Figure 4. (a). Workflow for screening double-atom catalysts with DFT calculations and machine learning for OER/ORR. [*55] (b-e). Screening for ORR on the surface of Ir-Pd-Pt-Rh-Ru HEA system. (b-c). The distribution of (b) *OH and (c) *O adsorption energies for periodic unit cells. (d-e**).** ML-predicted adsorption energies of (d) *OH and (e) *O plotted against DFT-calculated results. [56] (f-g). A two-dimensional volcano plot for CO_2RR (f) activity and (g) selectivity of Cu and Cu based compounds. [57] (h-j). High-throughput experiments and machine learning on Fe-N-C systems for ORR applications. (h-i). Illustration of high-throughput experiments: (h) robotic synthesis platform and (i**)** high-throughput planetary ball

mill. (j). An illustration of machine learning guided catalyst design. [58]

Recently, Sargent, et al. [57] employed a comprehensive approach that combines high-throughput calculations, machine learning, and experiments to develop the Cu-containing CO_2RR intermetallic electrocatalyst. The research began with high-throughput calculations of CO adsorption energies on various sites. Meanwhile, machine learning models were trained on the fly and directed high-throughput calculations towards the intermetallic compounds with ML-predicted optimal CO adsorption energies (Figure 4f and 4g). This close integration not only significantly increases the efficiency of high-throughput calculations, but also constantly improves the ML models. This leads to the theoretical discovery of the most promising Cu-Al candidate, which has been experimentally verified by the superior CO_2RR performance to ethylene. The successful identification of Cu-Al electrocatalysts well illustrates the value of high-throughput computations and ML in guiding the experimental exploration of diverse and complex systems.

In addition, Wilton, et al. [58] reported an approach for high-throughput synthesis of Fe-N-C electrocatalysts. To achieve this, a robotic synthesis platform (Figure 4h) and a high-throughput planetary ball mill (Figure 4i) were leveraged to efficiently synthesize a large number of Fe-N-C samples. Regression models were developed to surrogate the complex relationships between six input features characterizing catalyst synthesis conditions and their ORR activity in the first two cycles of staircase voltammetry (as shown in Figure 4j). The integration of high-throughput synthesis and data-driven regression modeling enabled to efficiently explore a vast chemical space and single out four ORR electrocatalysts that outperform the best one in the initial database.

5. Conclusions and Perspectives

In conclusion, significant advancements have been made in electrocatalyst design in recent years. High-throughput experiments, which are often complemented by automation or robotics, efficiently prepare and characterize samples, enabling parallel execution and rapid data acquisition. High-throughput calculations provide an efficient process to screen and predict the

properties of a broad range of materials, including stability, catalytic activity, and selectivity. The vast datasets obtained from high-throughput processes serve as invaluable resources for further integrating machine learning technologies. By harnessing machine learning technologies, essential descriptors and patterns can be extracted, giving rise to deep insights into the intricate relationships between intrinsic properties and catalytic performance of electrocatalyst candidates.

As the data-driven approaches are playing a pivotal role in the development of high-performance electrocatalysts, we would like to point out some potential avenues for future investigations.

i. High-throughput experiments make data acquisition more efficient than traditional methods. Central to the high efficiency is the general nature of developed synthesis approaches with respect to a certain type of materials like SACs and HEAs. [15, 16] New general synthesis approaches are demanded to experimentally reach diverse categories of materials in a high-throughput way. On the other hand, the use of robotic platforms could be beneficial.[58, **60]

   The reported high-throughput calculations usually leverage the imperial linear scaling relationships to speed up the screening of superior electrocatalysts towards multiple-electron reactions. Nevertheless, this may make it unlikely for high-throughput calculations to identify potential electrocatalysts whose catalytic performance could surpass that constrained by the linear relationships. Also, low-coverage models are often adopted. Taking into account the coverage effect is crucial to electrocatalysts which are catalytically active at intermediate or high coverages. Besides, it would be important to consider more factors of electrochemical reaction, such as solvent effect and electrolyte/catalysts interface, in the high-throughput first-principles calculation or machine learning, which would give us a more reliable prediction.

ii. While machine learning has demonstrated impressive performance for the electrocatalyst design from the search space of similar materials, its application is far from satisfactory when it comes to large search spaces consisting of diverse materials with various structures, elemental combinations, and composition ratios. This could mainly be ascribed to the extremely sparse sampling of the enormous materials space

that we are able to reach. [61] Therefore, it is highly desirable to develop ML models that can make the best use of the sparse sampling data points. Alternatively, the divide-and-conquer strategies are also worth considering while developing ML models. More specifically, the entire enormous materials space may be divided into a set of subspaces according to similarities between materials such as structural prototypes and elemental compositions. ML models are trained and optimized for each subspace and then may together constitute a compound ML model with improved performance.

iii. Last but not least, the close integration of high-throughput experiments, high-throughput calculations, machine learning and even robotics will make a huge difference to the design of advanced electrocatalysts. [58, 62] It is noteworthy that partial combinations, in particular between high-throughput calculations and machine learning, have been well demonstrated nowadays. [55, 56] However, full integrations have been rarely realized. Close cross-disciplinary collaborations among multiple research groups with their respective expertise in high-throughput experiments, high-throughput calculations, machine learning and robotics are indispensable to achieve our common and ultimate goals of efficiently designing advanced electrocatalysts and combating global warming.

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7. Declaration of interest

The authors declare no competing interests.

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