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AI-guided electro-decomposition of persistent organic pollutants: a long-awaited vision becoming reality?

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Existing and emerging persistent organic pollutants (POPs) constitute great threats to human health and cause great economic loss. The persistence of these chemicals made it necessary to develop proper mitigation strategies to remove or degrade them from the natural environment. Various methods for electrocatalytic degradation of POPs have been developed under lab conditions, while challenges including harmful byproducts, limited working time, *etc.* still limit their practical use. As the core of electrocatalytic degradation is the optimization of electrocatalysts, we outlined why high-entropy alloys could be ideal materials as catalysts in this perspective work, and further discussed how AI could be utilized in facilitating and accelerating the design–characterize–test cycle for electrocatalyst development. The potential challenges, negative effects and solutions of applying AI in this process were also discussed. By leveraging AI's power, it is expected that the vast potential of new catalyst materials including high-entropy alloys could be fully explored in a timely manner, which would contribute to developing a practical electro-degradation strategy for POP mitigation. A broad perspective of AI in designing high-entropy alloy catalysts was also included.

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Environmental significance

Existing and emerging POPs constitute great threats to modern society. It is necessary to develop proper mitigation strategies to remove or degrade them from the environment. Electro-degradation strategies showed promising potential in the mitigation of POPs, given reliable catalysts could be developed. High-entropy alloys could be, ideally, designed to catalyze reactions for each pollutant as they allow continuous yet accurate control of adsorption energy on their reaction surface. However, the time taken to design, characterize, and test catalysts limited the search for appropriate catalysts. In this perspective work, we outlined how AI could be utilized to develop high-entropy alloy based electrocatalysts in a timely manner and the challenges we might face, which shed light on practical POP mitigation strategies.

From both health and economic perspectives, existing and emerging persistent organic pollutants (POPs) greatly threaten human society. For example, per- and poly-fluoroalkyl substances (PFAS), a family of over 10 000 chemicals called "forever chemicals" due to their persistent nature and ability to accumulate in the food chain, were associated with various reproductive and developmental diseases as well as cancer formation. Meanwhile, it was not until 2020 that a single toxic molecule, *N*-(1,3-dimethylbutyl)-*N*′-phenyl-*p*-phenylenediamine quinone (6PPD-Q), was isolated and identified to be the cause of acute mortality for coho salmon associated with stormwater runoff, formerly known as Urban Runoff Mortality Syndrome.[1] Derived from rubber tire antioxidant 6PPD, 6PPD-Q was quickly found to exist ubiquitously in various environments including rainwater, urban watershed, sewage, roadside soil, indoor and outdoor dust, and atmospheric fine particles.[2] This pollutant was also found accumulated in the human body and is highly toxic at trace doses to several fish species of commercial and ecological importance. POPs including 6PPD-Q can resist natural degradation processes due to their inherent stability and resistance to degradation, making their removal difficult. Therefore, measures should be taken to mitigate POPs that remain in environments, to effectively alleviate the health and economic burden those POPs bring to human society.

Efforts have been made to achieve the degradation and removal of POPs under lab conditions *via* an electrocatalytic strategy.[3] Electrocatalysts are essential in determining the selectivity and degradation efficiency of POPs. However, the complex and diverse chemical structures of POPs make it challenging to find a desired electrocatalyst that can effectively break down the pollutant without producing harmful byproducts. Furthermore, the harsh working conditions of electro-

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degradation make it difficult to maintain long-term activity. Therefore, designing a satisfactory catalyst for POP mitigation remains a significant challenge in the field.

At the core of electrocatalyst design lies the "Volcano Curve",[4] which states that moderate adsorption energy of degradation targets is desirable for highly efficient electrocatalytic reaction. According to the theory, it is possible to design appropriate catalysts for each specific target (POPs and their degradation intermediates) according to their adsorption energy to improve the performance and selectivity. High-entropy alloys (HEAs), which allow continuous yet accurate control of adsorption energy on their reaction surface *via* a free combination of their composing metals, consequently become a promising choice as electrocatalysts for POP degradation. However, two major hurdles constitute a great barrier to HEAs' usage in real practice: the difficulty in synthesizing materials and the extremely time-consuming design–characterization–test cycle. In the following paragraphs, we will discuss how the revolutionary power of artificial intelligence (AI) could be utilized in accelerating the HEA design and synthesis for POP electro-degradation.

The great challenge for synthesizing HEA catalysts is to obtain a single phase. According to the Gibbs free energy equation,

$$\Delta G_{\mathrm{mix}} = \Delta H_{\mathrm{mix}} - T\Delta S_{\mathrm{mix}}$$

a more negative ΔG_{mix} is required to obtain a single-phase state with random metal atom distribution. Despite ΔH_{mix} and $T\Delta S_{\mathrm{mix}}$ being determined by the nature of each metal, it is a facile and efficient way to increase the temperature T and decrease ΔG_{mix}. To avoid metal particle growth, agglomeration, and/or phase separation, an ultra-fast annealing and cooling method was proposed to benefit the formation of HEA particles.[5] As experimental datasets of HEA synthesis accumulate in large amount, machine learning algorithms based on neural networks could be utilized to explore a wide range of synthesis conditions of silica and identify the optimal temperature, pressure, and time for alloy formation, as well as predict the likelihood of phase formation under different conditions.

After successful synthesis, HEA electrocatalysts are conventionally characterized by metallographic specimens and X-ray diffraction, followed by electrochemical characterization to obtain the structure–property relationship of the alloys. The performance of the electrocatalysts will then be tested by experiments, and the data obtained could be used as feedback to guide the next round of design synthesis. As such characterization–test–feedback loops have to be done in a one-by-one manner, the overall efficiency is greatly compromised due to the long yet time-consuming evaluation cycle, limiting the practical use of HEAs.

It is therefore long-awaited that AI could be used to predict and evaluate HEAs as electrocatalysts for each targeting POP and their intermediates to accelerate the overall cycle. By inputting data including elemental composition, crystal structure, and synthesis conditions from previous experimental datasets, and leveraging the power of machine learning algorithms such as neural networks, we may expect AI to predict the catalytic activity, stability, or other relevant properties of HEAs. Meanwhile, density functional theory (DFT) simulations, a quantum mechanical method for calculating the electronic structure and properties of materials, could be empowered by AI and used to perform in silica evaluation of HEAs. We might be able to "predict" the proper HEA for every single type of POP and guide the corresponding synthesis strategy of the catalyst purely based on existing datasets. In addition, synergistic effects exist in all alloy catalysts, with no exception for HEAs. Synergistic effects can arise from the electronic interactions between alloy elements, such as charge transfer, electron delocalization, and modification of the electronic band structure. The arrangement of atoms in the alloy can also result in synergistic effects. The presence of different elements can induce structural distortions, alter the surface morphology, create active sites with unique geometries and even impact the stability and durability of the electrocatalysts. The exploration of various alloy compositions and combinations is essential to identify the synergistic effects. As HEAs allow a free combination of their composing metals to achieve continuous yet accurate control of adsorption energy on the reaction surface, the synergistic effects between different components in HEAs will be even harder to interpret and predict. Consequently, an AI-assisted analytic and prediction pipeline will be powerful to understand the synergistic effects in HEAs (Fig. 1).

The functional groups of targeting pollutants are the major consideration factors in designing electro-degradation strategies. According to the "Volcano Curve" theory, moderate adsorption energy of degradation targets is desirable for highly efficient electrocatalytic reaction. If we want to achieve selective electro-degradation for targeted functional groups of organic molecules, it is necessary to achieve specific functional group enrichment and adsorption by controlling the physicochemical properties of the catalyst surface. Different functional groups can exhibit varying affinities for specific pollutants, influencing their adsorption behavior. The presence of specific functional groups can enhance or hinder the adsorption of pollutants, thereby impacting the overall degradation performance. More importantly, it is unavoidable that certain intermediate products will generate during the electro-degradation process *via* oxidation/reduction of certain functional groups of the target pollutants. The intermediate organic molecules may show a strong adsorption on the catalyst surface and poison the active sites by occupying the active sites. Tailoring the surface functional groups with proper design will help degradation or desorption of the undesirable intermediate products or reducing the adsorption rate of the intermediates. In conclusion, the functional groups of pollutants to be enriched and adsorbed on the surface of alloy catalysts have tremendous effects on the selectivity, stability, during, and activity of the electrocatalysts. A thorough understanding is necessary for proper design of the catalyst, which requires a combination of experimental studies, theoretical modelling, and computational simulations. AI-assisted catalyst design will greatly improve and accelerate the process and improve the overall performance of the electrocatalysts.

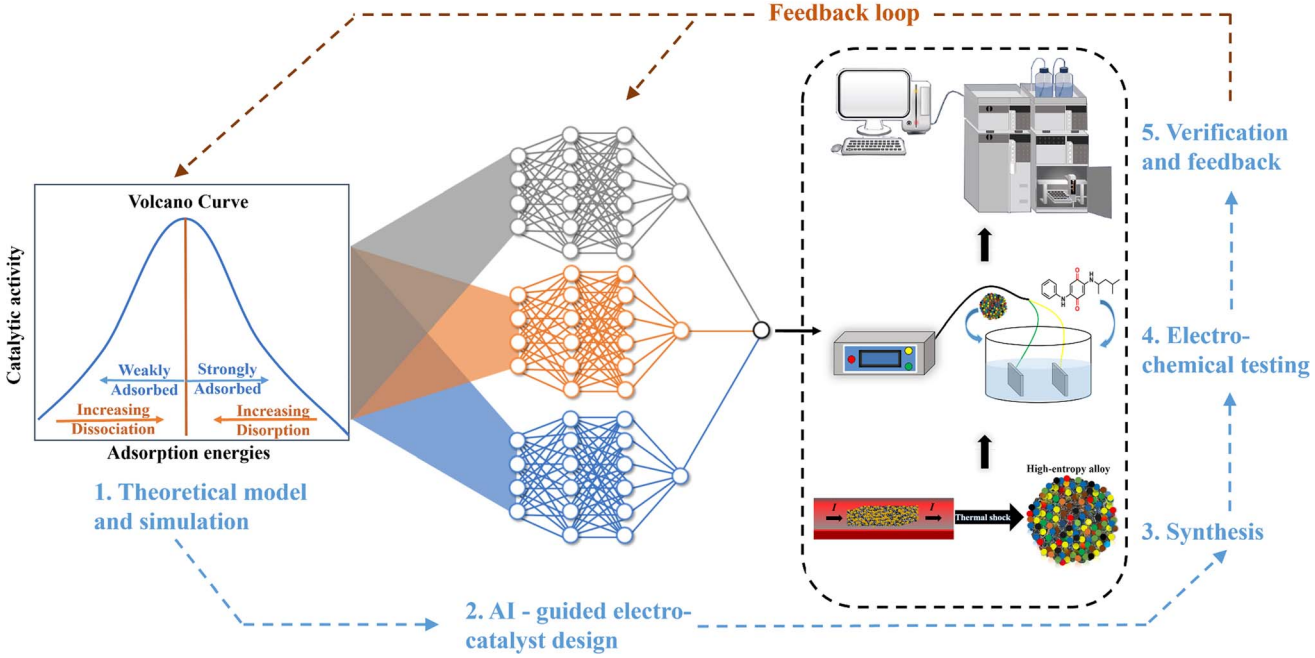

Fig. 1 Proposed AI-guided electrocatalytic decomposition strategy for POPs. AI could accelerate the overall design–synthesis–test cycle by facilitating the theoretical modeling, simulation, design, synthesis, and optimization processes for an optimized electrocatalyst. Given the experimental data of synthesized electrocatalysts, the AI-driven models will be in a continuous iterative upgrade.

Been said that applying AI in HEA-mediated POP degradation is not all smooth sailing. When applying AI-guided electro-decomposition on a larger scale, the first challenge is costs and resources for infrastructure and computational power. Significant investment in hardware, software and skilled personnel is required for implementing AI to robustly process large scale data. Second, the reliability and robustness of AI models need to be thoroughly tested, validated, and monitored constantly. The issue might be more crucial when scaling up the system to the industry level. Third, how to integrate the AI models with existing systems while following proper regulatory compliance might be a significant challenge. Established protocols and systems, particularly in the waste management industry, are bound by governmental regulations and guidelines. How to implement an AI-guided strategy while complying with current regulations may need extra efforts, particularly in solving challenges related to data privacy and safety with legal considerations. Last but not least, it is necessary to develop effective human–machine collaboration between AI systems and human operators, so as to leverage the strengths of both parties for an optimized electro-decomposition process.

It is worth noting that potential negative effects when applying AI in the designing process should be noticed as well. Poor quality data may compromise the accuracy and reliability of AI prediction. More importantly, deep learning models often lack interpretability, which means although the prediction might be correct, it is challenging to understand or explain the results. It therefore may further lead to another negative effect: overreliance on AI. It should be noted that AI should be only used as a complement tool while human expertise still plays a significant role in decision making. Lastly, as training of AI models in catalyst design may require access to sensitive or proprietary data, data security and privacy might be potential issues and should be monitored constantly. Progress in the characterization tools and knowledge of catalysts is still of utmost importance for catalyst design, while data validation and testing are required to guarantee the reliability of AI prediction. Addressing these challenges requires collaboration between AI experts, industry professionals, regulators, and policymakers. It is important to invest in research and development, establish standards and guidelines, and foster partnerships to successfully implement AI-guided electro-decomposition on a large scale and realize its potential benefits for industrial pollution control.

In conclusion, AI can play a pivotal role in designing and optimizing well-defined electrocatalysts. Through leveraging the power of machine learning, AI can be used to explore the vast potential of HEA electrocatalysts, *e.g.*, to identify the most promising optimization pathways, and to predict the performance in different environments. Specifically, as suggested in this perspective, the machine learning-guided HEA electrocatalyst design and thermal shock-based synthesis of electrocatalysts with an appropriate d-band center would be the key for successful and efficient mitigation of POPs in living environments, such as 6PPD-Q and PFAS. More excitingly, with the accumulating data and increasing calculating power, AI is expected to improve, or accelerate the HEA design in almost every aspect, and not to limit its application to degradation of environmental pollutants. For example, as the time for discovery

and optimization processes could be significantly reduced, it is possible to achieve customized catalyst design using HEAs. Due to the delicate control of surface adsorption energy of HEAs, in theory, HEAs could be used to catalyze every possible reaction with higher selectivity, stability, and efficiency. HEAs could, therefore, not only be used to degrade undesirable chemicals (pollutants, waste, or undesirable intermediates), but to promote the formation of desirable chemicals, such as high value-added chemicals, *e.g.*, hydrogen. Further optimization of HEAs would improve the selectivity of desirable product formation with lower catalyst costs, consequently achieving greater economic benefits. AI-guided HEA design strategies therefore could be widely applied in various industrial production scenarios and contribute to a better life of mankind in the future.

Conflicts of interest

There are no conflicts to declare.

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