Material and Composition Screening Approaches in Electrocatalysis and Battery Research

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In light of the global effort to transform the energy supply away from fossil fuels toward renewables, electrochemical devices have emerged as key technologies to store and convert energy as well as to convert waste products such as CO_2 into high-value chemicals. Progress in advancing these technologies (i.e., fuel cells, batteries, electrolyzers, or CO_2 conversion cells) hinges on the development and improvement of key materials, especially electrocatalysts and charge storage materials as well as electrolytes and transport materials. The complexity of the structure and composition of the state-of-the-art materials synthesized in recent years has made the search for new materials challenging. Trial-and-error-based screening of new materials is becoming less effective, as there are too many material combinations, synthesis parameters and processing routes to try experimentally. Hence, effective screening methods to designate experimental targets are required to advance the discovery of next-generation materials in an effective way. The development of advanced screening methods has led scientists on the hunt for the understanding of underlying relationships (e.g., structure-property-performance), and guiding principles (like the Sabatier principle). Additionally, the use of ever-more-powerful computational methods to avoid cumbersome, expensive experiments, as well as the utilization of machine learning and artificial intelligence techniques to re-create in-silico the intuition and experience of an experimenter, leads to a further increase of the efficiency of materials screening methods.

The articles comprised in this special issue highlight a broad range of applications, in which materials and composition screening is used. They range from advanced battery technologies (Dillenz, Kowalski, Tichter), to water splitting (Lim, Mukouyama, Zeradjanin) and CO_2 reduction (Malek, Verma) to general electrocatalysis (Kox, Ooka). The applied methods span the whole spectrum from computational methods like density functional theory (Dillenz, Kowalski, Lim, Verma) and ab-initio molecular dynamics simulations (Kox), continuum modeling (Mukouyama), artificial intelligence (Malek) as well as the interplay of theory and experiment (Tichter, Mukouyama).

Apart from the 8 Original Research articles, this special issue contains 2 review articles. Our editors Ooka, Huang and Exner have reviewed the famous Sabatier principle within the context of electrocatalysis. They have highlighted its limitations and challenges, and in doing so, have showed how moving beyond its current thermodynamic framework might lead to next-generation electrocatalysts. In the second review, Zeradjanin et al. have reviewed the current understanding of activity and stability trends of oxides for the anodic oxygen evolution reaction (OER). Their review brilliantly highlights that the topic is far from being concluded and ends with remaining fundamental questions and eight suggestions for future research directions.

Lim et al. partially followed one of the suggested directions by tuning the OER activity of transition-metal oxides via the strategic formation of a heterostructure with another transition metal oxide. They screened 11 transition metal oxides on a TiO2 substrate using DFT, finding that these heterostructures follow the universal scaling relationship of metal oxides, thereby confirming RuO_2 and IrO_2 as highly active OER catalysts.

Staying in the field of electrochemical water splitting, Mukouyama et al. investigated the hydrogen evolution reaction and highlighted the importance of quantifying the surface pH. They developed and demonstrated an effective continuum model, which converts partial differential equations to ordinary differential equations, allowing the the surface pH to be estimated in a computationally efficient way. The resulting model explains measured experimental voltammograms of both the hydrogen evolution as well as the more complex hydrogen peroxide reduction reaction.

Another work focusing on the catalyst-electrolyte interface is presented by Kox et al. Using ab-initio molecular dynamics simulations, they unraveled the effect of water and solvation on the structure and reactivity of Co_3O_4 (001) A-type and B-type surface terminations.

Moving to the application in batteries, Tichter et al. have investigated the electro-oxidation of VO^{2+} on glassy carbon electrodes, as encountered in redox-flow batteries. They performed stationary and rotating linear sweep voltammetry, which they combined with Koutecký-Levich analysis. While the observed concentration dependence of the ordinate intercept in the Koutecký-Levich plots was so far unexplained by the theory, they introduced a concept of finite rate constants leading to a theory that captures mass transport limitations, Butler-Volmer kinetics, and finite heterogenous kinetics simultaneously.

In solid-state lithium ion batteries, Kowalski et al. gave an overview and discussed the role of atomistic modeling in accurately predicting thermodynamic properties of Li_xFePO_4 orthophosphates as well as fluorite- and pyrochlore-type zirconates, key materials for energy storage and solid-state ion conduction. Dillenz et al. also used periodic density functional theory calculations to screen the migration of various charge carriers in spinel-type $MgSc_2Se_4$, a potential candidate for solid electrolytes in Mg-ion batteries. Screening the diffusion barriers of different ions in this material allowed disentangling structural and chemical factors in ion mobility. Not only the size and charge of the ion determines its mobility, but also charge redistribution and rehybridization caused by the migration of multivalent ions increase the resulting migration barriers.

Tackling the challenge to find suitable electrocatalysts for CO_2 reduction to CO, Verma et al. used computational screening of doped graphene electrodes. After establishing thermodynamically stable electrode materials, the CO_2 reduction reaction in alkaline media was studied. It was found that the CO_2 electrosorption and associated charge transfer along the decoupled proton and electron transfer mechanism significantly impacts the electrochemical performance, leading to their discovery of metal-doped 3 nitrogen-coordinated graphene as highly active electrodes.

Malek et al. made use of computational and experimental materials data in an artificial intelligence-based material recommendation and screening framework. This framework utilizes high-level technical targets, advanced data extraction, and categorization as well as data analytics and property-matching algorithms to recommend the most viable materials and reveal correlations that govern catalyst performance. This framework is demonstrated for certain classes of electrocatalyst materials for low or high temperature CO_2 reduction.

Overall, we thank all authors for their excellent, broad and multi-facetted contributions that highlight the importance and widespread application of diverse materials screening approaches and show up pathways for future energy materials discovery.

1 Conflict of Interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

2 Author Contributions

TK drafted the editorial. All authors discussed, reviewed, and approved the submission of this editorial

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