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Advancing energy materials through high throughput experiments and computation

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Modern megatrends in research such as automation and machine learning have far reached implications for the very way we conduct research. Ultimately these tools augment research and enable greater and faster knowledge generation.

Withing this themed collection titled "Advancing materials through high-throughput experiments and computation" in *Journal of Materials Chemistry A* we sought to highlight the transformative potential of integrating high-throughput experimentation in conjunction with artificial intelligence (AI), and machine learning (ML). This is to ultimately cross-fertilize automated, and data driven virtual and physical experiments. All of this is, to accelerate the discovery and proliferation of energy materials – which we dearly need in the wake of climate change and greenhouse gas emission mitigation. The submitted and accepted papers presented herein underscore the need for joint physical and virtual experiments to efficiently explore the vast chemical space to tackle what is possibly humanity's greatest challenge.

Papers range from the diverse fields of catalysis for water splitting or ammonia production over batteries, all the way to metamaterials. Herein we briefly summarize the awesome content of all manuscripts featured in this collection.

Cai *et al.* (**https://doi.org/10.1039/D3TA06211B**) employ an automated high-throughput aerogel synthesis system to rapidly generate and screen a series of $Fe_xCo_yLa_z$-based electrocatalysts for the oxygen evolution reaction, demonstrating the efficiency of combinatorial approaches in identifying high-performance compositions. The paper by Zhou *et al.* (**https://doi.org/10.1039/D3TA04899C**) explores the development of non-precious metal oxide catalysts for the acidic oxygen evolution reaction (OER), a critical process in green hydrogen production. The authors utilize a combinatorial approach to synthesize and characterize complex rutile-phase electrocatalysts in the Mn–Sb–Sn–Ti–O system, aiming to achieve high OER activity at reduced Mn concentrations for improved stability. The study highlights the potential of combinatorial approaches and strategic elemental combinations in the development of efficient and stable non-precious metal oxide catalysts for acidic OER. Jayarathna *et al.* (**https://doi.org/10.1039/D3TA05939A**) demonstrate the use of an active learning framework for the accelerated discovery of ammonia synthesis catalysts, highlighting the potential of AI in guiding experimental design and optimizing catalyst formulations.

The collection also delves into the integration of computational modeling and machine learning with high-throughput experimentation for enhanced materials discovery and characterization. Li *et al.* (**https://doi.org/10.1039/D4TA00982G**) employ high-throughput DFT calculations to generate a comprehensive dataset of alloy structures, enabling the investigation of material representations and the development of effective machine learning models for high-entropy materials. Miyagawa *et al.* (**https://doi.org/10.1039/D4TA00452C**) explore the potential of machine-learning force fields for predicting ion migration mechanisms in solid-state ion conductors, offering a computationally efficient approach for understanding and designing novel materials for energy storage.

The paper by Ojih *et al.* (**https://doi.org/10.1039/D3TA06190F**) explores the use of graph theory and graph neural networks (GNNs) to accelerate the discovery of new stable crystal structures with desirable properties for energy conversion and storage applications. The study demonstrates the potential of combining graph theory with pre-trained GNNs to expedite the search for novel materials with targeted properties.

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Jenewein *et al.* (https://doi.org/10.1039/D3TA06651G) showcase the power of multiobjective Bayesian optimization in guiding experimental design and optimizing catalyst formulations for improved activity and stability. The integration of *operando* mass spectrometry in their study further enhances the understanding of catalyst degradation behavior. Schaefer *et al.* (https://doi.org/10.1039/D3TA07220G) introduce a novel cyclical growth and *in situ* etch method in molecular beam epitaxy, enabling rapid screening of synthesis conditions for $(In_xGa_{1-x})_2O_3$ alloys and highlighting the potential of this approach in accelerating materials discovery and optimization. Farithkhan *et al.* (https://doi.org/10.1039/D4TA01919A) explore the impact of electron and microstructural engineering on the performance of trifunctional electrocatalysts for enzyme-free urea sensing and urea electrolysis applications. The authors systematically varied the composition and morphology of the electrocatalytic materials, allowing them to investigate a wide range of design parameters and their influence on the performance. This approach enabled the identification of the optimal combination of materials and structures for achieving the desired electrocatalytic properties. Yan *et al.* (https://doi.org/10.1039/D3TA06249J) further demonstrate the effectiveness of high-throughput experimentation by developing an automated platform for electrolyte formulation and conductivity measurements, facilitating the rapid screening and optimization of electrolyte compositions for improved battery performance.

Manuscripts in the collection also emphasize the critical role of data management and analysis in materials research. Röttcher *et al.* (https://doi.org/10.1039/D3TA06247C) present a comprehensive data management tool that streamlines data acquisition, analysis, visualization, and publication, promoting FAIR data principles and enhancing knowledge production. This toolset may serve as the catalyst for machine-learning studies downstream. That data management is critical for research and that it can act as a catalyst for more is also shown in the manuscript by Kraus *et al.* (https://doi.org/10.1039/D3TA06889G). Their paper focuses on bridging the gap between trust and control in research data management by integrating experimental automation with computational workflow management systems. The authors successfully interface off-the-shelf battery cycling hardware with the AiiDA workflow management software, enabling the control of experiments while ensuring data integrity through provenance tracking. The paper also details the development of user-friendly interfaces that facilitate experiment design, data visualization, and result analysis. The integration of job monitoring capabilities allows for automated supervision and early termination of experiments based on predefined criteria, leading to significant time and resource savings. Additionally, the authors demonstrate the ability to incorporate data from external sources into AiiDA, promoting a seamless transition from manual to automated labs. They conclude by highlighting the flexibility and adaptability of their approach, suggesting its potential application in various experimental workflows beyond battery research. Soleymanibrojeni *et al.* (https://doi.org/10.1039/D3TA06054C) present an active learning workflow coupled with a kinetic Monte Carlo (kMC) model to investigate the formation of the solid-electrolyte interphase (SEI) in Li-ion batteries. The workflow iteratively trains a Gaussian process classification model to predict the SEI composition based on reaction barriers, enabling efficient exploration of the vast parameter space and identification of key reactions controlling SEI formation. Lu *et al.* (https://doi.org/10.1039/D3TA06227A) employ high-throughput DFT calculations and leverage a database to screen for potential coating materials for $LiFePO_4$-based all-solid-state batteries. The authors evaluate the thermodynamic stability, electrochemical stability window, chemical reactivity, and ionic conductance of various lithium-containing compounds, ultimately identifying 41 promising coating materials. The study also provides insights into the interfacial compatibility between different chemical compositions, offering guidance for coating selection in various battery systems.

Ochoa *et al.* (https://doi.org/10.1039/D3TA05860C) introduce a novel high-throughput screening approach that integrates data-driven methods with physics-based models, enabling the efficient exploration of vast chemical spaces and accelerating the discovery of organic photovoltaic materials. Wieczorek *et al.* (https://doi.org/10.1039/D3TA07274F) present a combinatorial inert-gas workflow for high-throughput aging studies of hybrid perovskite thin films, showcasing the importance of controlled stability assessments in understanding material degradation behavior. Oliva *et al.* (https://doi.org/10.1039/D3TA03119E) utilize reinforcement learning to design shape-changing metamaterials, showcasing the potential of AI in optimizing material architectures for improved energy storage and other functionalities. The authors develop a reinforcement learning model that guides the placement of silicon beams in a virtual environment, aiming to maximize the theoretical storage capacity of the resulting battery electrode. The model's predictions are then experimentally validated using 3D-printed polymer structures, demonstrating the effectiveness of the approach in designing functional metamaterials. Miyazaki *et al.* (https://doi.org/10.1039/D3TA06719J) investigate the gelation conditions for zeolitic imidazolate framework (ZIF) gel synthesis using high-throughput screening and batch synthesis. The authors identify novel ZIF gels and demonstrate their potential for forming dense and mechanically rigid films on various substrates. The study also employs statistical modeling to evaluate the influence of synthesis conditions on gelation, contributing to the development of a unified strategy for MOF gel fabrication. Gu *et al.* (https://doi.org/10.1039/D3TA06892G) construct a comprehensive dataset of gold (Au) nanoclusters and employ a graph convolutional neural network (GCNN) model to predict their formation energies and assess their stability. The model's applicability is demonstrated through the successful synthesis of novel Au nanocluster structures,

highlighting the potential of deep learning in guiding the discovery and synthesis of stable nanoclusters.

This themed collection exemplifies the transformative power of high-throughput experimentation, AI, and ML in accelerating materials research. There is however still a need to move beyond the single lab and isolated setup and build modularly interconnected systems towards truly internationally distributed materials acceleration platforms.[1,2]

References

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