SOLID-OXIDE FUEL CELLS

Catalyst design with machine learning

Development of oxygen reduction catalysts is of key importance to a range of energy technologies; however, the process has long relied on slow trial-and-error approaches. Now, accelerated discovery of perovskite oxides for use as air electrodes in solid-oxide fuel cells is achieved with machine learning.

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A solid-oxide fuel cell (SOFC) is an electrochemical device that converts the chemical energy of a fuel (for example, hydrogen or syngas) into electricity and heat. SOFCs are both highly efficient and fuel-flexible[1]. They consist of an ion-conducting ceramic electrolyte stacked between a fuel-oxidizing anode and an air cathode. At the cathode, oxygen (O_2) in ambient air is reduced to oxide ions (O^{2-}), which travel across the electrolyte to the anode side where they react with fuel molecules, liberating electrons to the external circuit (Fig. 1).

Oxygen reduction at metal oxide cathodes is a kinetic bottleneck that often limits the process to high temperatures (800–1,000 °C); lowering the operating temperature to 500–800 °C through materials design is essential to improve the prospects of SOFC technologies. However, due to the intrinsic complexity of oxide materials with entangled structure, composition, and phase attributes, finding high-performance electrocatalysts of this type, preferably composed of earth-abundant elements, is a long, expensive, and tedious trial-and-error endeavour. Now, writing in *Nature Energy*, Shuo Zhai and colleagues in China report an experimentally validated machine learning approach to accelerate the discovery of perovskite oxides for oxygen reduction in solid-oxide fuel cells[2].

To begin, the researchers curated a small dataset of perovskite oxides from the literature on which to train machine learning algorithms to learn underlying composition–activity correlations, as has been attempted previously[3,4] albeit with different data sources. For each material they collated the polarization resistance — an activity metric — and various features relating to the metal ions in the perovskites, including: electronegativity, ionic radius, Lewis acidic strength, ionization energy, and tolerance factor (a predictor for the stability of perovskite structures). A wide range of machine learning algorithms were then employed to determine which showed

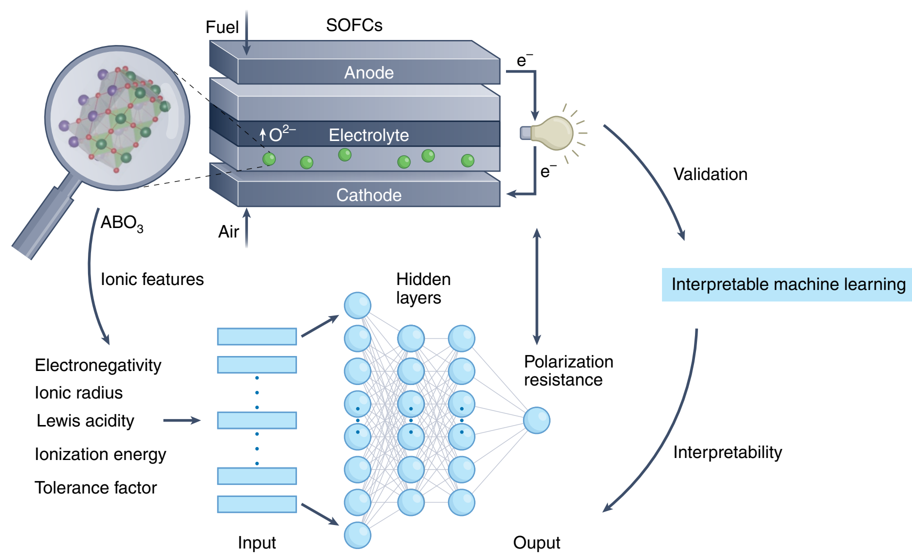

Fig. 1 | Interpretable machine learning for discovery of catalytic materials for solid-oxide fuel cells. Feature engineering of perovskite oxides leads to accurate machine learning models that can predict new materials while providing interpretation or design rules, a key component of explainable AI technologies in an integrated workflow for materials discovery.

the best generalizability in predicting the polarization resistance of materials. Both linear regression and highly non-linear regression (for example, artificial neural networks with multiple hidden layers between input and output) methods were tried, with the latter showing the best results.

In previous works, a variety of reactivity descriptors for perovskite oxides have been proposed to guide the design of new materials[5–7]. However, some of those features require time-consuming quantum-chemical simulations — the *p*-band centre of bulk oxygen atoms, for example — limiting their practical applications in large-scaling materials screening. One of the strengths of the work by Zhai et al. is that the descriptors used are easily accessible and thus a much larger materials space can be explored quickly.

By ranking the importance of features within the artificial neural network models, the researchers found that the Lewis acid strength of metal ions is crucial in determining the polarization resistance. Perovskite oxides typically contain two or more types of metal ion and have the general chemical formula ABO_3. A-site cations are often alkaline-earth and rare-earth metals; B-site cations are transition-metals of variable valence essential for modulating electron transfer during electrochemical reactions. From the researchers' analysis, a simple design rule emerged: promising oxygen reduction electrocatalysts should have a smaller A-site Lewis acidic strength and, to a certain extent, a larger B-site value. In other words, a polarization of the ionic Lewis acidity is needed across the A- and B-sites.

With this insight, the team suggests candidate materials by doping A- and B-sites with ions of choice that have a relatively low or high Lewis acidity — such as Cs^+ for A sites — while satisfying the charge neutrality constraint with fractional compositions.

Among those predicted to be promising, they successfully synthesize four of them (including $Sr_{0.9}Cs_{0.1}Co_{0.9}Nb_{0.1}O_3$) and find them to be more active than the benchmark material, $Ba_{0.5}Sr_{0.5}Co_{0.8}Fe_{0.2}O_{3-\delta}$. Density functional theory calculations suggest that the polarization of Lewis acidity across sites shifts the adsorption of electron-pair species toward B-site cations. This provides a unique site for facile formation of oxygen vacancies and labile oxygen species, which are essential for surface oxygen exchange and their transport at reduced temperatures.

The work from Zhai et al. highlights the importance of interpretable machine learning — machine learning models for which we can understand how final decisions were made — to accelerate discovery of catalytic materials. Broadly speaking, machine learning models can be made interpretable using three different approaches, including feature engineering, algorithm development, and post hoc analysis[8]. Zhai et al. showcase an example of the former: they engineer easily-accessible features that are rooted in the mechanistic understanding of physical processes.

Future development of interpretable machine learning is needed on the path towards explainable artificial intelligence (AI) technologies that humans can comprehend and trust. Open challenges include automatically extracting high-level feature representations of diverse materials with physical constraints, integrating established theories into learning algorithms that have seen promise for metal catalysis[9], and gaining actionable insights by analysing feature importance of machine learning models while being mindful about possible feature correlations.

In order to realize AI-automated design of catalytic materials with increasing complexity (for example: strain- and/or ligand-modified systems; heterostructures; and high-entropy materials) a large amount of high-quality data from high-throughput experiments and computational modelling are likely a prerequisite. Encouragingly, many data initiatives are emerging, such as the open catalyst project[10]. More importantly, deep fundamental understanding of the electronic properties of materials beyond the nearly free-electron approximation (in which electrons move almost freely through the crystal lattice of a solid) is a long-lasting challenge in oxide catalysis; this might hold the key to unlocking the full potential of large-scale materials data and shedding light on design rules for improved catalysts.

Notably, the materials space for perovskite oxides is immense: not only the composition, but also the phase, defect, and symmetry of structural motifs influence catalytic activity[1] and thus it is imperative to leverage tuning knobs beyond simplified ionic descriptors. Nevertheless, the work from Zhai et al. lays important groundwork to tackle materials design challenges with machine learning, and it will be fascinating to see how the computational and data sciences evolve over the coming years in accelerating the discovery of materials with increasing complexity. ❐

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Competing interests

The author declares no competing interests.