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Perspective

High Entropy Oxides: Next-Generation Air Electrodes for Reversible Protonic Solid Oxide Cells

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ABSTRACT: Reversible protonic solid oxide cell (P-SOC) operating at intermediate-temperature exhibits excellent potential as a power generation and green hydrogen production device in fuel cell and electrolysis cell modes because of the high conversion efficiency. However, the lack of efficient air electrodes is the main challenge to obtain P-SOC with remarkable performance. Typically, air electrodes should possess high proton, oxygen ion and electron conductivity, outstanding catalytic ability for oxygen reduction reaction and H_2O splitting, and also long-term durability. Recently, high entropy oxides (HEO) have become popular due to their various potential applications in terms of outstanding properties, including catalysis ability, conductivity, thermal stability, etc. HEO air electrodes have been confirmed to show good electrochemical performance in P-SOC, but the complex compositions and structure make it difficult to study HEO by traditional experimental methods. Machine learning (ML) has been regarded as a powerful tool in materials research and can solve the drawbacks in the discovery of HEO in a traditional way. In this perspective, we not only discuss the current utilization of HEO in P-SOC but also provide a possible process to use ML to guide the development of HEO.

Keywords: High entropy oxide; Air electrode; Proton conduction; Protonic solid oxide cell; Machine learning

Sustainable development depends on low-carbon fossil energy, the scale of zero-carbon energy, and the accelerated integration of multiple energy sources. Hydrogen energy has been considered as a promising energy carrier due to the advantages of clean, low volumetric energy density and high gravimetric density [1]. So, Efficient technologies of green hydrogen production and utilization are pursued to accelerate the innovation of energy. The combination of intermittent renewable energies and H_2O electrolysis strategies is one of the most important methods to achieve the supply of green hydrogen. Several types of electrolysis cells are studied in different operating temperatures based on the employed ionic conductors, such as alkaline water electrolysis cell (AWE) at < 100 ℃, proton exchange membrane electrolysis cell (PEMEC) at 80–150 ℃, protonic solid oxide electrolysis cell (P-SOEC) at 450–700 ℃ and oxide-ion solid oxide electrolysis cell (O-SOEC) at >700 ℃ [2–5]. Hydrogen generation in full-temperature can be realized via collaboratively working of the above cells. However, P-SOEC is still not as mature as other types of cells, even though it offers the advantages of not requiring noble catalysts and having high efficiency. At the same time, the reversible process of P-SOEC can be directly used as a fuel cell mode (P-SOFC) to achieve hydrogen-induced power generation. A typical protonic solid oxide cell (P-SOC) structure consists of a dense proton conducting electrolyte thin film of about several to dozen microns, a porous air electrode, and a porous fuel electrode of composite with nickel and electrolyte. Among these components, $BaZr_{1-x-y}Ce_xM_yO_{3-\delta}$ (BZCM, M = Y, Yb, etc.) is usually used as electrolyte materials [6,7]. Therefore, One of the main challenges for P-SOC is the lack of materials with high catalytic ability for air electrode reactions and excellent tolerance to high H_2O partial pressure, resulting in low utilization and poor stability of air electrodes [8,9]. Thus, most advances in P-SOC focused on air electrodes, and it's urgent to rationally design electro-catalytically active and robust air electrodes of P-SOC for hydrogen production and power generation at intermediate temperatures.

At an early stage, some O^{2-}/e^- conducting oxide from high-temperature oxide-ion solid oxide cells (O-SOC) are applied as air electrode materials for P-SOC, such as $Sm_{0.5}Sr_{0.5}CoO_{3-\delta}$ (SSC), $(LaSr)CoO_{3-\delta}$ (LSC),

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$La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-\delta}$ (LSCF) [10–13]. However, the air electrode reaction is limited to gas-air electrode-electrolyte triple phase boundary (TPB) because these oxides don't possess proton conductivity. To match the proton conductivity of the BZCM electrolyte, $H^+/O^{2-}/e^-$ single phase conducting oxides are proposed to extend the reaction area to the whole surface of the air electrode particle. Over the past twenty years, many advances have been made to increase the hydration ability (H^+ conduction), active reaction sites, electrocatalytic ability, and the thermal compatibility between air electrode and electrolyte, such as some excellent $H^+/O^{2-}/e^-$ conducting materials: (Pr, La, Sr)(Co, Fe, Mn, Ni)O_3, Ba(Fe, Zr, Zn)O_3, and (Pr)(Ba, Sr)(Co, Fe)O_6 [14–16]. These oxides have significantly increased both power density and current density in fuel cell and electrolysis mode. However, such oxides with Sr, Ba, and Pr elements are easy to segregate and agglomerate under high steam concentration, which will change the structure and composition of the oxide surface and affect the durability, active sites, and catalytic ability of the air electrode.

Recently, a novel class of air electrode material, high entropy oxides (HEO), has attracted significant attention due to the distribution of various cations in HEO, which could contribute to a broad range of high electrocatalytic performances. Entropy represents the degree of disorder in a material, which was first introduced by T. Clausius in 1854. Benefit from the research of high entropy alloys (HEA) in recent years, the HEO containing five or more elements with equal or nearly equal atomic fractions are also studied in terms of excellent electrical conductivity, thermal conductivity, magnetic properties, catalytic properties, etc [17,18]. HEO with a high configuration entropy would lead to a decreased Gibbs free energy, thus enhancing the stability of oxides, which is required by the air electrode of P-SOC. In addition to the thermal stability, the multi-elements can also accelerate the formation of oxygen defects, offering catalytic activity for air electrode reactions. Figure 1 shows the publications and citations for HEO and the application of HEO in P-SOC from 2013 to 2023. It is apparent that the studies on HEO were relatively few before 2015, indicating that HEO did not receive much attention.In 2015, Rost et al. highlighted entropy-stabilized oxides, which showed a random and homogeneous distribution of cations [18]. After this, the SCI papers about HEO increased greatly and had a number of 1329 in 2023, suggesting that HEO was getting more and more attention. However, among them, the SCI papers with a topic of P-SOC is only 9, indicating that the utilization of HEO as an air electrode or electrolyte for P-SOC is still in the very initial stage. In addition, the impact of configuration entropy on the protonic conductivity, electrochemical performance, structural durability, etc., is still unclear, and more effort needs to be done to accelerate the development of HEO in P-SOC.

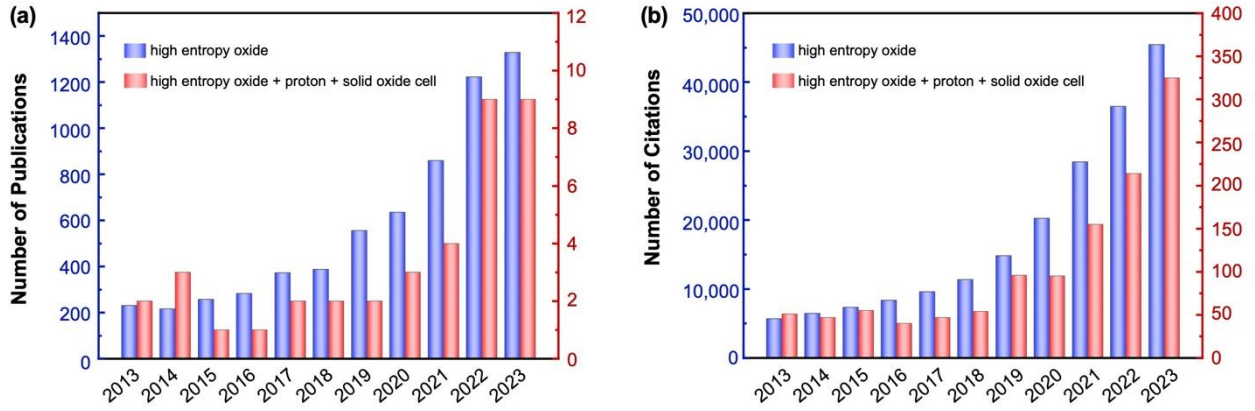

Figure 1. The number of (**a**) published SCI papers and (**b**) corresponding citations related to HEO and P-SOC with HEO from 2013 to 2023 (data come from Web of Science).

The crystal structures of HEO are closely related to their properties. The main crystal structures of HEO include rock-salt, fluorite, perovskite, spinel, and pyrochlore. After exploration for several years, the relationships between the composition, crystal structure, preparation methods, and properties have been established preliminarily. Several review papers have thoroughly discussed HEOs for O-SOC applications, covering synthesis methods, electrical and thermal properties, and electrochemical performance [19–22]. Most commonly used synthesis methods of HEOs in O-SOC are solid-state reaction and sol-gel method, as summarized by Wang et al. [21]. For example, homogeneous single solid-solution phases, $Sr(Zr_{0.2}Sn_{0.2}Ti_{0.2}Hf_{0.2}Mn_{0.2})O_3$, $Sr(Zr_{0.2}Sn_{0.2}Ti_{0.2}Hf_{0.2}Nb_{0.2})O_3$, and $(Sr_{0.5}Ba_{0.5})(Zr_{0.2}Sn_{0.2}Ti_{0.2}Hf_{0.2}Nb_{0.2})O_3$ were successfully fabricated at 1500 ℃ by solid-state reactions [23]. $La_{1-x}Sr_x$(Co, Cr, Fe, Mn, Ni)$O_{3-\delta}$ (x = 0, 0.1, 0.2, 0.3, 0.4 and 0.5) perovskite-type oxides were successfully prepared at 700 ℃ by sol-gel

method [24]. The impedance spectroscopy measurement identified that a total polarization resistance of 0.126 Ω cm^{-2} at 900 °C was achieved for $La_{0.7}Sr_{0.3}$(Co, Cr, Fe, Mn, Ni)$O_{3-\delta}$ air electrode, leading to a power density of 550 mW cm^{-2}.

When HEO is used for P-SOC, HEO-type air electrodes are usually derived from ABO_3 perovskites, as shown in Figure 2a. The A-site is randomly and homogenously occupied by different cations. Most HEO air electrodes are also synthesized via the sol-gel method. For example, by this way, $Pr_{0.2}Ba_{0.2}Sr_{0.2}La_{0.2}Ca_{0.2}CoO_{3-\delta}$ (PBSLCC) was designed by using typical cations (Pr^{2+}/Pr^{3+}, Ba^{2+}, Sr^{2+}, La^{3+}, Ca^{2+}) on A site, which demonstrated similar valence and ionic properties. When used as an air electrode of P-SOC, PBLCC exhibited outstanding activity for oxygen reduction and H_2O splitting reactions with a low polarization resistance of 0.26 Ω cm^2 at 600 °C of BZCM-supported symmetrical cell, contributing peak power density of 1.16 W cm^{-2} in fuel cell mode and current density of 1.75 A cm^{-2} at 1.3 V in electrolysis mode at 600 °C. Specially, the cell maintained a promising durability of 543 h at 1.0 A cm^{-2} at 600 °C even in the air with a higher concentration of steam (30%), which didn't show apparent segregations of nanoparticles or air electrode delamination, implying the high stability of PBSLCC [25]. Similar, Nd was added based on PBSLCC and formed $Pr_{1/6}Ba_{1/6}Sr_{1/6}La_{1/6}Ca_{1/6}Nd_{1/6}CoO_{3-\delta}$ (PBSLCNC), which achieved higher peak power density and current density of 1.21 W cm^{-2} and 1.95 A cm^{-2} under the same conditions, and also the cell kept stable operation without obvious decay after 200 h test under different steam pressure conditions [26]. In the case of the B site, $BaCo_{0.2}Fe_{0.2}Zr_{0.2}Sn_{0.2}Pr_{0.2}O_{3-\delta}$ (BCFZSP) was prepared and employed as an air electrode from the famous oxide $BaCo_{0.4}Fe_{0.4}Zr_{0.1}Y_{0.1}O_{3-\delta}$ (BCFZY). BCFZSP shows integrated $H^+/O^{2-}/e^-$ conduction and yields peak power density and current density of 0.68 W cm^{-2} and 0.92 A cm^{-2} (1.3 V) at 600 °C [27]. Although papers studying HEO as an air electrode are rare, the reported electrochemical performances are superior to most O^{2-}/e^- and $H^+/O^{2-}/e^-$ conducting oxides with low or medium entropy, as summarized in Figure 2b. These results indicate that HEO provides a new strategy for designing air electrodes with outstanding catalytic activity and thermal stability for revisable P-SOC. In addition to air electrodes, HEO was also used as electrolyte, Gazda et al. investigated the possible proton conduction in $BaZr_{0.2}Sn_{0.2}Ti_{0.2}Hf_{0.2}Ce_{0.2}O_{3-\delta}$, $BaZr_{0.2}Sn_{0.2}Ti_{0.2}Hf_{0.2}Y_{0.2}O_{3-\delta}$, $BaZr_{1/7}Sn_{1/7}Ti_{1/7}Hf_{1/7}Ce_{1/7}Nb_{1/7}Y_{1/7}O_{3-\delta}$, and $BaZr_{0.15}Sn_{0.15}Ti_{0.15}Hf_{0.15}Ce_{0.15}Nb_{0.15}Y_{0.10}O_{3-\delta}$ materials [28]. These successful cases of HEO applications in P-SOC indicate that further optimization of the composition and microstructure of HEO will allow higher properties. Nevertheless, the design of HEO, especially the choice of cations at different sites, largely relies on reported air electrode oxides. At the same time, it will take a long time and high cost for repeat experiments to screen HEO and investigate the interaction of each metal cation because of thousands of possible compositions.

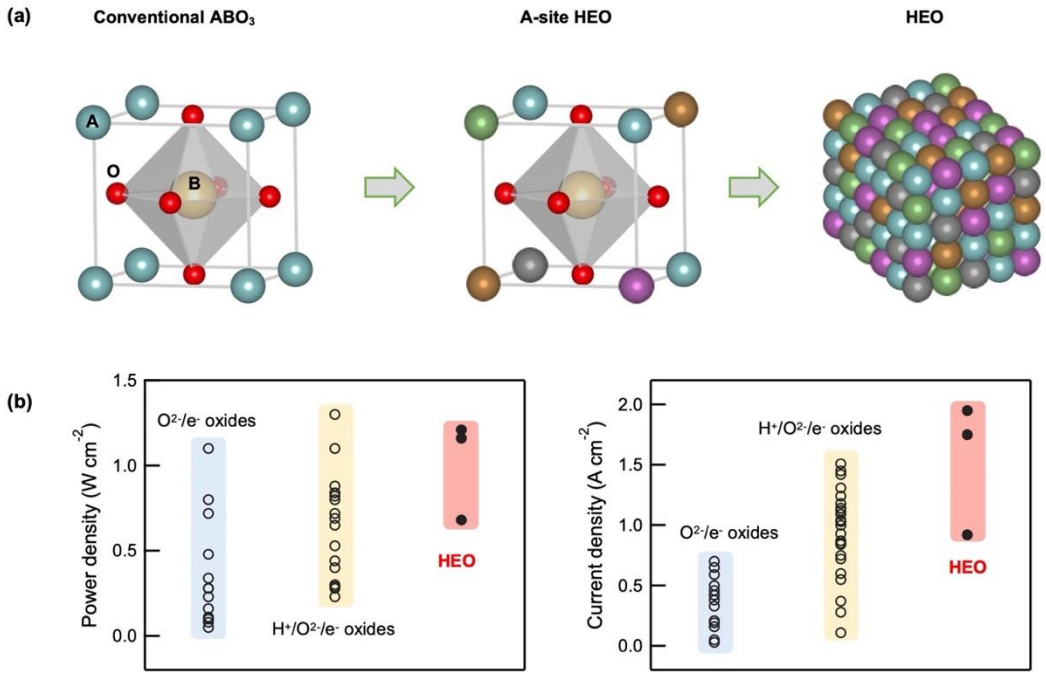

Figure 2. (**a**) Structure of HEO derived from ABO_3 perovskite; (**b**) Summary of peak power density at 600 °C and current density at 600 °C under 1.3 V of P-SOCs with different types of air electrodes.

Material properties are closely related to chemical compositions, crystal structures, physical properties of elements, and other multi-dimensional data. It is expected that material properties can be reliably predicted, thus providing theoretical assistance and guidance for the design of materials. Recently, material science, catalysis, etc., have been

revolutionized by machine learning (ML) models due to the development of artificial intelligence. ML offers advantages in accurate prediction from large amounts of data, low cost by automating processes, handling complex data sets, and so on. According to the specific problems, data features, algorithm performance, and application scenarios, there are some typical supervised learning algorithms commonly used in ML, such as (1) decision trees (DT) display decision rules and classification results in a tree-like data structure; (2) random forest (RF) combines the output of multiple DT to reach a single result and can be used to handle both classification and regression problems; (3) support vector machines (SVM) can be used for liner or nonlinear classification, regression, and even outlier detection tasks; (4) K-Nearest neighbors (KNN) uses proximity to make classifications or prediction about the grouping of an individual data point; (5) neural networks is a model making decisions in a manner similar to the human brain [29,30]. In the case of P-SOC and oxide-ion solid oxide cell (O-SOC), several works have successfully predicted air electrodes in terms of hydrated proton concentration, oxygen reduction activity, and thermal expansion coefficient by using ML [31–34]. For instance, Zhai and his coauthor demonstrated an experimentally validated ML-driven approach to the discovery of outstanding air electrodes, which indicated that ionic Lewis acid strength (ISA) was critical to enhance the surface exchange kinetics of oxides. In this way, $Sr_{0.9}Cs_{0.1}Co_{0.9}Nb_{0.1}O_3$ (SCCN) was selected from 6871 distinct compositions and achieved an extremely low area-specific resistance (ASR) of 0.01 Ω cm^2 at 700 ℃, contributing to peak power density of 1.52 W cm^{-2} at 600 ℃ [31]. However, the available papers in P-SOC with the utilization of ML to screen new materials or forecast the properties are still scarce.

As mentioned above, the complex structure and composition of HEO cause challenges in studying its various possible compositions by using traditional experimental and computational methods, including density functional theory (DFT). Fortunately, the maturing ML techniques make it possible to explore HEO air electrodes based on experiment data from low or medium-entropy oxides, structure data from DFT like the open quantum materials database (OQMD), materials projects, and so on. Here, the OQMD is a database of DFT-calculated thermodynamic and structural properties of materials, which offers the formation energy, decomposition energy, and stability of materials [35,36]. Materials project also provides open web-based access to computed information on known and predicted materials, which includes density, magnetic properties, electronic structure, vibrational properties, etc [37]. Moreover, ML can not only build HEO compound systems from different cations but also can directly make a prediction in terms of HEO properties, such as, formation energy, oxygen vacancy concentration, proton conductivity, etc. by training reasonable and accurate ML models. Recently, many works combining high entropy compounds with ML were reported [38–45]. For instance, Divilov et al. developed a convolutional algorithm and introduced a disordered enthalpy-entropy descriptor to represent the balance between entropy gains and enthalpy costs, enabling the discovery of novel high entropy ceramics [38]. Mints et al. developed an experimentally obtained dataset of 350 nanoparticles to find the optimal catalyst AuIrOsPdRu for oxygen evolution reaction (OER) [41]. In the case of HEO, Seyedsaeed et al. made four robust ML models, including adaptive boosting (AdaBoost), categorical boosting (CatBoost), RF, and eXtreme Gradient Boosting (XGBoost) to predict the selectivity of the oxidation reactions. The results indicated that the XGBoost model exhibited the highest accuracy and the noble metal-free $(CoFeMnCuNiCr)_3O_4$ HEO nanoparticles grafting on reduced graphene oxide displayed excellent catalytic performance for aerobic and solvent-free oxidation of benzyl alcohol [42]. An integrated explainable ML algorithm with high throughput first principles and experimental verification was used to screen low thermal conductivity $A_2B_2O_7$-type HEO. In this way, $(Sc_{0.2}Y_{0.2}La_{0.2}Ce_{0.2}Pr_{0.2})_2Zr_2O_7$ was selected from 6188 $(5RE_{0.2})_2Zr_2O_7$ HEO, which performed low experimental thermal conductivity of 1.69 W m^{-1} K^{-1} and was close to the predicted value (1.59 W m^{-1} K^{-1}) [44]. Not only the target properties of HEO but also the crystal structure can be predicted by ML. Liu et al. successfully established the crystal structure prediction model for HEO, whose feature importance in descending order was anion-to-cation radius ratio, the difference in Pauling, the difference in Mulliken electronegativities, atomic size mismatch, element&content, sintering method, and entropy of mixing, being a good agreement with the law of crystal chemistry and previous experimental results [43]. To conclude, a combination of HEO and ML has been confirmed as an effective and promising strategy to study novel HEO materials and can be extended to HEO air electrodes for P-SOC.

Here, as shown in Figure 3, a continuous ML process of data collection, feature engineering, algorithm, iteration, optimization, and evaluation is given for HEO air electrode prediction [5,46]. (i) The problems related to HEO should be confirmed, which can be one or more properties, compositions, and structures. (ii) Establishing high-quality database, including the literature and DFT data, that should be representative and reasonable. (iii) Making feature engineering, i.e., the process of transforming collected data into corresponding information for use by ML models. The features should be the key points of HEO, such as coordination numbers, chemical bonding, atomic coordinates, etc. ML models can be explicable by understanding these features. (iv) ML model selection, training, and analysis, that is, a proper ML

algorithm or comparison of several algorithms, the model learns the characteristics and patterns of data and aims to minimize the training error. (v) Model performance validation and adjustment to reduce the risk of overfitting or underfitting. At the same time, the interpretability between features and target properties should be given. (vi) Application, predicting HEO air electrode, preparation, and measuring corresponding properties to verify the result of ML. In this way, not only the discovery of HEO can be realized, but also the structure-property relationships of HEO can be comprehensively understood.

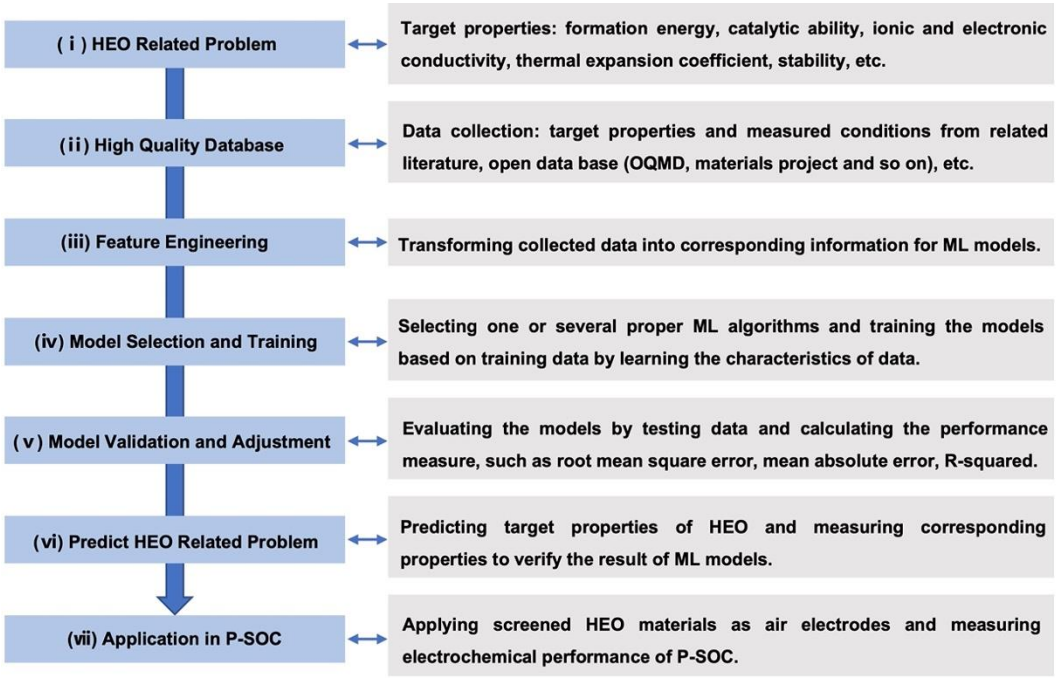

Figure 3. Schematic of an ML training process for HEO air electrode.

In summary, this perspective focuses on the development of high-performance air electrodes for P-SOC at intermediate temperatures, which requires outstanding stability, catalytic ability, and conductivity. The concept of HEO provides new possibilities for air electrode materials. However, there are still many challenges to the design of HEO air electrodes. First, the complexity of composition makes it time-consuming to prepare material and perform experimental verification. Second, the relationship between the element properties and HEO electrochemical performance needs in-depth understanding and explanation. Third, the contribution of entropy stabilization to P-SOC stability is not clear. The establishment of explainable ML models makes it possible to predict the formation energy of oxide, oxygen evolution reaction, oxygen reduction reaction, ionic and electronic conductivity, thermal expansion coefficient, reaction enthalpy of HEO and steam, and so on. In this way, a promising and efficient strategy can be provided to discover high-performance HEO air electrodes enhancing the electrochemical performance of P-SOC.

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Author Contributions

Conceptualization, C.T.; Methodology, C.T.; Validation, X.Z. and F.Z.; Formal Analysis, Q.S.; Investigation, C.T.; Resources, N.W. and S.Y.; Data Curation, C.T.; Writing—Original Draft Preparation, C.T; Writing—Review & Editing, N.W. and S.Y.; Supervision, N.W. and S.Y.

Ethics Statement

Not applicable.

Informed Consent Statement

Not applicable.

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Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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