

High-throughput screening of $\text{Sr}_x\text{A}_{1-x}\text{Fe}_y\text{B}_{1-y}\text{O}_3$ perovskites for low-temperature chemical looping air separation using graph neural networks

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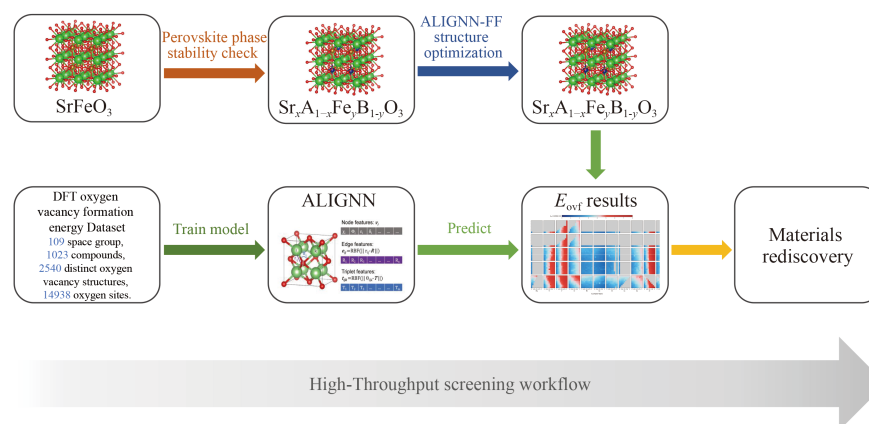
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Abstract: Low-temperature Chemical Looping Air Separation (CLAS) is a promising technology for producing oxygen-enriched gas streams. It utilizes the redox properties of solid oxygen carriers to selectively capture and release oxygen from the air. Oxygen vacancy formation energy (E_{ovf}) can be used to evaluate the ease of oxygen release by the oxides. In

this study, the applicable range of E_{ovf} for CLAS oxygen carriers was determined to be <2.3 eV using the thermodynamic calculations. A graph neural network (GNN) was trained and used to predict the E_{ovf} . By predicting E_{ovf} with the GNN model, a screening from the 3649 compositions of $\text{Sr}_x\text{A}_{1-x}\text{Fe}_y\text{B}_{1-y}\text{O}_3$ perovskites was conducted to discover the CLAS oxygen carriers. The prediction results indicate that the doping of Ba and Ca ions at the A site and Co ions at the B-site can effectively reduce the E_{ovf} of the perovskite structure. The feasibility of screening the CLAS oxygen carriers using the E_{ovf} method was validated through the rediscovery some known materials. These results support the effectiveness of GNNs in accelerating the discovery of CLAS oxygen carriers.

Key words: chemical looping air separation; oxygen vacancy formation energy; graph neural network; material discovery



Chemical looping air separation (CLAS) is an attractive technology for industrial applications, owing to its high efficiency, low energy consumption, and the potential to integrate with thermoelectric power generation cycles to reduce CO_2 emissions^[1–3]. CLAS operates using oxide-based oxygen carriers that can reversibly absorb and release O_2 in response to changes in the oxygen partial pressure or

temperature. As shown in Figure 1, when the oxygen partial pressure is low, the lattice oxygen in the oxide is released in the form of gaseous oxygen, accompanied by the reduction of the oxide. Subsequently, the reduced oxide can be regenerated in air or oxygen-rich stream. Repeating this process in a cyclic manner enables a continuous air separation and oxygen production.

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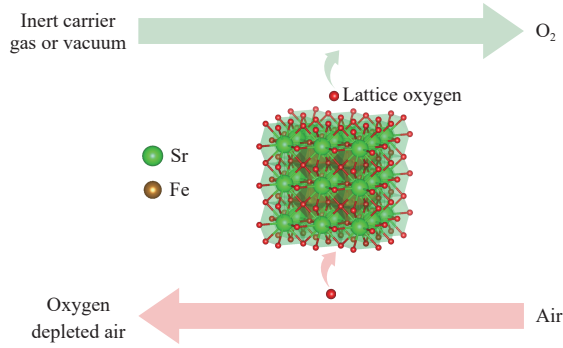


Fig. 1 Schematic illustration of the chemical looping air separation (CLAS) process with $\text{SrFeO}_{3-\delta}$ perovskite as the oxygen carrier

Perovskite materials possess superior oxygen transport capabilities, high chemical stability, and tunable redox properties, which render them ideal oxygen-carrier candidates for chemical looping (CL) processes^[1,4–6]. $\text{SrFeO}_{3-\delta}$ with a perovskite structure has been studied for low-temperature CLAS reactions (400–650 °C) due to its excellent oxygen release and absorption capabilities^[2,7]. The perovskite structure of $\text{SrFeO}_{3-\delta}$ provides the possibility of doping substitution atoms at the A-site or B-site to further regulate and enhance the material's redox properties^[7–8]. For instance, the doping of A-site with Ba^[9] or Ca^[8,10] and B-site with Co^[11–12] of SrFeO_3 has demonstrated an enhanced CLAS performance. However, the search for optimal doping elements and concentrations still relies largely on conventional heuristics and trial-and-error methods.

Oxygen vacancy formation energy (E_{ovf}) can be used to evaluate the redox performance of CLAS materials^[13–15]. Oxygen carrier materials with lower E_{ovf} are more likely to release lattice oxygen and have a better performance in low-temperature CLAS^[16–17]. Density functional theory (DFT) method is commonly used to calculate the E_{ovf} of oxides in crystal structures^[18–19]. The E_{ovf} can be calculated through Equation (1):

$$E_{\text{ovf}} = E_{\text{def}} - E_{\text{perf}} + \mu_{\text{O}} \quad (1)$$

where E_{def} is the total energy of the supercell with an O vacancy, E_{perf} is the total energy of the pristine supercell, μ_{O} is the O chemical potential. However, calculating the E_{ovf} using DFT is expensive and time-consuming.

Machine learning methods have been researched for rapid prediction of E_{ovf} . Linear models based on DFT descriptors such as band gap and formation energy can predict E_{ovf} with a mean absolute error (MAE) of 0.39 eV, without the need to construct a supercell with an oxygen vacancy^[20–21]. The unified atomistic line graph neural network force-field (ALIGNN-FF), trained with formation energy data of the defect-free crystals, can be directly applied to predict the E_{ovf} with an MAE of 0.87 eV^[22–23]. The defect graph neural network (dGNN) model, trained with E_{ovf} data calculated by DFT, predicts the E_{ovf} of oxides (containing elements present in the training set) with an MAE of 0.52 eV^[14].

In this study, an ALIGNN model was trained using a curated dataset of E_{ovf} and used for the prediction of E_{ovf} .

The range of E_{ovf} suitable for CLAS oxygen carriers was estimated using thermodynamic calculations. High-throughput screening for CLAS was performed on 3649 $\text{Sr}_x\text{A}_{1-x}\text{Fe}_y\text{B}_{1-y}\text{O}_3$ perovskites through stability judgment and E_{ovf} prediction. The feasibility of the E_{ovf} method for screening CLAS oxygen carriers was confirmed through materials rediscovery.

1 Methodology

1.1 Machine learning

The charge-neutral E_{ovf} dataset^[14,24] utilized in this article was based on DFT-computed data using the Generalized Gradient Approximation (GGA) with Hubbard U correction. The dataset comprises 48 cationic elements, 102 space groups, 901 compounds, 2329 distinct oxygen vacancy structures, 9342 oxygen sites (see Section S1 in Supplemental Information). The distribution of elements in the dataset is shown in Figure 2(a).

The Atomistic Line Graph Neural Network (ALIGNN) demonstrated superior performance over other graph neural networks in multiple atomistic prediction tasks by incorporating bond angle information^[23,25]. As shown in Figure 2(b), the ALIGNN was trained to predict the E_{ovf} at all oxygen sites within the crystal structures. Details on the ALIGNN can be found in the related paper^[23,25]. In brief, each node in the atomistic graph is assigned 9 input node features based on its atomic species: electronegativity, group number, covalent radius, valence electrons, first ionization energy, electron affinity, block and atomic volume. The inter-atomic bond distances are used as edge features with a radial basis function up to a cut-off of 8 Å. A periodic 12-nearest-neighbor graph construction was used. This atomistic graph is then used for constructing the corresponding line graph using interatomic bond-distances as nodes and bond-angles as edge features. The dataset was partitioned into training, validation, and test sets at a ratio of 80:10:10, and an ALIGNN model was trained to predict the E_{ovf} .

1.2 Thermodynamic calculations

The E_{ovf} predicted by the ALIGNN corresponds to the zero-temperature limit. Therefore, thermodynamic calculations are required to determine the range of E_{ovf} values that are suitable for various chemical looping applications. In light of the comparable E_{ovf} across distinct sites in the ideal perovskite structure^[14,26], it is reasonable to assume that the properties of oxygen vacancies have a degree of similarity among these sites. At a specified temperature T , the Gibbs free energy for the formation of oxygen vacancies in the crystalline structure is expressed as follows^[27]:

$$\Delta G = n * h_{\text{ovf}} - T \Delta S_{\text{cf}} \quad (2)$$

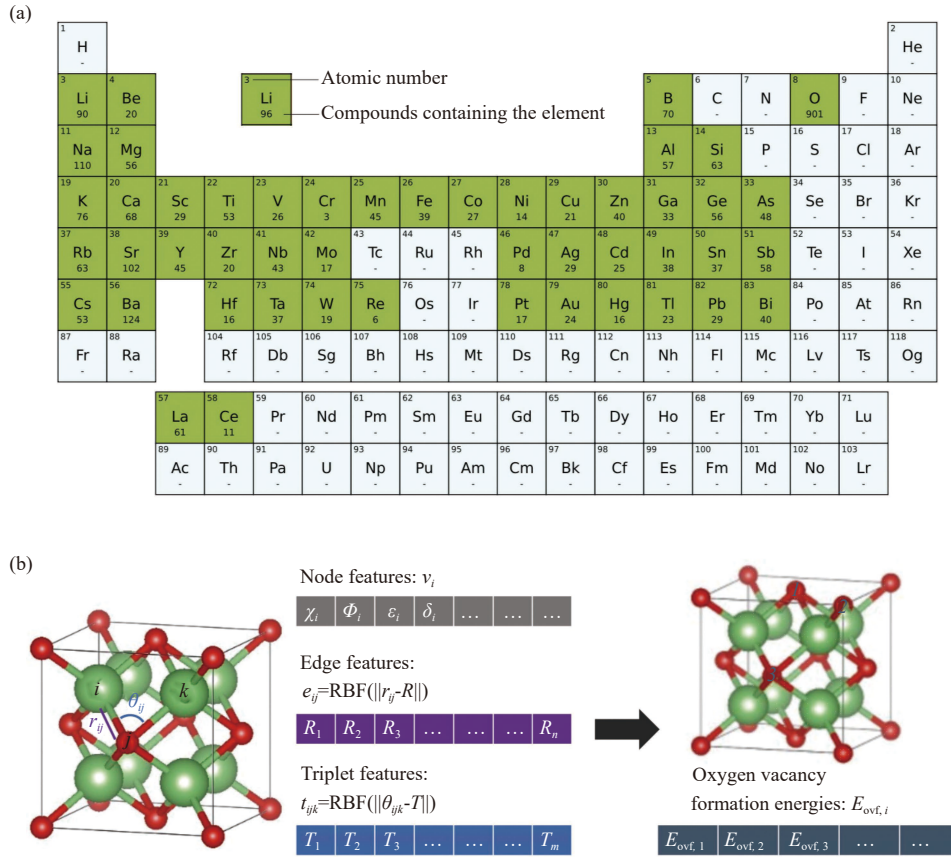


Fig. 2 (a) The distribution of elements in the dataset, (b) Schematic diagram for predicting the E_{ovf} using the ALIGNN model

where n represents the number of vacancies, h_{ovf} denotes the formation enthalpy per defect (including contributions from vibrational enthalpy and entropy for each defect), and ΔS_{cf} is the configurational entropy. Assuming a total number of N atoms in the system (where $N \gg n$), the structural entropy is derived *via* Stirling's approximation as follows^[27]:

$$\Delta S_{\text{cf}} = k_B \times n \times (\ln N - \ln n) \quad (3)$$

In the context of solids within the zero-temperature limit, the volume change is negligible, and thus the h_{ovf} can be reasonably approximated by the E_{ovf} ^[28]. The chemical potential $\Delta\mu$ is introduced to determine the Gibbs free energy of oxygen vacancy formation under the relevant thermodynamic constraints^[14]:

$$\Delta G = n \times (E_{\text{ovf}} + \Delta\mu) - T \times k_B \times n \times (\ln N - \ln n) \quad (4)$$

when the system reaches equilibrium, the condition $\Delta G = 0$ leads to the following expression for the dimensionless oxygen vacancy concentration:

$$c_v = \frac{n}{N} = e^{-(E_{\text{ovf}} + \Delta\mu)/(k_B T)} \quad (5)$$

Given a specific oxygen vacancy concentration, the corresponding $\Delta\mu$ at the thermodynamic constraints can be determined by inverting the above expression^[14]. By comparing the $\Delta\mu$ curves corresponding to different E_{ovf} values on the Ellingham diagram, the applicable ranges of E_{ovf} for different chemical looping applications can be identified^[13–14].

1.3 High-throughput screening for $\text{Sr}_x\text{A}_{1-x}\text{Fe}_y\text{B}_{1-y}\text{O}_3$

The $\text{Sr}_x\text{A}_{1-x}\text{Fe}_y\text{B}_{1-y}\text{O}_3$ perovskite structures were constructed by substituting A- and/or B-site cations in SrFeO_3 using the pymatgen package^[29]. Table 1 summarizes the dopant types and concentrations investigated in this study. By exhausting all possible combinations of A- and/or B-site dopants in the selected cation network, 3520 perovskite models were constructed. Through a preliminary screening process, some of these combinations were filtered out due to their inability to form stable perovskite phases. Firstly, the total valence of the compositions was examined to determine whether it can be zero (see Section S2 in Supplemental Information). Besides charge neutrality, a modified tolerance factor $\tau = \frac{r_O}{r_B} - n_A \left(n_A - \frac{r_A/r_B}{\ln(r_A/r_B)} \right)$ ^[30] was applied to estimate the stabilities of the perovskite structures, where r_O , r_A , r_B represent the radii of oxygen anion, and the A- and B-site cations (see Table S1 in Supplemental Information). n_A and n_B are the oxidation states of the A- and B-site cations, respectively. The modified tolerance factor (τ) carries more chemical information and exhibits better predictive ability than the classical Goldschmidt tolerance factor^[4,30].

After the initial construction of the doped crystal structures, the fast structure optimization was performed using the ALIGNN-FF^[23] model, for which the model parameters were directly adopted from the literature^[23] and

shared publicly. The ALIGNN-FF was developed using both energies and forces for applications, such as, in fast structure optimization. Then, the optimized crystal structures were input into the ALIGNN model which is then trained for E_{ovf} to predict the E_{ovf} for each oxygen site. The average of all the E_{ovf} values for each crystal structure was calculated and used as an indicator to evaluate the redox performance. The process is illustrated in Figure 3.

2 Results and discussion

2.1 The range of E_{ovf} for oxygen carrier selection

As shown in Figure 4, the Gibbs free energy of several key reactions is used to distinguish different chemical looping reactions^[13,31]. The data of reactions was from calculations with the FactSage software. For a target oxygen vacancy concentration of $[\text{V}_\text{O}] = 1\%$, Figure 4 shows the oxygen vacancy formation chemical potential $\Delta\mu$ at various temperatures for different values of E_{ovf} . It has been shown that the trends of E_{ovf} across different perovskite oxides remain the same for different extents of oxygen vacancy^[32]. Therefore, it is reasonable to estimate the performance of the oxygen carrier by using E_{ovf} of the chemical potential corresponding to $[\text{V}_\text{O}] = 1\%$. From Figure 4, it can be inferred that for CLAS, the suitable E_{ovf} values should be approximately less than 2.3 eV when the temperature is higher than 500 °C. Previous studies have indicated that materials with E_{ovf} less than 2.5 eV are suitable for thermochemical O_2 storage and pumping/separation^[13,15], which is in agreement with the estimated results. Additionally,

this estimation method can also be applied to assess the suitability of materials in fields such as chemical looping full oxidation (CLFO), chemical looping with oxygen uncoupling (CLOU), and chemical looping syngas production.

2.2 The training results of the ALIGNN model for predicting E_{ovf}

The ALIGNN model trained for predicting E_{ovf} was tested. The oxides in the test set did not exist in the training set or the validation set. As shown in Figure 5, the ALIGNN model achieved a mean absolute error (MAE) of 0.26 eV in predicting the E_{ovf} values at different oxygen sites of the oxides in the test set, and the coefficient of determination (R^2) reached 0.94. The closer the MAE is to 0 and the R^2 is to 1, the higher the accuracy of the model. This result outperforms the linear model using the DFT descriptors (0.39 eV)^[20–21] and the dGNN model based on a simpler convolutional architecture relative to the ALIGNN (0.52 eV)^[14].

2.3 High-throughput screening of $\text{Sr}_x\text{A}_{1-x}\text{Fe}_y\text{B}_{1-y}\text{O}_3$

The screening results from the 3649 compositions of $\text{Sr}_x\text{A}_{1-x}\text{Fe}_y\text{B}_{1-y}\text{O}_3$ perovskites are presented in Figure 6. Most of the compositions containing ions of Na, K, Mg and Cu were filtered out due to the inability to achieve electrical neutrality. It is noted that the nonstoichiometric cases of perovskites ($\text{Sr}_x\text{A}_{1-x}\text{Fe}_y\text{B}_{1-y}\text{O}_{3-\delta}$) were not considered in the preliminary screening process. The different grids in the Figure 6 may represent the same chemical composition.

Table 1 A- and B-site dopant elements, x and y in $\text{Sr}_x\text{A}_{1-x}\text{Fe}_y\text{B}_{1-y}\text{O}_3$ investigated

Variable	Parameter values
A-site elements	Na, K, Ca, Ba, La
B-site elements	Mg, Ti, V, Cr, Mn, Co, Ni, Cu, Ge, Zr, Sn
x	0, 0.125, 0.25, 0.375, 0.5, 0.625, 0.75, 0.875, 1
y	0, 0.125, 0.25, 0.375, 0.5, 0.625, 0.75, 0.875, 1

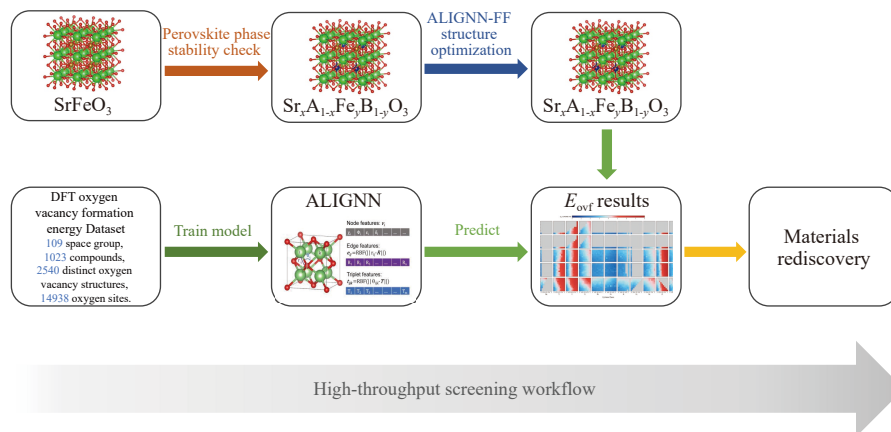
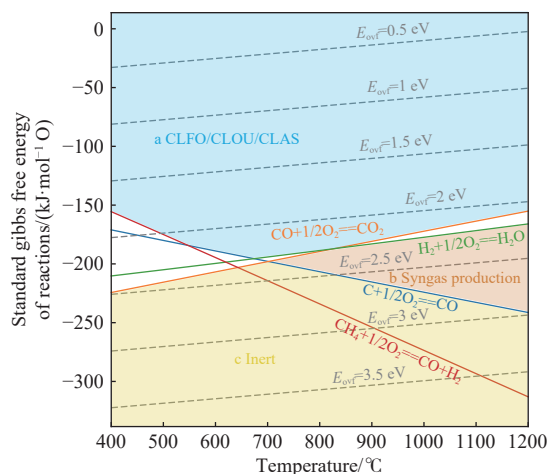
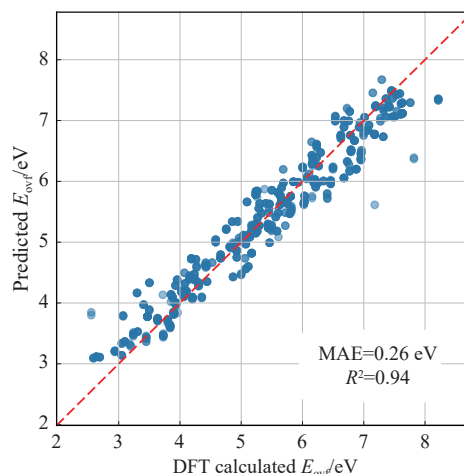


Fig. 3 The flowcharts for high-throughput materials screening

Fig. 4 The range of E_{ovf} for chemical looping

The slight differences in the average E_{ovf} for the same material, such as $\text{Sr}_x\text{Ba}_{1-x}\text{Fe}_1\text{Ti}_0\text{O}_3$ and $\text{Sr}_x\text{Ba}_{1-x}\text{Fe}_1\text{V}_0\text{O}_3$, may be attributed to random errors introduced during the crystal structure optimization process. For the $\text{Sr}_{1-x}\text{Ca}_x\text{FeO}_3$, it is known that the E_{ovf} first decreases and then increases with the increase of the Ca^{2+} concentration from 0 to 0.5^[10]. This is consistent with the predictions. In certain compositions, such as $\text{Sr}_{0.125}\text{Ba}_{0.875}\text{CoO}_3$, the doping of the Ba ion at the A-sites also leads to a decrease in E_{ovf} . The doping of the Co ion at the B-sites shows a pronounced trend of decreasing E_{ovf} with an increasing concentration. Among all the compositions with a predicted average E_{ovf} below 2.0 eV, the Co ion is consistently present (see Section S4 in Supplemental Information).

Fig. 5 The results of the ALIGNN model for predicting E_{ovf} on the test set

2.4 Rediscovery of low-temperature CLAS materials

Through the screening criterion $E_{\text{ovf}} < 2.3$ eV, a series of known low-temperature CLAS oxygen carriers were rediscovered, as shown in Table 2. This indicates that the E_{ovf} can be used to accelerate the material discovery for the low-temperature CLAS process. Still, this method has certain limitations. The E_{ovf} can be used to assess the ease of oxygen release from the oxides, but it cannot be used to evaluate the material's re-oxidation extent. For example, although the predicted E_{ovf} for BaCoO_3 is as low as 1.44 eV, it is difficult to be fully oxidized after oxygen loss, making it unsuitable for low-temperature CLAS^[12].

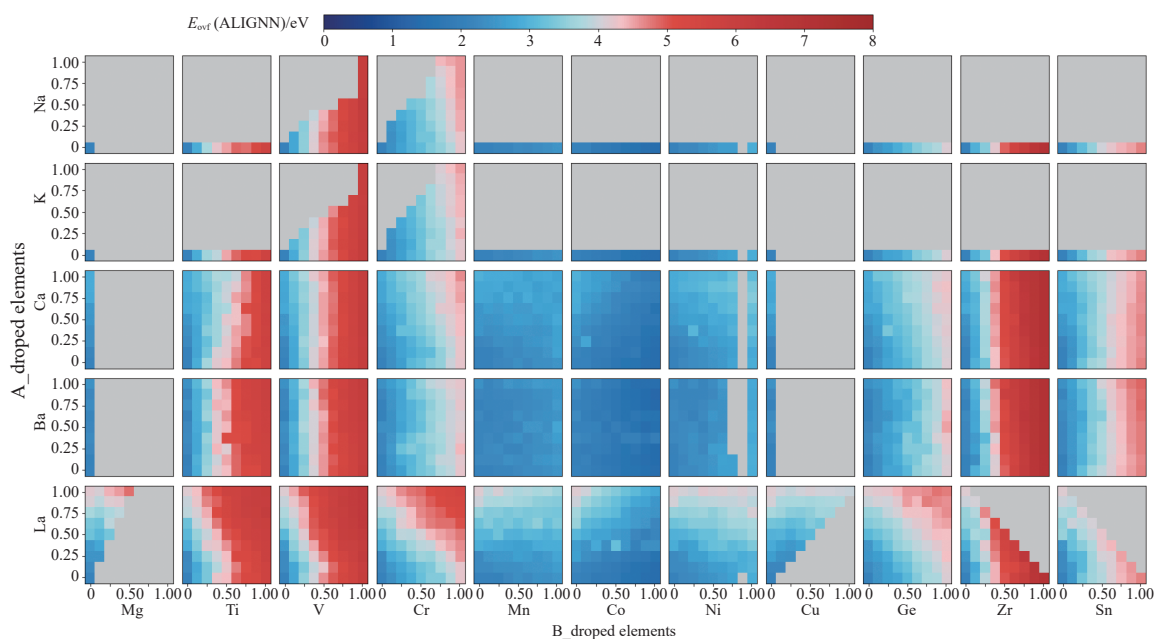
Fig. 6 The predicted average E_{ovf} of the $\text{Sr}_x\text{A}_{1-x}\text{Fe}_y\text{B}_{1-y}\text{O}_3$ perovskites (The axes represent the doping elements and their proportions at A-site and B-site. The grey grid represents compositions determined to be unable to form stable perovskite structures)

Table 2 The rediscovered perovskites oxygen carriers for low-temperature CLAS

Composition	CLAS performance	Predicted E_{ovf} /eV
$\text{SrFeO}_{3-\delta}$ [3]	Oxygen carrying capacity was 0.5% at 500 °C (0–0.2 atm p_{O_2}), the onset temperature for O_2 release was around 400 °C	2.16 (SrFeO_3)
$\text{SrFe}_{0.25}\text{Co}_{0.75}\text{O}_{3-\delta}$ [3]	Oxygen carrying capacity was 1.3% at 500 °C (0–0.2 atm p_{O_2}), the onset temperature for O_2 release was around 290 °C	1.64 ($\text{SrFe}_{0.25}\text{Co}_{0.75}\text{O}_3$)
$\text{Sr}_{0.8}\text{Ca}_{0.2}\text{Fe}_{0.4}\text{Co}_{0.6}\text{O}_{3-\delta}$ [7]	Oxygen carrying capacity was 1.11% at 450 °C (0–0.2 atm p_{O_2})	1.77 ($\text{Sr}_{0.75}\text{Ca}_{0.25}\text{Fe}_{0.375}\text{Co}_{0.625}\text{O}_3$)
$\text{Sr}_{0.76}\text{Ca}_{0.24}\text{FeO}_{3-\delta}$ [33]	Oxygen carrying capacity was 1.2% at 500 °C (0.05–0.2 atm p_{O_2})	2.24 ($\text{Sr}_{0.75}\text{Ca}_{0.25}\text{FeO}_3$)
$\text{Sr}_{0.75}\text{Ba}_{0.25}\text{FeO}_{3-\delta}$ [9]	Oxygen carrying capacity was 0.73% at 400–600 °C (0.2 atm p_{O_2})	2.06 ($\text{Sr}_{0.75}\text{Ba}_{0.25}\text{FeO}_3$)

3 Conclusions

Based on the graph neural network for predicting the E_{ovf} , this work proposes a fast method for the screening of low-temperature CLAS oxygen carriers, and conducts a high-throughput screening from the 3649 compositions of $\text{Sr}_x\text{A}_{1-x}\text{Fe}_y\text{B}_{1-y}\text{O}_3$ perovskites. Through the thermodynamic estimation, it was found that the E_{ovf} value for the CLAS material should be less than 2.3 eV. An the ALIGNN model was trained for the prediction of the E_{ovf} and the test set shows that the model has a MAE of 0.26 eV. In the high-throughput screening, the doping of the Co ions at the B-site was found to be able to effectively reduce the E_{ovf} of the perovskite structure. And the doping of specific proportions of Ba and Ca ions at the A site can also contribute to lowering the E_{ovf} . Under the screening criterion of $E_{\text{ovf}} < 2.3$ eV, the model rediscovered several previously reported oxygen carriers for the low-temperature CLAS. This work demonstrates that using GNN to predict E_{ovf} can accelerate the discovery process of CLAS materials. In a broader context, the presented methods may also accelerate the discovery of materials for other related chemical looping applications, such as chemical looping full oxidation and chemical looping syngas production.

Declaration of Competing Interest

The authors declare no conflict of interest.

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使用图神经网络高通量筛选用于低温化学链空气分离的 $\text{Sr}_x\text{A}_{1-x}\text{Fe}_y\text{B}_{1-y}\text{O}_3$ 钙钛矿

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摘 要: 低温化学链空气分离 (CLAS) 是一种很有前景的富氧气流生产技术。它利用固体载氧体的氧化还原特性选择性地捕获和释放空气中的氧气。氧空位形成能 (E_{ovf}) 可用于评估氧化物释放氧的难易程度。本研究通过热力学计算确定了 CLAS 载氧体的 E_{ovf} 适用范围 < 2.3 eV, 训练了一个图神经网络 (GNN) 并用于预测 E_{ovf} 。通过 GNN 模型预测 E_{ovf} , 筛选了 3649 种 $\text{Sr}_x\text{A}_{1-x}\text{Fe}_y\text{B}_{1-y}\text{O}_3$ 钙钛矿成分。结果表明, A 位 Ba 和 Ca 离子的掺杂以及 B 位 Co 离子的掺杂可以有效降低钙钛矿结构的 E_{ovf} 。通过对已知 CLAS 材料的重新发现, 验证了使用 E_{ovf} 方法筛选 CLAS 氧载体的可行性。这些结果表明了图神经网络预测氧空位形成能在加速 CLAS 载氧体材料发现中的有效性。

关键词: 化学链空气分离; 氧空位形成能; 图神经网络; 材料发现

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