Designing High Entropy Oxides for Fuel Cell Using Machine Learning Potential

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High-entropy materials are a new frontier of functional materials for applications such as thermal protection, catalysis and batteries [1]. Because of the very large chemical space, computational materials modelling can help in identify new high high-entropy ceramics with optimal properties [Fig 1]. In this talk, we report our efforts to develop an automated framework that integrates machine learning methodologies with high-throughput computational processes, enhancing the efficiency of discovering novel high-entropy oxides suitable for fuel cell applications. This framework has its foundation in a recent dynamic workflow management system, called PerQueue [2]. PerQueue allows a new level of flexibility in integrating Density Functional Theory (DFT) simulations for the estimation of structural and kinetic properties, with machine learning models, to predict and explain the origin of these properties. The calculation of kinetic properties (via the Nudged Elastic Band, NEB, method [3]), in particular, are still orders of magnitude more computationally demanding than the estimation of the basic structure of the materials. In addition, the intrinsic disorder of these materials requires many barriers to be calculated for each composition, which is unfeasible.

Machine learning models can help us in this. Starting from the MACE foundation model, [4, 5] we have fine-tuned it using our unique dataset composed of several thousands of unique high-entropy oxe-perovskite materials. The dataset contains pristine materials as well as single oxygen vacancies and transition state structures. The aim of this approach is multifold. From one side, we assess how many structures of each category is needed to train a MACE model to have a good predictive power [Fig 2.] (mean absolute error below ~12 meV/atom), reducing the amount of expensive calculations to include in favor of cheaper ones; from the other, we want to define a model to predict high-entropy oxides with low-oxygen diffusion barriers (i.e., candidates to be used as cathode materials in proton conducting fuel cells). Finally, we develop an explainable AI model, in the framework of the SISSO model, to identify the origin of low/high oxygen diffusion barriers, defining criteria for the design of high-performance oxygen conductors.

References

[1] Oses, C., Toher, C. & Curtarolo, S. High-entropy ceramics. Nat. Rev. Mater. 5, 295–309 (2020).
[2] B. H. Sjølin, W. S. Hansen, A. A. Morin-Martinez, M. H. Petersen, L. H. Rieger, T. Vegge, J. M. García-Lastra and I. E. Castell, Digital Discovery, 2024, , 3(9), 1832– 1841.
[3] G. Henkelman, B. P. Uberuaga, H. Jónsson, J. Chem. Phys. 2000, 113, 9901–9904.
[4] I. Batatia, et al., arxiv, Chemical Physics and Materials Science (2024).
[5] I. Batatia, D. P. Kovacs, G. N. C. Simm, C. Ortner, and G. Csanyi, MACE: Higher order equivariant message passing neural networks for fast and accurate force fields, Adv Neural Inf Process Syst (2022).

Figures

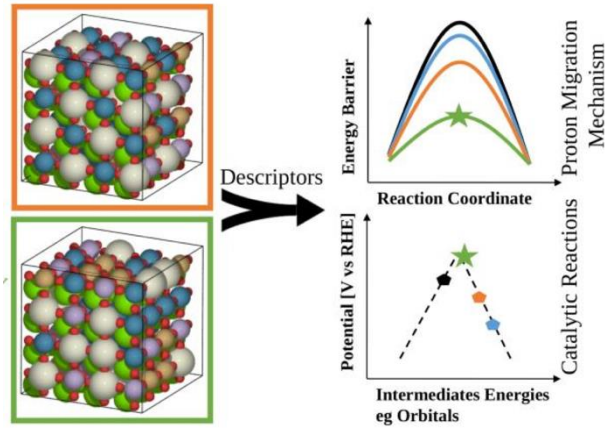

Figure 1. Ordered (top/orange) and disorder (bottom/green) oxides. Our goal is to use disorder to generate structures with improved properties for fuel cell.

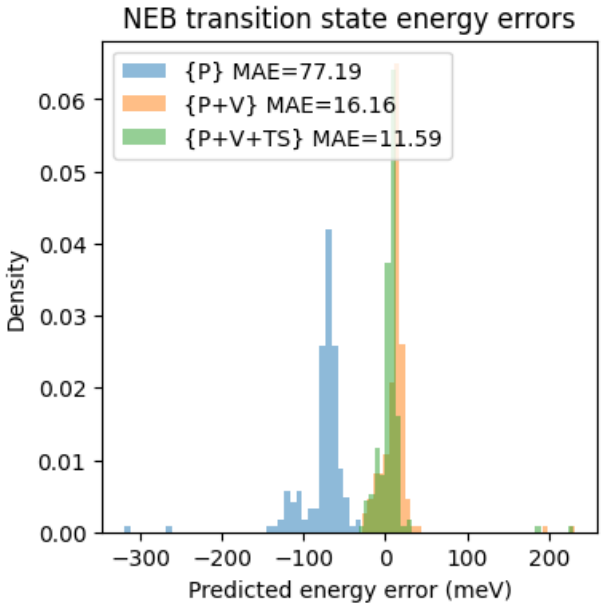

Figure 2. The predicted energy error of the NEB transition state from finetuning the MACE model on three different datasets. We have finetuned with datasets containing pristine- (blue); pristine and vacancies- (orange); pristine, vacancies and transition state (green) structures. The corresponding MAE is reported in the legend.