Accelerated Discovery of Perovskite Solid Solutions through unsupervised material fingerprints and Automated Materials Synthesis

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Abstract

Accelerating perovskite discovery and synthesis is vital for advancing wireless communication and biosensors. The complexity of compositional disorder and diverse chemical spaces poses significant challenges. To address this, we present an automated framework combining machine learning (ML), robotic synthesis, and high-throughput characterization. An unsupervised deep learning strategy identifies material fingerprints from chemical compositions, enabling predictions of crystal symmetry and facilitating analogical materials discovery. This approach streamlines the search across ~600,000 compounds with a 94% success rate. Validated through rapid synthesis of solid solutions like (BaxSr1-x)CeO3, the platform reduces processing times to minutes, bridging gaps between established knowledge and unexplored materials.

Introduction

Perovskite oxides, with their general ABO_3 crystal structure, are an essential class of materials due to their unique properties, such as high ionic conductivity and ferroelectricity, which make them applicable in a variety of technologies, including ferroelectric photovoltaics, solid oxide fuel cells, electrocatalysis, and resistive switching memories. [1] Traditionally, ordered materials were considered superior to their disordered counterparts, but recent studies have shown that chemically disordered or alloyed perovskite oxides often outperform ordered ones, offering enhanced functionality for a wide range of applications. This realization has led to a surge in interest in exploring and synthesizing disordered perovskite oxides, yet the vast compositional space and complex synthesis pathways have made this a challenging endeavor.
To address this, ML has emerged as a powerful tool for accelerating material discovery by predicting the properties and stability of materials based on large datasets. After making these predictions, the next challenge is synthesizing materials into single-phase. To assess phase stability under various conditions, CALPHAD computational methods and density functional theory (DFT) are frequently used.[2] These techniques typically assume an equilibrium thermodynamic state, which may not fully account for the complexities of perovskite oxides, necessitating careful analysis. Additionally, integrating temperature effects into ab-initio simulations is a demanding process, requiring methods such as quasi-random structures or molecular dynamics. These approaches are often difficult to apply to ML-predicted materials due to the limitations in available synthesis data. To overcome these challenges, artificial intelligence (AI) and collaborative lab robots have been integrated into "self-driving laboratories" (SDLs) to accelerate material discovery and synthesis. Figure 1 presents a comprehensive customizable SDL loop for the synthesis and characterization of perovskites, highlighting the process through a combination of both AI and expert scientific input.

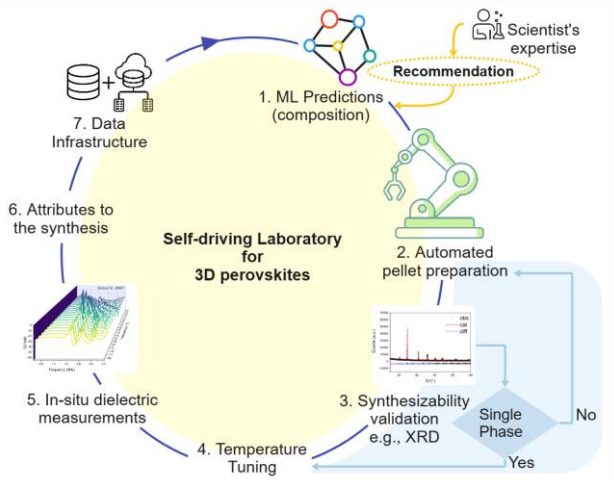

Figure 1. The workflow concept. Starting from the top right and moving clockwise, the sequence initiates with ML-models recommending sample library compositions, followed by automated pellet creation via the solid-state reaction, method, synthesizability validation (e.g., XRD), temperature tuning, real-time dielectric property measurements, and analysis linking dielectric attributes to the synthesis process. The cycle concludes with the updating of archives and the generation of AI-driven predictions for comprehensive cycle management and decision-making.

Method

In this study, we used ML to screen a vast library of potential perovskite compositions, focusing on disordered quaternary perovskites. The ML models were trained using a dataset that included over 1,700 perovskites and 227 non-perovskites from the Inorganic Crystal Structure Database (ICSD). A gradient boosting classifier, trained on this data, achieved a 94% accuracy rate, allowing us to identify nearly 5,000 promising candidates from a pool of 600,000 possible compositions.
To refine these predictions, we employed data mining techniques, including composition embeddings from variational autoencoders (VAE) and Mat2vec. The embeddings helped rank candidates based on their similarity to known perovskite structures, while expert feedback and existing literature were incorporated to prioritize materials with high synthesis feasibility.

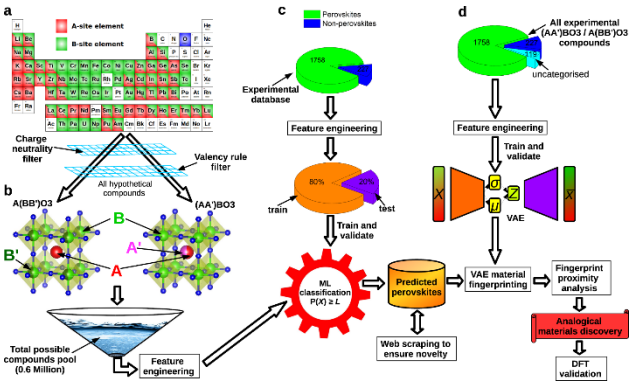

Figure 2. a) Elements that can fully or partially occupy octahedral and interstitial sites. b) Average unit cells of and materials, usually realized by XRD analysis, c) A gradient boosting classifier is iteratively trained on sampled experimental data, and d) training VAE model.

A selected set of these ML-predicted materials was synthesized using an automated approach, ASAP, as reported in the reference. ASAP systematically explored sintering variables i.e., sintering hold time, sintering temp, heating- and cooling- rates.[3]
The conditions were optimized through a contour plot, identifying ranges where single-phase perovskite structures were successfully synthesized. Experimental validation with X-ray diffraction (XRD) confirmed phase purity in the synthesized materials, illustrating the effectiveness of the ML-guided approach and automated laboratory.

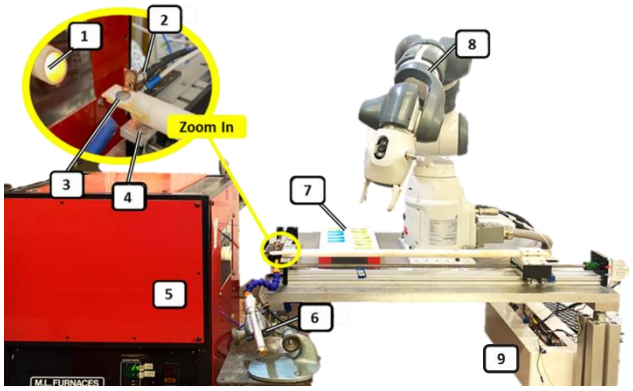

Figure 3. The ASAP experimental setup for automated sintering and dielectric characterisation of perovskites. 1) Tube furnace, 2) automated dielectric sensor connected to vector network analyser and MATLAB graphical user interface, 3) sample holder and sample under test, 4) infrared temp sensor, 5) automated high-temperature furnace, 6) vortex tube, 7) pellet tray, and (8) cobot.

Results

Among different selected compositions for testing, two selected compositions, $(Ba_{0.8}Sr_{0.2})CeO_3$ and $(Ba_{0.4}Sr_{0.6})CeO_3$, were successfully synthesized as single-phase materials. XRD analysis revealed that $(Ba_{0.8}Sr_{0.2})CeO_3$, sintered at 1400°C for 10 minutes, and $(Ba_{0.4}Sr_{0.6})CeO_3$, sintered at 1250°C for 25 minutes, achieved a single-phase perovskite structure. A contour plot, Figure 4, shows the relationship between sintering conditions and XRD phase purity indicating that single-phase perovskite materials could be synthesized under a variety of conditions. Specifically, the plot demonstrated that high sintering temperatures (>1350°C) for shorter times (<20 minutes) and lower temperatures for longer times could both yield single-phase structures.

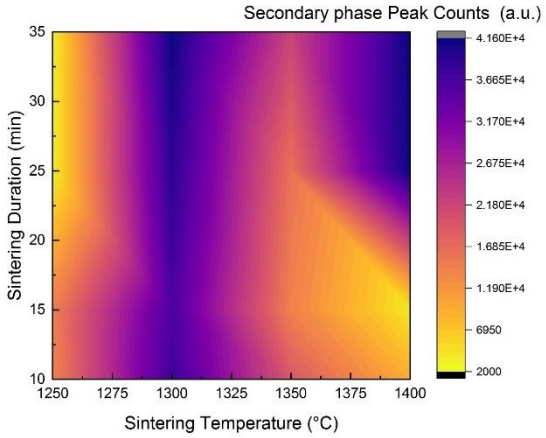

Figure 4. Process-Structure analysis for ML predicted composition. The contour plot of automated rapid sintering conditions displays the investigated range of variables and their impact on the XRD secondary phase peak of ML-predicted composition $(BanSr_{1-n})CeO_3$.

Discussion

The successful synthesis of ML-predicted compositions highlights the potential of machine learning in guiding the discovery and synthesis of new materials, particularly in the case of complex disordered systems. The results demonstrate that ML can effectively reduce the trial-and-error nature of material discovery, allowing for a more systematic exploration of compositional spaces and synthesis conditions. However, the study also underscores the limitations of relying solely on ML predictions. Synthesis challenges, such as non-stoichiometry and the need for careful control of processing conditions, remain significant. Incorporating human expertise and feedback into the ML workflow can help address these challenges, ensuring that ML predictions are grounded in experimental feasibility. Further work will focus on enhancing the AI models, expanding the dataset using the automated setup, and exploring additional synthesis strategies to discover even more high-performance perovskites.

References

[1] Ihalage, A., Hao, Y. (2021). Analogical discovery of disordered perovskite oxides by crystal structure information hidden in unsupervised material fingerprints. *npj Comput Mater* **7**, 75.
[2] Vecchio, K. S., Dippo, O. F., Kaufmann, K. R. & Liu, X. (2021). High-throughput rapid experimental alloy development (HT-READ). Acta Mater 221, 117352.
[3] Omidvar, M., Zhang, H., Ihalage, A.A. et al. (2024). Accelerated discovery of perovskite solid solutions through automated materials synthesis and characterization. Nat Commun 15, 6554.