Synthesis of cuprate superconductors predicted from a closed loop machine learning model to accelerate materials discovery

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Mechanisms for superconductivity below a critical temperature in materials with a superconducting phase transition have not been established, restricting the design and production of new high-temperature superconductors. In this work, we investigate the implementation of generative AI in conjunction with experimental data to produce a closed loop machine learning model to accelerate the discovery of superconductors. We use solid state and hydrothermal processes to synthesize predicted cuprate superconductors. Characterization of resulting diffraction patterns and isolation of unknown phases contributes raw data and human classification to the machine learning model to improve future materials predictions. Possible new phases were synthesized based on AI predictions; however, superconducting properties from the realized compounds were not observed.

Introduction

Materials that exhibit superconducting properties at ambient conditions have potential for a wide array of applications including efficient electric power generation and transportation, quantum computing, and high-resolution MRI devices. Superconductivity is described by the ability to conduct electrical current with zero resistance and expel an external magnetic field below a critical temperature (T_C). Currently, materials that undergo a phase transition to a superconducting state typically do so only when cooled to temperatures between 0 K and 10 K via liquid helium, greatly limiting the general applicability of superconductors.

High-temperature superconductors are materials with transition temperatures from 30 K to 140 K. Cuprate superconductors are a family of such materials and have the highest known T_C's at ambient pressure. The layered structure of charge reservoirs and copper oxide (CuO_2) planes characteristic of cuprates is thought to allow for superconducting current to flow in the CuO_2 planes.

Despite many theories of superconductivity, the mechanism for this property is not defined for all superconductors, making it difficult to propose and synthesize new materials with superconducting properties. In this study, we explore the possibility of synthesizing materials predicted in a closed loop machine learning approach to accelerate the discovery of novel superconductors.

The closed loop process begins with a generative artificial intelligence (AI) model, a collaboration with the Johns Hopkins University Applied Physics Laboratory and Microsoft, predicting millions of complex chemical formulas and structures for possible superconductors, varying from three to eight different elements in the composition. A down selection process occurs to isolate materials in the cuprate family with feasible structures and with the highest transition temperatures. Choosing a synthesis method and subsequent parameters, including reagents and heating profile, we attempt to synthesize the predicted material. After characterization of the sample, the synthesis and characterization process can be iterated multiple times to reach a desired product. The samples are tested for superconductivity near 0 K, and the experimental data, both raw data and human interpretation, are fed back into the machine learning model.

Methods

The predicted compounds were synthesized by solid state synthesis: combining the stoichiometric ratios of selected carbonate or oxide reagents with a mortar and pestle to form a homogenous powder. The powder was then placed in an alumina (Al_2O_3) crucible and annealed in a furnace ramping up to temperature at 200 °C/h, dwelling at 800 °C for 12 h, and ramping down at

200 °C/h. After recombining the powder, a similar annealing process was repeated with a dwell temperature of 900 °C.

Solid state synthesis is a common method for initial materials creation, but hydrothermal synthesis provides more parameters to tune and was explored as a secondary approach to solid state synthesis. The addition of a solvent in a PTFE lined vessel, housed in a hydrothermal bomb, can change the acidity, solubility, and oxidizing potential of the reagents, altering the reaction that occurs. For predicted materials that do not preferentially form in air, hydrothermal techniques can be a means to achieve alternate oxidation states. Pressure can also be tuned by varying the amount of solvent in the vessel.

Samples were characterized after each annealing step with a Powder X-Ray Diffractometer (PXRD) to determine phases present in the powder. An x-ray source is diffracted by the sample and sensed by the x-ray detector, characterizing the chemical composition and structure type. Diffraction patterns were verified with existing phases found in the Bruker Eva database and Inorganic Crystal Structure Database (ICSD). Patterns that did not fit with available data indicate the possible synthesis of a new phase.

Tests for superconductivity were performed with a Magnetic Property Measurement System (MPMS) to quantify the alternating current (AC) susceptibility vs temperature as the material was cooled from 300 K to 2 K.

Results

Majority unknown phases were observed for several samples targeting AI predicted and down selected cuprate superconductors (Figure 1). By omitting reagents and varying the stoichiometric ratios, we explored the vast phase space of the predictions to identify the elements and quantities of reagents composing the unknowns. Unknown patterns varied with the two heating profiles and were often realized or intensified at the higher temperature. The unknown phases were specified as such and reinputted into the machine learning model. Most targeted materials were a mixture of known binaries, ternaries, and quaternaries, essentially functioning as null results for the machine learning model. No phase pure unknown samples were discovered but additional effort spent isolating the unknown patterns could achieve this result, leading to successful crystal growth and characterization. Previously unidentified diffraction patterns are valuable results for the advancement of generative AI and could suggest the realization of new materials.

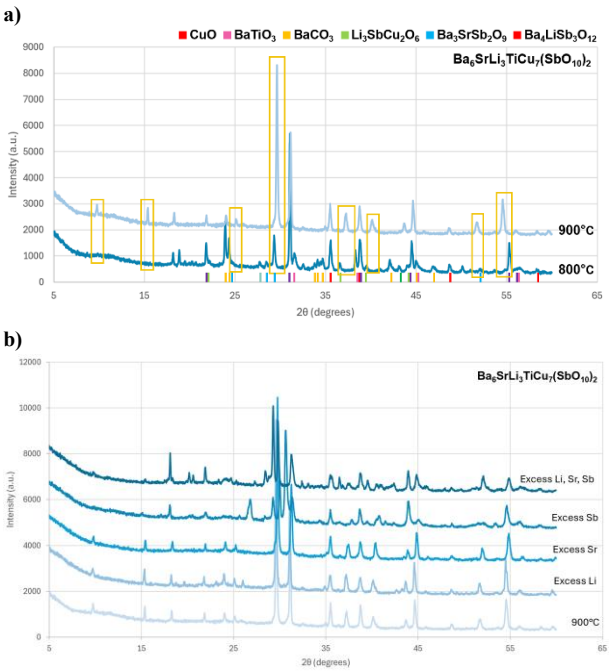

Figure 1. (a) PXRD scans from synthesis of predicted material $Ba_6SrLi_3TiCu_7(SbO_{10})_2$ at different heating profiles. Scans show both a mixture of known patterns and a majority phase unknown, prevalent at 900°C and highlighted by yellow rectangles. (b) Attempt to isolate unknown peaks by tuning the stoichiometric ratio of reagents. Excess antimony alters the pattern. Excess lithium and strontium preserve the unknown peaks, indicating they are present in the unknown composition.

MPMS measurements concluded that the predicted and synthesized compounds are not superconducting (Figure 2). Superconductivity is indicated by a sharp downward slope to a negative AC susceptibility value as the temperature approaches 0 K, illustrating the expulsion of a magnetic field. The complex compositions and structures of the predictions prevented the formation of the intended materials via solid state and hydrothermal synthesis; thus, the predicted structures may be favorable for superconductivity but require further synthesis attempts.

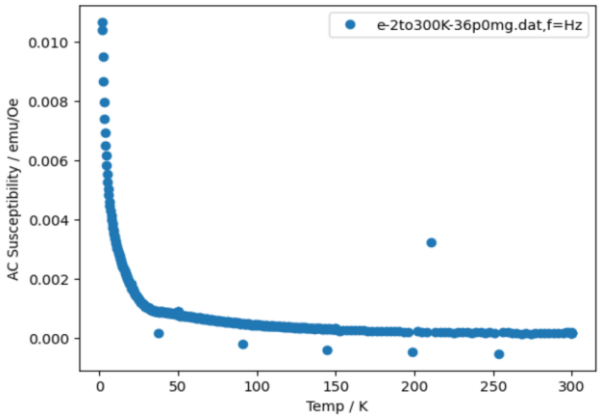

Figure 2. Superconductivity results of a sample containing synthesis attempt of $Ba_6SrLi_3TiCu_7(SbO_{10})_2$ annealed at 900°C (Figure 2). Test shows no superconducting properties.

Conclusion

Lack of knowledge about superconducting mechanisms makes the realization of novel high temperature superconductors challenging. Generative AI, in combination with experimental data, could advance the discovery of materials hosting specific, advantageous properties. Solid state synthesis was used as an initial method to create predicted materials, and we identified and refined several unknown phases of the resulting samples. Despite not finding any superconducting results, possible new phases have been synthesized based on AI prediction. Continuing to isolate these unknown peaks of interesting compounds to achieve phase pure samples can provide better experimental data to train the ML model and improve predictive power for the advancement of materials science.

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