AI-Assisted Search for High Temperature Cuprate Superconductors

Steven May
School of Chemistry and Biochemistry, Georgia Institute of Technology

Many cuprates have been shown to exhibit superconductivity at relatively high temperatures. Artificial intelligence (AI) has been used to predict the critical temperatures of a variety of superconducting materials, and it is thought that it could be used to predict the structures of novel high temperature superconductors, such as cuprates. The syntheses of AI-predicted cuprate superconductors were carried out via solid-state and molten salt flux (MSF) synthesis methods, and superconductivity was measured using a Magnetic Properties Measurement System (MPMS). The results of these syntheses were fed back into the AI to improve its future predictions.

Introduction

The pursuit of high temperature superconductors is a prominent area of materials research due to their applications in energy transmission and a variety of electronic technologies. Cuprates are a family of superconductors which are of particular interest due to their high critical temperatures, which are believed to arise from their copper oxide layers[1]. It is hypothesized that AI could be employed to accelerate the discovery of high temperature cuprate superconductors. Specifically, AI models can be used to predict novel crystal structures that are thermodynamically plausible and structurally compatible with superconducting behavior. Throughout this summer, solid-state and MSF synthesis methods were used in synthesis attempts of AI-predicted candidate cuprate compounds.

Methods

To synthesize the target cuprate compounds via solid-state reaction, stoichiometric amounts of precursor powders (metal oxides and carbonates) were weighed and thoroughly mixed using a mortar and pestle until homogeneous. The mixed powders were placed in alumina (Al_2O_3) crucibles, then heated in air using furnaces at 800 °C for 12 hours. After cooling, the samples were reground and reheated to 900 °C for 12 hours. Heating and cooling were conducted at a rate of 200 °C/hr. Powder X-ray diffraction (XRD) spectra were collected after each heating step to monitor phase evolution, and MPMS was used to check for superconductivity.

Since carbonates (CO_3^{2-}) typically decompose into carbon dioxide (CO_2) gas and O^{2-} at high temperatures, sealed quartz (SiO_2) tubes were used to target AI-predicted compounds containing carbonates to prevent atmospheric interaction. Stoichiometric amounts of precursor powders were ground thoroughly and placed into crucibles, then the crucibles into quartz tubes. The quartz tubes were first evacuated using a vacuum pump while being flushed with argon to remove residual air. Once evacuated, the tubes were flame-sealed to maintain an inert atmosphere during heating. The sealed tubes were subjected to the same treatment as outlined above for the other solid-state reactions, apart from a slower heating and cooling rate of 33 °C/hr to prevent sudden increases in pressure that could rupture the tubes. The products were characterized as outlined above.

To target $LaCuO_3$, MSF was used because to form the high-valent transition metal ion Cu^{3+} requires highly oxidizing conditions, such as those found in fluxes composed of potassium hydroxide (KOH) and potassium iodide (KI). $LaNiO_3$ and $NdNiO_3$ were chosen to be synthesized as indicators of oxidizing conditions due to their similarity to $LaCuO_3$ and because they are easier to form. Stoichiometric amounts of precursor metal oxide powders were added

to alumina crucibles. Fluxes with KOH:KI ratios of 1:0, 3:1, and 1:1, respectively, were used to fill the crucibles. Samples were heated in air to temperatures of 500 °C and 700 °C, respectively, for 12 hours. Heating and cooling were conducted at a rate of 200 °C/hr. After cooling to room temperature, the solidified products were washed multiple times with deionized water using vacuum filtration to remove residual flux, then dried overnight at ambient temperature. The solid products were characterized using XRD.

Results & Discussion

Solid state synthesis, as outlined in the methods section above, was used to synthesize 26 AI-predicted cuprate compounds, and five of them yielded materials with structures not found in Bruker EVA, the Inorganic Crystal Structure Database (ICSD), or in the literature. After the initial synthesis, the unknown phases were targeted by systematically altering the stoichiometry to probe a multi-dimensional phase space to determining their composition and structure. No new compounds were isolated due to time constraints.

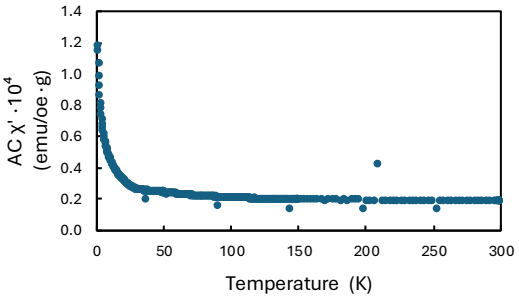

Figure 1. *MPMS of mixed cuprate samples synthesized via solid-state showing lack of superconducting signal.*

MPMS results indicate that superconductivity was not observed in any of the solid-state synthesized samples, which may be due to the unsuccessful formation of the AI-predicted crystal structures. This possibility is supported by the observation that only a small number of unknown phases were identified relative to the total number of compounds targeted. Additionally, given the complexity of the target compositions, often containing up to seven elements, it is plausible that the synthesized unknown phases did not incorporate the full intended stoichiometry, potentially deviating from the predicted structures required for superconductivity.

Tube sealing was used to target the AI-predicted cuprate $Li_2Cu_5CO_8$. At first, stoichiometric amounts of lithium carbonate (Li_2CO_3) and copper(II) oxide (CuO) were placed directly into the quartz tube for reaction. The products of this reaction were Li_2CO_3, CuO, and lithium silicate (Li_2SiO_3), indicating that the only reaction that occurred was between the Li_2CO_3 and the quartz tube. The synthesis was attempted again by first placing the precursor powders into a crucible, then placing the crucible into the quartz tube. XRD analysis revealed that only CuO and Li_2CuO_3 were present, indicating that no reaction occurred. Similar oxycarbonate compounds have been synthesized using flowing carbon dioxide gas at various partial pressures[2-4], but for the scope of my project this summer, that was beyond the capabilities of the McQueen lab. Attempts to synthesize $Li_2Cu_5CO_8$ were abandoned, as we decided to focus on other compounds with the limited time available.

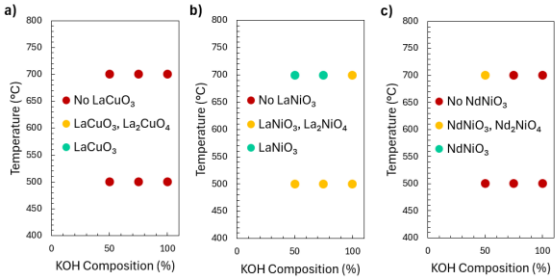

Figure 2. a) *Results of $LaCuO_3$ flux synthesis.* ***b)*** *Results of $LaNiO_3$ flux synthesis.* ***c)*** *Results of $NdNiO_3$ flux synthesis.*

Molten salt flux did not yield $LaCuO_3$, but the results did demonstrate which conditions are more favorable for the formation of RMO_3 (R = rare earth metal, M = Cu, Ni) in general. $LaNiO_3$ was formed on its own at 700 °C with 1:1 and 3:1 KOH:KI fluxes, and alongside La_2NiO_4 (with Ni^{2+} instead of Ni^{3+}) in all other tested conditions. The presence of La_2NiO_4 is an indicator of weaker oxidizing conditions. $NdNiO_3$ was formed alongside Nd_2NiO_4 at 700 °C with a 1:1 KOH:KI flux, and it was not formed on its own under any conditions. These results suggest that higher temperatures and lower KOH:KI ratios lead to conditions more favorable for the formation of RMO_3. The investigation of higher temperatures and

increased KI concentration was not conducted due to time constraints. If a synthesis method that formed $LaCuO_3$ powder was identified, we intended to repeat it with a slower cooling rate to grow crystals.

Conclusions

AI predictions were used to target novel cuprate superconductors, which were synthesized using both solid-state and MSF methods. While no superconductivity was observed in the synthesized products, several previously unreported phases were produced. MSF synthesis, though unsuccessful in yielding the target compound, provided useful insight into the oxidizing capabilities of KOH-KI fluxes. Further work should be conducted to isolate novel compounds and to explore the KOH-KI phase space for optimizing oxidation. The data reported in this study is valuable for refining AI predictions and improving synthetic strategies, paving the way for future discoveries in the search for high temperature cuprate superconductors.

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