Identifying high-Tc transmon candidates with minimal thickness

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Abstract

Qubit coherence times are the largest factor currently limiting quantum supremacy. While candidate materials have been identified and utilized in transmon (superconducting qubits) based circuits, the current coherence time record of 0.3 milliseconds in superconducting Tantalum remains insufficient for most problems. A recent discovery McLellan and Dutta et. al [1] demonstrated that decoherence effects on qubits could be split into three contributors: $1/3^{rd}$ being caused by the (typically Sapphire) substrate, $1/3^{rd}$ being caused by environmental and external effects, and $1/3^{rd}$ being caused by oxide layers grown on the material surface. Based on this information, this project focused on the creation and expansion of metrics that quantify the desirability of a material based on its predicted oxide amounts and predicted critical temperature. The gradient boosting algorithm XG Boost was used for predictions, and the Nelder-Mead optimization algorithm was used to solve the Convex Hull minimization problem posed when calculating energetically favorable oxide amounts.

Introduction

A previous metric for oxide favorability was developed by previous REU student Koppel [2] for all binary metal alloys and most metal oxides. Limitations to this approach included a limited list of oxides, the usage of only the most energetically favorable oxide (instead of multiple oxide layers), and the limited list of materials (1562 binary alloys). This work enhances the oxidation metric by extending the material database to include the Supercon and Materials Project databases (~170000 materials total, inclusive of repeat entries), the oxide database to include all possible oxides based on elemental composition, and solving for the optimal amount of each oxide layer. Predictive models from ASU's Hong Group were used to obtain material and oxide melting temperatures to find the percentage of removable oxide, and a new model based on Hamadeh's work trained to find the critical temperatures of our materials.

Methods

An outline of the table of input dataset entries for calculations performed is below.

Material	Tc (Kelvin)	Oxide Metric	% Meltable Oxide	Formation Energy	Elemental Info.

Oxide	Melting Temp	Formation Energy	Elemental Info.

All information about materials were obtained from Materials Project and the Supercon dataset. For entries with missing formation energies, a machine learning model (See Section A) was used to predict the overall oxide metric.

A. Oxidation Metric – Favorability of a material to form an oxide layer

Each material in our combined Supercon + Materials Project dataset was parsed into its elemental composition. Then, entries in a new dataset were made by including the following:

For Supercon entries only:
Atomic radius, Fusion Heat, Thermal Conductivity, Number of Valence electrons, Tc
Elemental data for all entries:
Atomic number, period, group, Gas, Solid, Liquid, atomic mass, boiling point, density, melting point, electron affinity, Pauling electronegativity, first ionization energy.
*Gas/Solid/Liquid were binary indicators for the state of the material in room temperature.

A database of oxides with information in the table above was also created from all materials containing Oxygen in Materials Project. Then, the following processes were executed.

1. Find list of all potential oxides
   a. Parse the chemical formula of the material and find all oxides whose formulas have elements strictly inclusive to the material's elements
2. Solve Convex Hull Minimization
   In order to find the most energetically favorable combination and amount of oxides, we needed to solve an optimization problem presented as follows. For sake of clarity, let us assume that there are n oxides in the list generated previously, and a total of m elements that compose our material.
   a. Create matrix n by m matrix M from the list of all potential oxides, with each row containing the amount of each element in the parent material.
   b. Create a m by 1 vector C from the parsed formula of the material that contains the amount of each element that is used.
   c. Create a n by 1 vector E that contains the net formation energy of each oxide in order.
   d. Remove repeat oxide entries from dataset and configure initial Convex Hull optimization to remove unnecessary oxides.
   e. Create an n by 1 matrix A by taking the dot product of the transpose of M and C. This provides an initial guess for our optimization process to work with.

At this point, we have the necessary ingredients to solve our minimization problem. By further imposing the conditions of preservation and positivity of mass (Negative amounts of materials were not allowed), a Nelder-Mead optimization could be performed with a max iteration of 100000 steps. Due to the nature of the Nelder-Mead algorithm, strict constraints could not be placed. Instead, heavy conditional functions were designed to prevent the breakage of these constraints and multiplied with the loss function of the algorithm for proper constraints. For the positivity constraint, the sum of all negative coefficients in the iterated A, which we will call X,

was used in the weighting function 1 / 24^X. For the preservation of mass, the cartesian distance between the vector C and the dot product of the transpose of M and the iterated A was set as *Y*. This was then used in the weighting function 1 / 8^Y. This produced generally good constraint fidelity while optimizing the net oxide formation energies. However, many outliers that violated these constraints exist.

These optimized net oxide formation energies were now used in the following equation:

$$\Delta H_{Material} = Q_{Formation\ Energy} \quad \Delta H_{Oxides} = \sum A \cdot E \quad \text{(Net Energy)}$$

$$Metric = 1 - \frac{\Delta H_{Oxides} - \Delta H_{Material}}{\Delta H_{Oxides}}$$

This metric quantifies the energetic favorability for a material to form an oxide. If it has a value below 0, the material is more likely to form an oxide, if it is greater than 0, it is less likely to form an oxide. For materials without a provided material formation energy, this oxidation metric was instead predicted using the elemental composition of each material. The output of these predictions is discussed in the Results section.

B. % Meltable Oxide – Amount of easily removable oxide

Melting heats for all oxides and materials were obtained through ASU's Hong Groups' melting temperature predictor API. Combined with the optimized A vector containing the optimal amounts of each oxide, we could determine which oxides had a predicted melting temperature less than that of their parent material. By multiplying each oxide's predicted amount by their elemental composition, and calculating the total mass of the removable oxide, we could obtain a single % figure by calculating: *Oxide melted mass / Net Oxide mass*

C. Critical Temperature

The critical temperature for materials were only available on the Supercon dataset. As such, predictions for Tc had to be made for the materials from Materials Project. Adapting a predictive XG Boost model [3] to our dataset, it was found that the critical temperature of materials in the SuperCon dataset could be predicted with a RMSE of ~9.4. The settings used for XG Boost were adapted from Hamidieh's approach [3]:

Evaluation Metric: RMSE, Max Depth: 16, Minimum Child Weight: 1, Colsample_bytree: 0.50, Learning rate: 0.02, Number of boosting rounds: 375.

Figure 1: Actual Tc (Kelvin) vs. Predicted Tc (Kelvin) for samples in Supercon

The finalized weights were used to predict critical temperatures for all entries in the Materials Project section of our dataset.
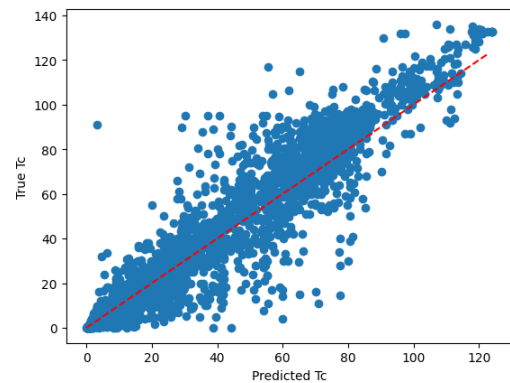
Results

We found that, despite being able to predict the critical temperature of our materials relatively accurately, machine learning models failed to converge when predicting the Oxidation Metric calculated in section A. Instead, convergence was found for the simplified oxidation metric initially proposed in Koppel's work. Graphs of convergence lie below.

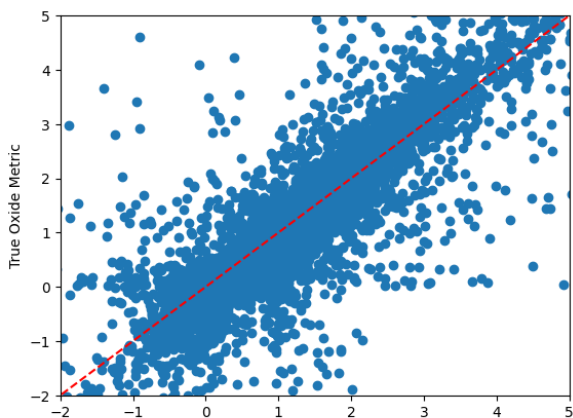The simplified oxidation metric consistently showed an RMSE of <10 across different training

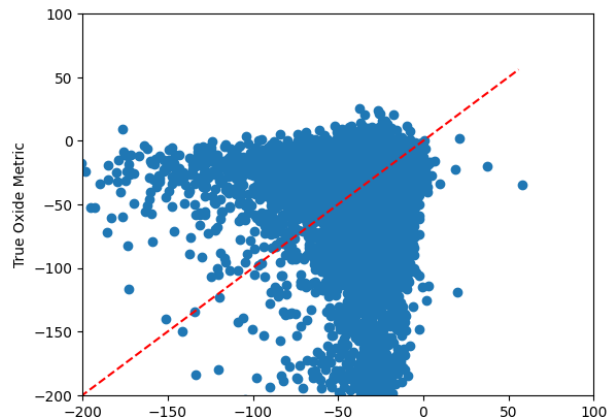*Figure 2: Simplified Oxidation Metric*

Figure 3: Optimized Oxidation Metric

sets, while the optimized oxidation metric had a consistent RMSE of > 40. On a visual basis, it is clear that while some correlation is found in calculation of the optimized oxidation metric, it is a poor correlation indeed. This suggests that the formation of oxides, and their optimal energy configuration, is a complex problem that cannot easily be solved with solely a material's elemental composition information. Despite this failure in predictability, the oxidation metric can still be calculated for new entries through a workaround. Instead of predicting the oxidation metric outright, the formation energy of the material can be predicted (This has been proven to be possible with an RMSE < 9 for our data) and the net oxide formation energy optimized with the process in Methods section A.

Conclusion

This work has developed a collection of metrics to analyze materials on a large scale. By locating materials grouped within a close Cartesian distance to each other (within the context of our three metrics), clusters with similar superconducting properties can be identified. As the materials project is inclusive of many theoretical materials, this provides valuable initial insights into which candidates may be useful for high Tc, high coherence time superconductors. Additional products such as the solution to the Convex Hull problem provide initial estimates for oxide layer thicknesses. It remains an open, important question of why exactly oxide layers promote qubit decoherence. Initial ideas point towards the two-level systems created by dangling electrons in the oxide layers, noise created by rough interfaces, or external microwave / phonon vibrations amplified in between the oxide layers. To approach this goal, simulations are currently underway on Tantalum oxidation.

References

[1] 'Chemical profiles of the oxides on tantalum in state of the art superconducting circuits', McLellan and Dutta et. al., Jan 2023

[2] 'Machine-guided Design of Oxidation Resistant Superconductors for Quantum Information Applications', Koppel et. al.

[3] 'A Data-Driven Statistical Model for Predicting the Critical Temperature of a Superconductor', Hamidieh 2018

Acknowledgements