Stable Structures in AlCuNi and AlNiTi

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(Dated: April 1, 2021)

I. INTRODUCTION

The study of materials is not something new. Throughout the history of mankind, the role discovering new materials has played on the progress of humanity is apparent from the bronze age, all the way to the information age. New technology does not follow far behind new materials, meaning that new materials lead to new technology.

There are many reasons computational techniques are used when trying to find new materials. One reason being that it helps eliminate a lot of the cost required to try and experimentally make new materials. They also allow for the exploration of new materials in ways that have not been done yet. For example, we may have not developed the technology to make a certain material in a lab, but as long as the models are accurate, knowing more about that material may assist in the development of the technology needed to make it.

The systems being investigated are Aluminum-Copper-Nickel and Aluminum-Nickel-Titanium. The motivation for choosing these systems is that finding stable structures in them could lead to the discovery of superalloys. A superalloy is a material that can withstand extreme temperatures and are in high demand in aeronautics.

II. METHODOLOGY

There are two main methods that were used to look for these stable structures. Those methods are, density functional theory and cluster expansion. Density functional theory solves the Schrodinger equation using electron density rather than the position of each electron. This significantly decreases the computation time needed to solve the equation. This is done by transforming it from an equation of $3N$ variables where N is the total number of electrons in the system to an equation of just three variables.

$$\widehat{H}\Psi(r_1, r_2, ..., r_N) = E\Psi(r_1, r_2, ..., r_N)$$

$$\Downarrow$$

$$\widehat{H}\rho(r) = E\rho(r)$$

Density functional theory is implemented using software called Vienna Ab initio Simulation Package (Vasp). VASP lets users calculate different properties of a material including the total energy of the system, which is what we needed.

Cluster expansion is the other technique that is used. This method takes interactions be-

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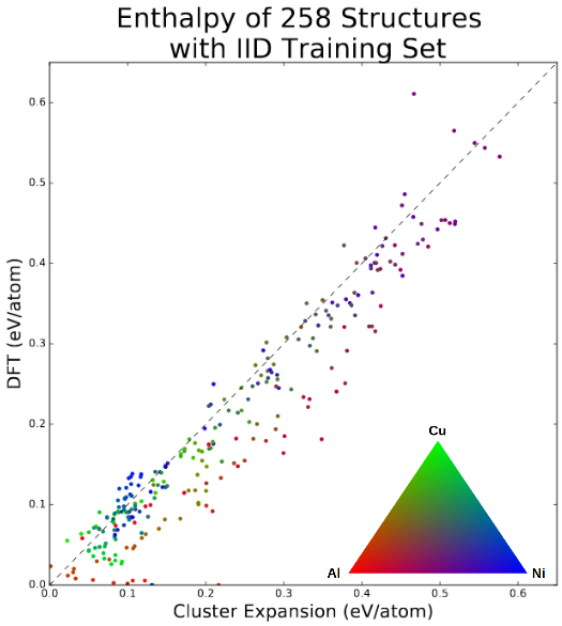
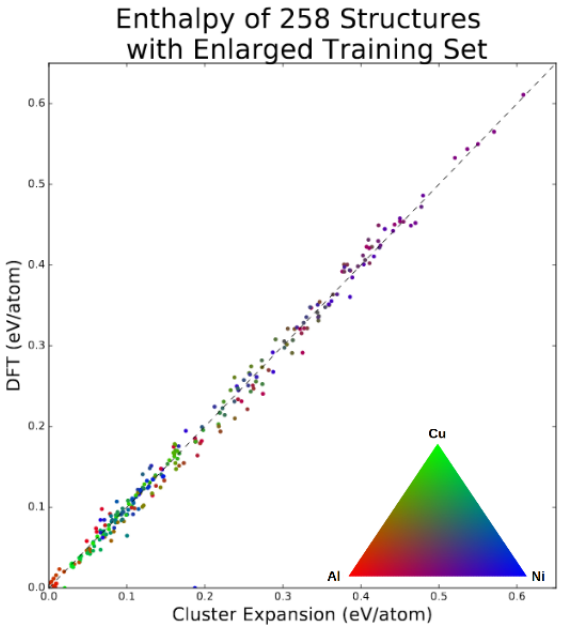
tween groups of atoms and finds the energy that each cluster contributes to the total energy. This is implemented through UNiversal CLuster Expansion (UNCLE).

How exactly do these two pieces of software help to look for new materials? The two systems that are being investigated have approximately 80,000 possible structures with unit cells ranging from one to ten atoms per unit cell. Out of the possible 80,000 structures, 500 independent and identically distributed (i.i.d.) structures were chosen. This means that the chosen structures were picked independently of one another, and the probability of choosing a certain structure does not depend on what was already picked. The total energy of these 500 i.i.d. structures is then found using density functional theory in VASP. Now that information is known about some of the 80,000 structures, a model can be trained, and attempt predict the energy of the remaining structures. This is what UNCLE does, it uses cluster expansion to form a model and predict the energy of all the possible structures.

III. RESULTS

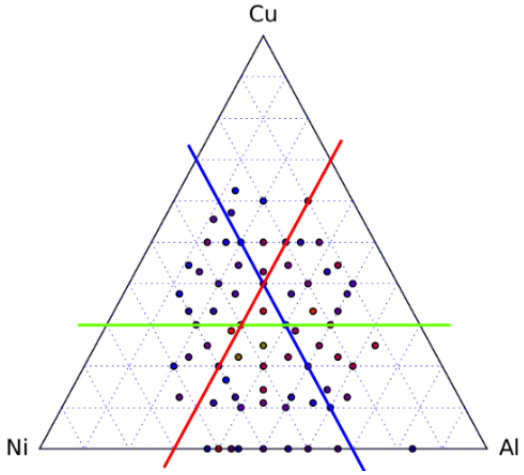
For Aluminum-Copper-Nickel we used 250 of the 500 i.i.d. structures to train the model. Using 250 gave us an average prediction error of $27\frac{meV}{atom}$. This error is considerably high. Applying cluster expansion to ternary systems is something that is still being developed, however it has been used on binary systems for a long time. An error less than around $6\frac{meV}{atom}$ is good for a binary system. Comparing the error we got for AlCuNi to what is acceptable for a binary system is what allows us to say the error is high. There are a few techniques that were used to try improve this high error. Increasing the size of the training set is the first thing that was attempted. Due to time constraints, getting another 500 i.i.d. structures was not possible. Therefor, the lowest energy for each concentration was found using VASP. This set of 258 structures will become our validation set, and will be used to compare results with. The energies found for these 258 structures were then added to the training set. Below are images comparing the formation enthalpy of the validation set with two different training sets of data.



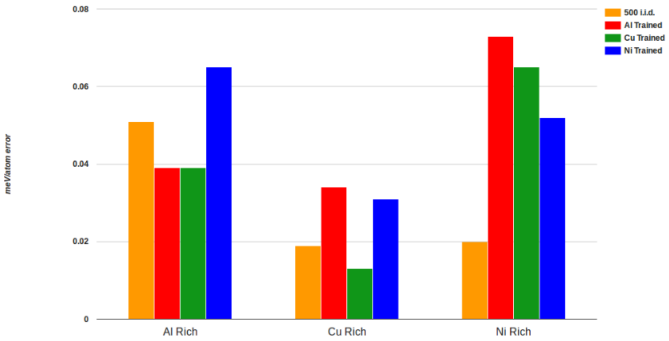


It can be seen that doing this improved the accuracy of the predictions. This is to be expected because we added the validation set to the training data.

The next technique that was attempted in order to improve the errors was partitioning the training data. This was done by separating the original training set by concentration. The original 500 i.i.d.'s were split into three new training sets. Aluminum rich, copper rich, and nickel rich, all which contained every structure that had at least thirty percent of that element respectively. Thirty percent was chosen so that no structures from the original training set would be left out. However, because of this there will be the same structures in multiple of the new training sets. Due to time constraints, the original training set had to be partitioned rather than selecting 500 new i.i.d. structures for each partitioned region. Below is how the data was partitioned.



Everything to the right of the red line is aluminum rich, to the left of the blue is nickel rich, and above the green is copper rich. Each region represents a new set of training data. Training the model on certain element rich structures provided some interesting results. Aluminum and copper behaved as expected but nickel did not. It was expected that if the model is trained on copper then the predictions on copper rich structures should become more accurate. The same expectations can also be applied to aluminum and nickel. As the chart below shows that is not the case.



Two of the three training sets reflected what was predicted. Training on aluminum and cop-

per rich structures improved the predictions for those respective structures. Training on nickel rich data not only made the predictions for aluminum and copper rich structures less accurate, but also the nickel rich structures. This is something that is very interesting and is being further investigated.

IV. CONCLUSION

From this research it has been learned that much improvement can be made to the prediction of ternary allows when using cluster expansion. Possible stable structures were found but due to the high error it can not be said for certain. Applying the techniques that we did allowed us to improve the error but not as much as preferred.

There is still more techniques to be applied to attempt to lower the error of the predicted energies. However the techniques that were applied not only provided more results but also lead to new ideas. The technique where the original training set was partitioned is being expanded on. A new training set of data with 500 i.i.d. structures for each concentration is currently being used to predict the remaining energies. If these predictions behave as expected then a model will be made to use all of the predicted energies and find the most accurate one.