Discovering New Superalloys Using Machine-Learned Interatomic Potentials

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It is more important today than ever before to accelerate the discovery of new revolutionary materials. Advances in computing, transportation, spaceflight, and all other technology sectors are happening faster than ever before, and demand is on track to surpass supply for energy, food, and other resources if big advancements are not made in the next 50 years. New computational methods have been developed to assist in that effort with great success. A Moment Tensor Potential (MTP) is a machine-learned interatomic potential that can predict material properties at a fraction of the cost of density functional theory while maintaining comparable accuracy. This is shown proved useful in creating phase diagrams for ternary alloys. In this work, MTP has been used to identify a new stable intermetallic phase in the Co-Al-W high-temperature superalloy system which may provide improvements in efficiency and performance from jet turbine engines, nuclear powerplants, and find itself useful in other high-temperature industrial uses.

Contents

1 Introduction

The history of mankind is also the history of materials science. The discovery of superior materials has played a key role in the development of construction materials, weapons of war, agricultural tools, and most recently personal computing devices.

The utilization of new materials through new extraction, manufacturing, and distribution processes has had a profound impact on every culture through every age. Just as bronze working was a superior technology to stoneknapping, iron was a superior material to bronze. Civilizations that learned to work iron became the dominating geopolitical forces of the time and were able to project their power and influence due to their advanced military and agricultural technologies.

The most recent technological revolution is commonly referred to as the *Information Age*. The first silicon transistors was manufactured in 1947 and resulted in the 1956 Nobel Prize in Physics.[1] Tens of billions on transistor devices are manufactured every year and are used today in everything from home and mobile electronics, kitchen appliances, cars, and even farming. In just the last six decades, global smartphone usage has reached such high rates that some markets in Western Europe and North America have been saturated.[2] There are few technologies that have had such a profound impact on civilization in such a short amount of time. By enabling access to instant information, these technologies have empowered minorities, women, and the poverty-stricken throughout the world.

The discovery and development of new materials and the technologies born from those innovations has shaped the world we live in, and as more advanced technologies are developed we will need new materials to manufacture them.

Many methods exist to accelerate the discovery of new materials; experimental scientists and engineers have had great success in using the scientific process and iterative experimental designs, building from their knowledge and past results. However, experimental methods are too slow to keep pace with the rapid evolution of microelectronics and space exploration.

Scientists are using a variety of computational methods to simulate material properties long before they need to expend the resources to manufacture it in a lab. One such approach is a first-principles method called density functional theory (DFT) which revolutionized materials science upon its invention in 1964[3,4] DFT allows the simulation of materials while accounting for quantum mechanical effects. However, these calculations are greatly limited in the size, complexity, and time scale of the structures that they are able to evaluate. The complex and often chaotic structures of next-generation materials result in extremely computationally demanding DFT calculations which are impossible to compute on all but the most powerful supercomputers available today. The need for calculations to be completed within a reasonable amount of time means that scientists will often use different simulation methods that trade accuracy for speed. However, molecular dynamics, mesoscale modeling, and finite element analysis are all limited by the narrow time and length scales in which they operate and are unable to treat quantum mechanical effects in their calculations resulting in inherent inaccuracies.

Recent progress in machine learning methods has opened many new doors in computational materials discovery. Later in this text, the concept of a machine-learned interatomic potential trained using data from DFT calculations will be explored, bridging the gap between quantum mechanics and atomistic simulation. These models can achieve quantum-accurate simulations on structures containing tens of thousands of atoms. The aim of this work is to clearly establish the methods used to build a moment tensor potential and demonstrate its utility in accelerating materials discovery.

Taking Flight

One of the greatest achievements in the history of human civilization was the discovery of powered flight. In the time since Wilbur and Orville Wright first took flight in the sprucewood-framed and fabric-lined “Wright Flyer" in 1903, humans broke the sound barrier, created affordable commercial air travel, landed a man on the moon, and this week (at the time of writing) remotely flew a helicopter on Mars.

The invention of efficient turbojet engines revolutionized commercial aviation, and the extreme operating conditions inside the combustion chamber has pushed modern metallurgy and materials science to the absolute limit. Due to microscopic defects in materials in the structure of all metals, extreme forces at high temperatures begin to deform them in a destructive process known as creep. Most metals stretch like taffy

at temperatures far below their melting point which is a death-knell for high RPM turbine blades.

The Sky is Not the Limit – The Metal is

In an effort to prevent creep in turbine blades, new nickel alloys were developed that resist creep at temperatures exceeding 600° C.

The extraordinary high-temperature mechanical strength, oxidation resistance, and creep resistance of nickel superalloys uniquely qualifies them for use in the most stressful conditions inside jet turbines, rocket engines, nuclear steam turbines, and other machines that experience extreme environments. Nearly all superalloys in use today belong to the nickel-base alloy families, such as Inconel, René, Hastelloy, and Waspaloy. Each of these alloys gains its high-temperature strength through the formation of an intermetallic phase called $L1_2$.

An intermetallic compound is a type of metallic alloy with an ordered crystal structure that maintains a well defined stoichiometry; they form when the ordered state is energetically favorable when compared to a disordered solid solution.

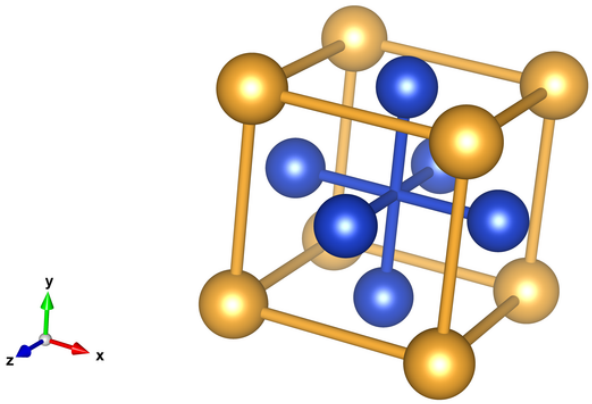

Figure 1: The $L1_2$ crystal structure is in the cubic crystal family and has a composition of A_3B_1. Atom species A lies on the faces of the cube, and species B lies on the corners. This is very similar to the face centered crystal structure.[5]

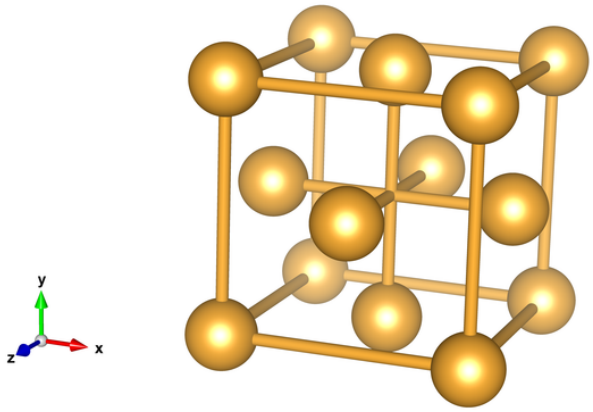

Figure 2: The FCC crystal structure serves as a parent prototype for the $L1_2$ crystal[5]

The primary strengthening mechanism of Ni-based superalloys starts with the formation of an intermetallic $L1_2$-Ni_3Al γ' phase. The $L1_2$ structure is pictured in figure 1. Compare the arrangement of atoms to that of a standard FCC crystal in figure 2.

The placement of a second atomic species on the faces of the crystal allows the $L1_2$ structure to exist within a bulk FCC structure with minimal distortion.

This phase's coherent precipitation from the continuous face-centered cubic γ-Ni matrix lowers the interfacial energy of the system, resulting in cube-like structures throughout the crystal. Coherent boundaries between crystals is an energetically favorable state, as the small mismatch between lattice sizes introduces very little strain. This process lowers the interfacial energy of the structure, which is a function of the difference in surface energy between two phases. A simple example is how water droplets tend to spread out if placed on paper; their spread lowers the total energy between the water and the paper.

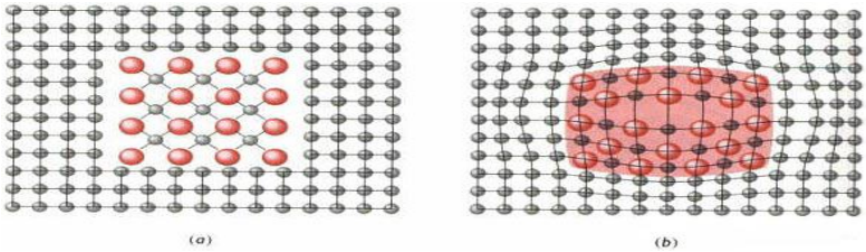

Figure 3: A large mismatch in the sizes between two neighboring crystals will result in a large amount of strain at the interface. Low lattice mismatch results in less strain, and is preferred in precipitation mechanisms in solids.[6]

The formation of the two-phase γ/γ' microstructure (the small microscopic structure of a material visible under an optical microscope) pictured in figure 4 is crucial in determining if an alloy will be a suitable candidate for high-temperature application, and many efforts are being made to improve the thermodynamic stability of these phases.

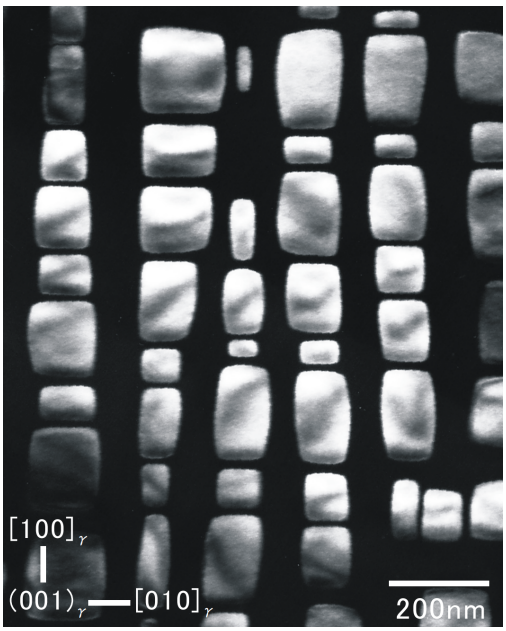

Figure 4: The Figure 1. TEM image of cuboidal γ' precipitate particles aligned along <100> directions in a γ matrix of Ni-8.5Al-5.4Ti alloy aged at 1213 K for 2.7×10^3 sec[7]

A New Superalloy is Born

In 2006, Sato, et. al, discovered a two-phase γ/γ' formation in the Co-Al-W system, morphologically identical to the microstructure found in nickel superalloys.[8] This discovery led to an explosion of research on the topic of cobalt alloys due to their higher melting point and superior oxidation resistance than nickel alloys. This makes them much better suited in high-temperature applications.[9]

As stated before, the formation of the γ' precipitate is the primary strengthening mechanism of Ni-and Co-based superalloys. The formation of an antiphase boundary (a result of the coherent precipitation of the $L1_2$ phase in the FCC matrix) prevents the migration of microscopic defects throughout the material.

Saal and Wolverton, at Northwestern University, determined that the cobalt γ/γ' microstructure was metastable.[10] Wolverton, et al., performed a computational investigation using special-quasirandom structures to approximate the $L1_2$ structure in a disordered γ matrix and found the formation enthalpy of the structure to be 66meV above the convex hull. As shown in figure 5, a convex hull is a way to measure the outermost values of a set of points. In the context of phase diagrams, the convex hull maps a structure's composition and energy. If a structure is on the convex hull (considering only structures with negative total energy), then it is thermodynamically stable. Structures that lie near the convex hull are not thermodynamically stable, however due to slow self-diffusion or other slow kinetic processes, these unstable structures may take hours, days, or even years to fully decompose. We can try to develop an intuitive understanding of what a convex hull is by looking at figures 6 and 7 and identifying which structures are stable Formation enthalpy is a measure of the thermodynamic stability of a chemical compound; crystals with a low formation enthalpy are stable and are possible to observe in nature (though not all stable crystals will be due to kinetic processes that govern their formation).

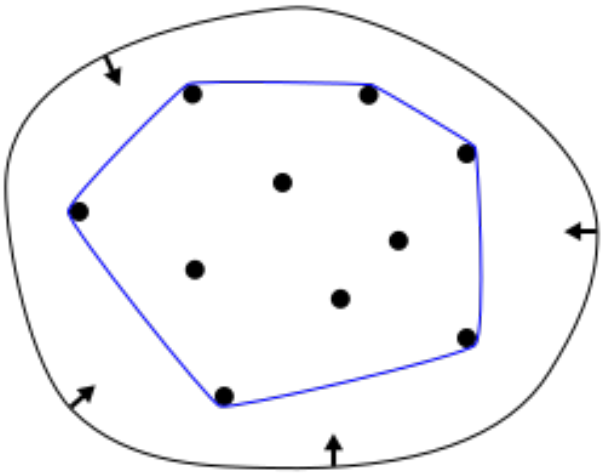

Figure 5: The convex hull algorithm is a geometric model that finds the outermost set of points that enclose all other points in a set with the smallest possible perimeter. An intuitive way to think about the convex hull is how stretching a rubber band across the nails in a geoboard.

It is metastable likely due to the small difference in energy between the the Co_3Al,W_1 γ' phase and the stable phases. This results in a remarkably slow dissolving process.[10]

Similarly, laboratory experiments showed that the γ' phase decomposes rapidly, relative to the required service lifetime for jet engine parts. Annealing treatments at 900° C indicated that the γ' phase decomposes into the γ-Co, $D0_{19}$-AlCo, and B2-W during extended (+1000 hours) periods at high temperature.[11] There is not a conclusive explanation for the decomposition yet, though self-diffusion velocities were high enough that it can reasonably be stated that the decomposition is not limited by the rate of diffusion of the present elements.

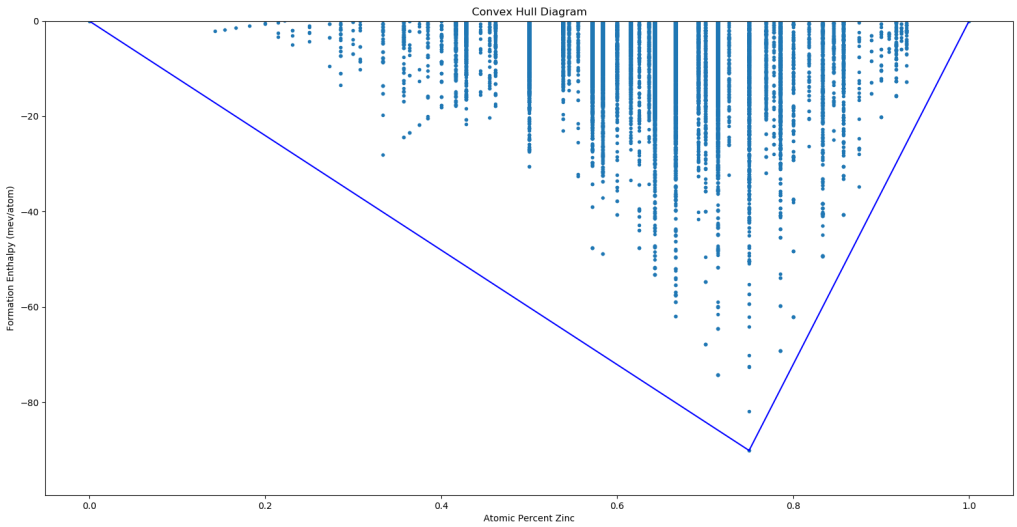

Figure 6: As a demonstration, observe that the convex hull for the Co-Zn binary alloy shows few structures with a low enough formation enthalpy to form stable compounds. Most of the possible arrangements of the atoms result in an unstable configuration and over time will dissociate into the stable structures that lie on convex hull

Accelerating Materials Calculations

As described earlier in the text, density functional theory (DFT) is a widely used computational method in the materials modeling community for its ability to accurately compute the electronic structure of simple chemical systems. Once DFT achieved accuracy high enough for use in quantum chemistry in the 1990s through the use of exchange correlations, computers became a valuable tool in describing the electronic structure of materials.

Simply put, DFT computes an approximate solution to Schodinger's equation, calculating the total energy of a structure, as well as the forces and stresses acting on that structures. Since engineering properties such as elasticity, bulk modulus, as well as thermal and electrical properties can be calculated from DFT results, it has played an important role in accelerating the discovery of new materials; especially relevant today given that silicon transistors are being manufacture on the scale of seven nanometers. Unfortunately, DFT calculations become exponentially more expensive as more and more atoms are added to simulations. This limitation makes true high-thoughput investigations of three, four, or more elements in a single system effectively impossible to complete.

As a potential workaround to the limitations of DFT, machine learning is often heralded as the next big development in the world of computational materials discovery for its potential to predict properties without fully evaluating the electronic structure properties. The Moment Tensor Potentials, developed by Alexander Shapeev, are a class of systematically improvable polynomials fit to the single, binary, and dihedral interactions between atoms in a lattice. Once trained, a MTP is able to predict the properties of a crystal structure with DFT accuracy using several orders of magnitude fewer computational resources. MTP can increase the breadth and speed of our investigations by two or three orders of magnitude by training on a small number of DFT calculations and predict properties for the structures that would be otherwise impossible to evaluate with DFT. By using an active-learning framework to determine the ideal training data for a given system, we can build an interatomic potential using relatively little training data. This potential then can determine equilibrium positions, energies, forces, virials, and stresses of any configuration within that alloy.

Previous work at BYU

In 2017, then graduate student Chandramouli Nyshadham completed a high-throughput computational search for stable γ' phases that would identify new cobalt based ternary superalloy candidates[12] using DFT calculations. This paper discovered six new superalloy candidates – filtered by price, availability, toxicity, among other metrics – which could be superior alternatives to the nickel-based alloys in use today. These simulations were confirmed by metallurgists at Northwestern University that found the predicted γ' precipitates in Co-Ta-V and Co-Nb-V and efforts are being made to stabilize the γ/γ' microstructures in these alloys.[13]

In 2019, then BYU PhD candidate C. Rosenbrock, published the results of using MTP to investigate the Ag-Pd alloy, and developed techniques on how to build a composition-temperature phase diagram from first principles.[14] Many of the methods described in this thesis are derived from this work.

As of the writing of this paper, MTP has been used to analyze several cobalt based ternary alloys including Co-Al-W, Co-Nb-V, Co-Ta-V, Co-Ni-Ti, and Co-Mn-Zn, resulting in the identification of several stable phases not reported in metallurgical databases like AFLOW[15] and ASM Alloy Phase Diagram Database.[16] In the following sections, I enumerate the required steps to train a moment tensor potential, including the creation of the training and testing databases, as well as the use of the Machine-Learned Interatomic Potential (MLIP) software[17] which is critical for the proper setup of a MTP. At the time of writing, there are limits to the capabilities of the public version of MLIP that restrict it to training an MTP on only a single atomic species. The authors of the MLIP package are working hard to bring MLIP-V2 to the public which enables multi-species training.

2 Methodology

This chapter will discuss the process of training a moment tensor potential and testing it against known materials and their properties. The process of selecting training data, evaluating properties from first principles quantum mechanics, and fitting the moment tensor potential to these properties is explained here. Given that the MLIP package handles most of these processes, we will focus on developing a higher-level understanding of the algorithm and how each step can affect the outcome of the potential. The details of how to run each of these steps are given in Appendix 1, and in the repository for AutoMTP hosted on GitLab which contains all of the code necessary to begin building moment tensor potential projects (except for the proprietary MLIP codes, which are core to the project. If you have access to these codes then you're good to go).

Setting up the active-learning framework

Once an alloy system has been selected for investigation, we begin by enumerating a comprehensive list of crystal structures that will serve as the training and testing datasets for our MTP.

First, using a program called Enumlib,[18] enumerate a list of crystal structures containing every derivative superstructure containing four or fewer atoms in the cell. There are several parameters needed to enumerate the list properly, contained in a filed called `struct_enum.in`. An example input file is given in the appendix for a face-centered cubic prototype. All parameters needed are given there, as well as an example input file. Enumerating each derivative superstructure for FCC, BCC, and HCP lattices for a ternary alloy will result in 309 structures, from which the structures using in the training set are selected.

The MLIP program reads each structure in this list and determines which structures hold the most valuable information using a D-optimality algorithm, which put simply, minimizes the determinant of the information matrix formed by the dataset to be relaxed. Understanding the nitty-gritty of the D-optimality algorithm is important if you wish to dive into the mechanics of how the structures are selected; please refer to K. Gubaev's work on using MLIP to train interatomic potentials from 2018.[19] Typically MLIP will select between 30 and 100 structures for the initial training set, though other training set lengths are possible. Once the structures have been selected we move on to using DFT to calculate their physical properties

Generating training data using density functional theory

As mentioned in the introduction of this paper, DFT is used to calculate electronic properties for quantum systems. Properties such as total energy, stresses, and forces are directly calculated for a given crystal system (it has many applications, but our research focuses solely on crystal properties).

When each of the structures has been calculated we feed in each DFT output file into MLIP for the training process.

Training algorithm

The training step fits a complicated polynomial function[17] to the DFT output utilizing the atomic positions, energy, forces, and virial stress. This polynomial is fit to each one-, two-, and three-body (dihedral) interactions within the crystal lattice. Each structure contributes information about the local atomic environment, and MTP learns how to interpolate between the datapoints.

Structure catalog relaxation

Once the training step of the potential is completed, we can use the potential to predict the properties of the structures in the relaxation set. This "relaxation set" is the remainder of the structures from the original database of 309 structures, and MLIP will attempt to perform an ionic relaxation.

MLIP performs a relaxation of the entire enumerated dataset using the recently trained MTP, which returns values for energy, forces, and stress for any given crystal structure. This relaxation process calculates each of the aforementioned properties, and applies a time-evolution to the system. If the ions in a structure are subject to an unbalanced force, its position is adjusted proportionally to its corresponding force vector and atomic mass with a timestep on the order of one half of a femtosecond.

If MLIP determines that the MTP is extrapolating a structures properties beyond a predetermined amount, those structures are added to a new dataset which is used as training data for the next training cycle. The outcome of this relaxation process has two possibilities:

1. The structure in question was fully relaxed and the ions are in their equilibrium positions
2. The structure either failed to converge or the extrapolation grade was too large; resulting in the addition of the structures to the training set and further development of the MTP

3 Results and Analysis

Predictions with a "coarse" potential

Following the MTP framework laid out by Gubaev, et. al,[19] and summarized in the methods section of this report, I began the training process for the Co-Al-W superalloy. The following process is built off the steps given in the introduction.

The initial training data is taken from the list of derivative superstructures of the FCC, BCC, and HCP crystal structures, as explained in the previous section. Using the Enumlib package, enumerate these superstructures for a ternary alloy. This creates a list of integers, base-zero, with three elements, eg. 0/1/2. At this point, no chemical properties are associated with this list, instead, their species is assigned after the enumeration step before they are calculated in DFT.

Each of the nearly 200k structures in the Co-Al-W system (with up to 10 atoms in the unit cell) were enumerated. From this set, we took the 309 structures containing 1-4 atoms per cell to use as a the test set, and began the active learning procedure. We began the procedure by using density functional theory to compute the energies, forces, and stresses for the 309 structures from the test set.

The calculations have been performed using the ab-initio total-energy and molecular-dynamics program VASP (Vienna ab-initio simulation program) developed at the Institut für Materialphysik of the Universität Wien.[20,21] These calculations are static calculations, therefore no ionic or electronic steps were taken. The density of the k-points was set to 1800 per Angstroms^{-3}.

Then, data generated from the static DFT calculations were added to the MTP using the MLIP training function. After training, the MTP attempted to relax the structures into their equilibrium positions using the LBFGS algorithm.

Once the interatomic potential learned how to relax each of the structures in the initial set of 309 crystals, we expanded the list of structures to include larger unit cells. This process was repeated through 18 iterations and when completed, was capable of relaxing all 200k structures without extrapolation.

Formation enthalpy computation

The moment tensor potential predicted the equilibrium positions and total energies for each of the 200k structures enumerated in the ternary system. Hess's law can be used to determine the stability of structures based on a weighted sum of their formation enthalpies.

$$\Delta H_f = E^{\text{total}} - \sum_i^N \frac{n_i}{N} \cdot E_i^{\text{total}}$$

Since the formation enthalpy per atom is a measure for the stability for each configuration at T=0 K we can easily identify candidate crystal structures for new alloy phases.

The training error (defined as the discrepancy between MTP predictions and DFT calculations) is $\sigma =$ 15 meV/atom, which is too high to be confident in many of our predicted stable phases and needed to be refined.

Convex Hull analysis

Using MTP, I generated the ternary phase stability diagram of Co-Al-W as a 0 K isothermal phase diagram, fig 7. The chemical composition of each configuration is represented by its position in the triangle, with the pure elements at each of the vertices. The Z-axis represents the formation enthalpy of the configuration as calculated in the previous section, where the pure elements lie at Z = 0 meV/atom.

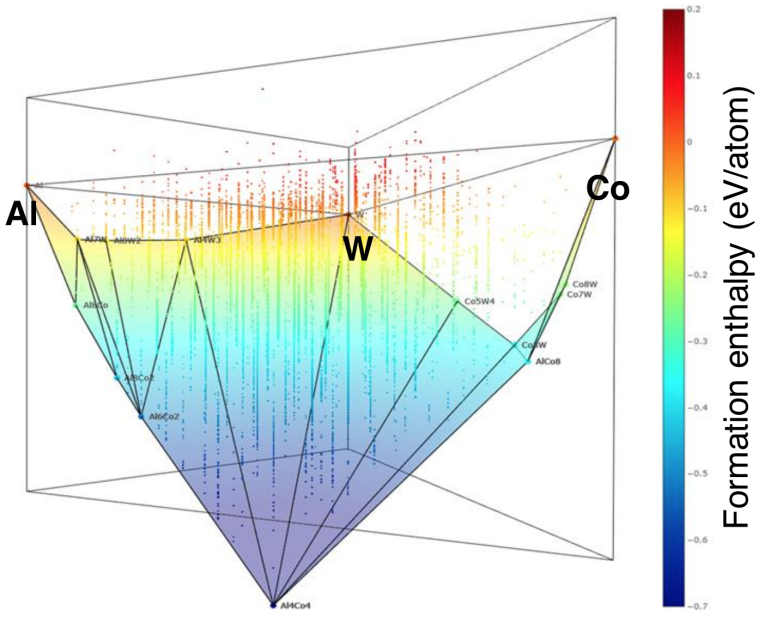

Figure 7: A 3-dimensional view of the 0 K convex hull shows compares the formation enthalpy of each structure, indicating which structure are likely to be stable at higher temperature.

Refining the potential

In an effort to decrease the uncertainty of our total energy predictions from our first interatomic potential, we built a new interatomic potential, training with more parameters in the potential. The training data for this potential would only include structures already suspected to be close to the convex hull, as this would provide the most relevant training data for our purposes.

Choosing new training data

We created a list of every structure within 4σ of the convex hull, where σ is the calculated training error given in the MLIP training output file. We used about 1650 structures as the training data for our model which included each of the structures within 4σ of the original convex hull. This set of structures serves the same purpose as the relaxation set from the first step but theoretically contains all of the information necessary for the potential to learn how to predict the stable phases.

We repeated the training process as detailed above to create a new potential. After 12 iterations, the potential was able to fully relax all the structures found within 4σ of the convex hull of the "fast" potential with a training error of less than 9 meV/atom which increases our confidence in the predictions.

Equilibrium positions were calculated, as well as formation enthalpy, and we found that MTP predicted several new ground states in the alloy system that have not been investigated in the AFLOW database.

New stable structures identified on the convex hull

The two-phase γ/γ' microstructure that imbues high-temperature strength to superalloys can only be observed in a very narrow composition window. We can find the two-phase region in the cobalt-rich corner of the phase diagram as observed by Sato, et al.[8]

A small number of structures containing 8, 12, and 16 atoms were found on the convex hull using the moment tensor potential. In order to be sure that we had found structures that accurately represent the real structures of the alloy, we relaxed each structure using DFT and compared the result to the prediction. These results were all within 2meV/atom, giving high confidence in the model's predictions.

Determining the properties of the new stable structures

Using the refined potential, we have predicted a family of $L1_2$ structures in the Co-rich corner of the convex hull. The depth of these structures is no greater than 2.5 meV/atom. Each of these structures contains cobalt atoms on the face-sites of the cell making up 75% of the total atoms. The lowest-energy structure predicted contained equal parts Al and W on opposite corner sites of the cell. In order to verify that the $L1_2$ structures predicted to exist on the convex hull by MTP were an accurate result, we computed the energies of each structure using density functional theory (VASP). The DFT results corroborate the story told by MTP and led us to find another structure on the convex hull, this time with a composition of $Co_{12}Al_1W_3$ which further lowered the convex hull by 6 meV/atom.

MTP predictions are supported by experimental findings

The identification of a thermodynamically stable ternary phase in Co-Al-W disagrees with the current literature on the subject and appears to clearly classify the γ' structures in the findings of Sato, et al.[8] as unstable, but with a slow kinetic reaction preventing their rapid decay.

Stabilizing γ' with a fourth element

Several including Kobayashi, Lass, and Sato, show that adding small concentrations of tantalum, titanium, hafnium, and other transition metals can increase the stability of the gamma prime structures.

Future work will focus on testing these additions, however initial results from a DFT analysis performed by myself and C. Nyshadham in the summer of 2019 are given below.

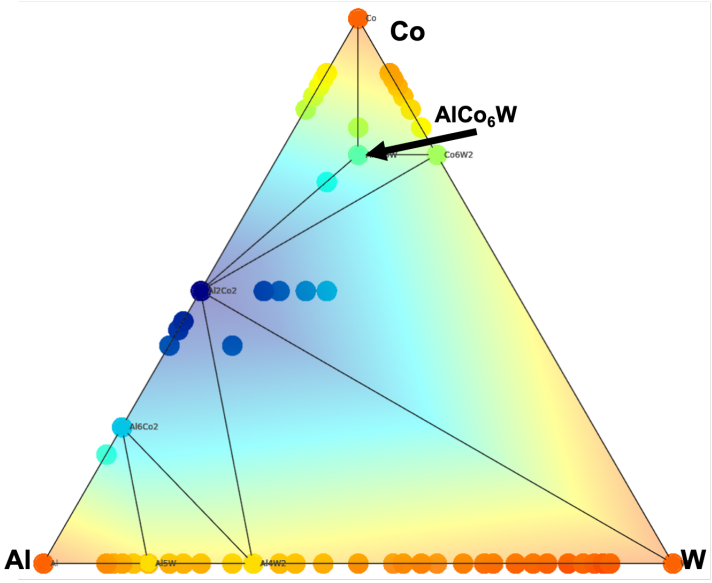

Figure 8: A 2-D projection of the convex hull clearly shows a new stable ternary phase. The phases in this region are derivatives of the $L1_2$ crystal structure which indicates that a γ/γ' region could exist.

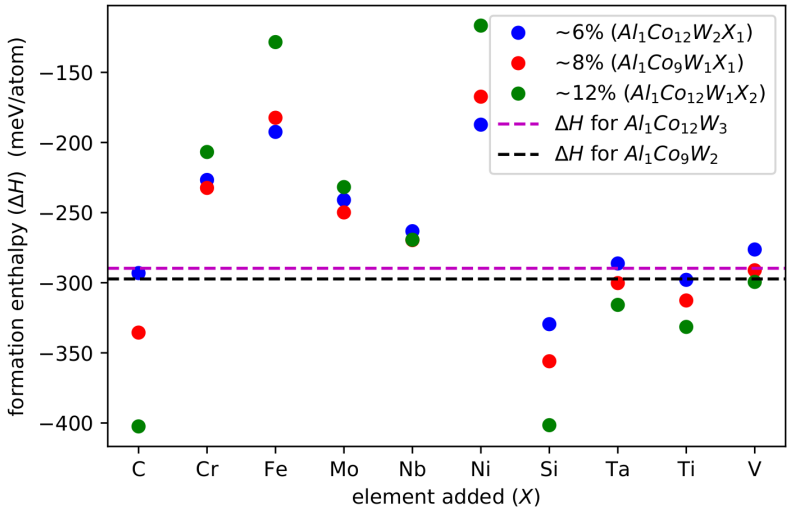

Figure 9: The addition of a quaternary element change the γ' formation enthalpy with respect to the Co-Al-W convex hull

Determining which elements can lower γ' formation enthalpy

In 2019, C. Nyshadham calculated the formation enthalpy of several 4-element systems with respect to the Co-Al-W convex hull and found untested elements could have stabilizing properties. We computed the formation enthalpies of several new families of $L1_2$ structures containing a fourth element addition whose concentration ranged from 6 to 12 atomic percent. Each of these systems were calculated using the structures found on the convex hull in figure **??**_chull2d_chullhe Co-rich section of the diagram. Fourth-element atoms were added in small concentration to various tungsten lattice sites and computed using a fully relaxed DFT calculation using otherwise the same INCAR parameters in VASP as the MTP training data. In figure 9 We see clearly that several new structures have a lower formation enthalpy with respect to the convex hull formed by Co-Al-W. The most promising candidates for further study are the systems containing tantalum, titanium and vanadium, which supports experimental research by Kobayashi, et al.[22] While carbon and silicon show very low formation enthalpy, they tend to diffuse to the grain boundaries in these types of alloys and generally avoided in superalloy metallurgy due to the weakened boundaries.

4 Summary

MTP is a promising candidate in using active learning algorithms to accelerate materials discovery. While still requiring DFT calculations, and subsequently suffering from some of the same bottlenecks as before, MTP is able to accurately predict properties for large structures that are impossible for DFT. Several alloy systems have been investigated using MTP with great success. The ionic relaxation of 200,000 crystal structures with MTP (including the training cycle) required less than 0.1% of the total computational resources as an equivalent effort purely with DFT, opening the doors to analyzing larger and more complicated alloys in the future. MTP will likely prove to useful in molecular dynamics simulations given its high accuracy, however, it is still too slow for simulations involving structure evolution over periods as long as one microsecond. Despite these shortcomings, MTP deepen our understanding of grain boundary properties, dislocation movement, and other chemical and mechanical properties of materials.

Acknowledgments

This work would not have been possible without the generous support from the Department of Physics and Astronomy at Brigham Young University for funding my research throughout the years.

I would also like to thank my research adviser, Dr. Gus Hart, for all the time and attention that he provides to his undergraduates.

I have made a point to try and include revisions for all the feedback that was given to me from the start of the project. Between catching unfinished sentences, grammatical errors, and incomplete ideas, this feedback has been super useful. I especially want to thank Dr. Nielsen for her support and kind words that she's given me during the COVID-19 crisis and helping me through a really difficult adjustment period.

5 Appendices

5.1 Enumerating an FCC crystal using ENUMLIB

```
fcc
bulk
 0.500 0.500 0.00
 0.500 0.00 0.500
 0.00 0.500 0.500
3 -nary case
 1 # number of points in multilattice
 0 0 0 0/1/2  # d-vector
1  5  # starting and ending cell_sizes
1e-07  # finite precision parameter
full  list of labelings
```

- Line 1 is a comment line and should be human-readable.
- Line 2, specifies the period boundary conditions of the following structures and that the following parameters are not a single set of atoms but rather a recurring lattice. Use `bulk` here.
- Lines 3-5 specify the parent lattice vectors, typically these are given either as the normalized lattice vectors or clearly readable values.

A script called `makeStr.py` processes the structures using Vegard's law to estimate the lattice constant for each system after the structures are enumerated.

- Line 6 takes an integer value indicating the number of atomic species to include in the enumeration
- Line 7 indicates the number of atom sites in the lattice. FCC and BCC have one atom, while HCP has two atom sites
- Line 8 is the location of the atom site in lattice coordinates. The following parameter is an explicit list of the atomic species that are allowed to reside on that site (species a labeled with base-0). In our case above, species 0, 1, and 2 all are allowed to lie on the atom site. Any additional atom sites will be listed in the same manner on the next lines.
- After the atom site specifications, Enumlib requires a list of the size of the lattices to enumerate.

It is best to start by enumerating small structures for the initial training sets, typically all the structures between 1 and 5 atoms in the unit cell (as shown above).

- Finite precision parameter. Don't set too large or symmetry will break, but too small of a parameter will result in very long calculation times
- The final part is a list of concentration restrictions. We avoid setting concentration restrictions for training a potential, but if you want to know how to set it up read the documentation for enumlib

  Once the `struct_enum.in` file is set up, run the `enum.x` executable, compiled from the Enumlib package, in the same directory as the input file. It will generate a file called `struct_enum.out` which is later interpreted by the `makeStr.py` script.

5.2 AutoMTP software

My personal project that has been in development since the summer of 2019 during my time at the Ames National Laboratory has been building a wrapper for MTP, in an attempt to make its use a clear and intuitive process.

Please see the repository located on my GitLab page for access to the code and a README that explains the use of the package. Keep in mind that if the full version of MLIP has not been released to the public at the time of reading this paper, you will need to contact Alexander Shapeev about access.

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