**Supplementary Information**

# Files in Folder

all\_data\_noCB.csv –literature dataset file formatted and used for training and testing; carbon and boron compositions were removed. This dataset has 4 repeat entries that were not discovered until after all experiments were done. This original data file is kept to account for how the current model used was created.

final\_TPOT\_model.py – model output from TPOT run after 50 generations

SI TPOT Regression Model.ipynb – jupyter notebook used to create final\_TPOT\_model.py and to plot parity plot

interactive\_visualization.html – UMAP interactive visualization of literature dataset, all simulated compositions, and final chosen compositions.

interactive\_visualization.ipynb – jupyter notebook used to create UMAP plot

Table\_S1.csv – literature dataset with a reference to each entry

Table\_S2.csv – data simulated with Thermo-Calc and ML model for all new compositions

additional\_figures.ipynb – jupyter notebook used to create remaining plots

# UMAP Visualization

To visualize the spread of sampled compositions, we use a dimensionality reduction technique known as Uniform Manifold Approximation and Projection (UMAP) to reduce the space of sampled compositions to two dimensions. A full interactive visualization can be opened from interactive\_visualization.html. While performing this reduction, we scale up the mole fraction of aluminum and nickel by 5x and 10x, respectively, so that the sample compositions and the literature data compositions separate out into discernible clusters. The resultant projection shows all the literature data points used in training and testing of the regression model, as represented by crosses. The color bar represents the log10 of each point. The labeled ovals highlight distinguishing characteristics of each cluster. All the successfully simulated randomly sampled compositions are in the same space. Clusters with high amounts of Al and Ni are distinctly greener in color, which is indicative of lower mass change values compared to alloy clusters with low or no Al and Ni. The set of allows that passed through each screening step is also plotted. The final seven alloys selected are also represented as circles. In the plot, the legend can be clicked to toggle a legend item on or off.

# Pre-Oxidation Microstructure and Thermo-Calc Comparisons

## *LM1* –Al35Cr15Fe30Mn10Nb10

For the alloy LM1, a Thermo-Calc simulation shown in **Figure 1a** predicts that the equilibrium alloy is a two-phase system. This correlates with what is observed in the BSE SEM image in **Figure 1b**. There are two distinct regions: the dark matrix and the brighter particles. The dark matrix regions show some slight inhomogeneity, but it is unclear whether this is due to the need for a longer homogenization treatment. The higher magnification BSE image also more clearly distinguishes some of the oxide inclusions within the larger particles regions.

**Table 1** compares the composition of the Thermo-Calc simulation to the composition measured with EDX. The dark matrix composition resembles the BCC B2 composition since they are both Fe and Al rich with low amounts of Nb. The brighter particle composition resembles the C14 Laves phase since they are both Nb rich.

## LM2 – Al40Co15Cr20Fe25

**Figure 2** shows that LM2 is predicted to be single phase system from ~1350 °C down to just below 1000 °C. The BSE image shows that the homogenized alloy is single phase. The light difference in contrast is not distinguished to be different compositions when analyzed with EDX. The light contrast may be due to another phase or polishing scratches.

## LM3 – Al30Co45Cr10Fe10Ta5

The alloy LM3 is predicted to be a two-phase system in equilibrium at the homogenization temperature of 1200 °C with the phases BCC B2 and Heusler L21. **Figure 3** shows the Thermo-Calc equilibrium simulation along with an BSE image of the homogenized alloy. The grey bulk region looks to be homogenous along with some dark oxide inclusions. The bright particles, rich in Ta, could be a second phase or due to incomplete homogenization or mixing.

**Table 2** compares the predicted phase compositions to the bulk and bright particle compositions. The bulk region is aligned closer to the BCC B2 predicted composition, but the bright particles do not match the Heusler L21 composition. In the Thermo-Calc simulation, the only phase that is Ta- rich is the C14 Laves phase but that phase is predicted to be stable at lower temperatures.

## LM4 – Al25Co35Cr20Mn20

At 1200 °C, the Thermo-Calc simulation in **Figure 4a** shows that the alloy is single phase down to the temperature of 1100 °C. The BSE image taken of the homogenized alloy sample in **Figure 4b** shows that the alloy is single phase. Some dark inclusions are seen in the alloy as well.

## LM5 – Al25Mn25(20)Ni30(33.3)Si5(7)V15

*(numbers in parentheses indicate measurements substantially different from expected)*

**Figure 5a** shows that the LM5 alloy composition is predicted to be a two-phase system down to a temperature of ~1150 °C. This correlates to the BSE image in **Figure 5b** where light and dark phases are observed.

**Table 3** compares the predicted compositions of the phases to the EDX measured compositions of the lighter bulk phase and the dark particles. The measured compositions do not closely match the predicted compositions. The particles are V and Si rich and more closely match the “Cr3SI\_A15” phase, which has a composition of V3Si, but is predicted to be stable at a lower temperature. The bulk phase does not match the “BCC\_B2” phase since most of the V and Si are concentrated in the particles.

## HM1 – Al15Cr15Nb20Ni5Ti45

**Figure 7a** shows that the HM1 alloy is predicted to be a two-phase system down to ~850 °C. The BSE image in **Figure 7b** shows that there is a bulk phase and slightly darker plate-like precipitate phase. Additionally, this alloy also has the highest amount of oxide and nitride inclusions, which are the black particles in the BSE image, of the all the alloys.

**Table 4** shows that the predicted “BCC\_B2” phase composition closely matches the EDX measured bulk composition since they are both close to the nominal composition. The plate composition does not match the C14 Laves composition predicted with Thermo-Calc since only the Al content is similar.

## HM2 – Al35Fe10Mn20(17.5)Nb10V25(27)

*(numbers in parentheses indicate measurements substantially different from expected)*

**Figure 8a** shows that the HM2 alloy is predicted to be a single phase “BCC\_B2” down to 600 °C where the limit of the simulation was taken. This is not reflected with the BSE image in **Figure 8b** since the homogenized alloy has two distinct phases. The inconsistency between the CALPHAD simulation and the experimental observations emphasizes that the composition region that HM2 exists in is not well defined for the database used.

**Table 5** shows the EDX measured composition of the homogenized alloy. The dark matrix phase is V rich while the bright particle phase is Nb rich.

# References for the Literature Dataset

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