

2.1.4 Special quasirandom structures (SQS)

To calculate the energies, enthalpies of formation and elastic properties across the entire binary and ternary composition range, varying compositions of special quasirandom structures (SQS) were used. The SQS are small supercells used to mimic randomly substituted structures in terms of correlation functions. The binary and ternary bcc SQS, used in the present work, were previously generated by Jiang et al. [73, 74]. The relaxation of these SQS structures is complicated because local atomic relaxations can cause the structure to lose the desired lattice symmetry which is far from the original bcc lattice. To preserve symmetry, the calculations were carried out with three different relaxation schemes: 1) the cell volume, cell shape, and ionic positions are simultaneously relaxed, 2) the cell volume and shape are simultaneously relaxed, and 3) only the cell volume is relaxed. The relaxed structure with the lowest energy that preserved the bcc symmetry was used. There are two ways to verify that whether the SQS is still bcc or not after the relaxations. The first is to merge different elements into one element for the SQS structure, and then, use codes available to check the symmetry or space group (such as VASP [62] and phonopy [75]). The second is to visualize the structure directly using a visualization software and compare the symmetry to the unrelaxed bcc structure. For the present work, the relaxed structures were plotted in visualization software using and compared to the unrelaxed structure. After the relaxation, at least five different volume structures were generated and the ions were allowed to relax. This yields the different volumes needed for the EOS fitting described above, which allows a better prediction of the different properties as a function of composition.

2.1.5 High-throughput partition function

This section introduces a theoretic framework to predict the formation of a solid with a mixture of multiple microstates. Our theoretic framework implies that the competition of stable and metastable microstates, results in an increase of entropy as a function of temperature. The increased entropy is what stabilizes the formation of the metastable phases similarly to how entropy stabilizes Ti in the bcc phase. The combined helmholtz energy can be expressed by [76]:

$$F_c = -k_B T \left(\sum \frac{Z_i}{Z_c} \ln Z_i - \sum \frac{Z_i}{Z_c} \ln \frac{Z_i}{Z_c} \right) \quad (2.24)$$

where Z_i represents the partition function a state expressed by:

$$Z_i = e^{-\frac{F_i}{k_B T}} = \sum_k e^{-\frac{E_{ik}}{k_B T}} \quad (2.25)$$

where F_i is the helmholtz energy of state i and E_{ik} is the energy eignevalues of microstate k in the i state. Z_c represents the combined system expressed by:

$$Z_c = e^{-\frac{F_c}{k_B T}} = \sum_j e^{-\frac{E_{cj}}{k_B T}} \quad (2.26)$$

where F_c is the Helmholtz energy of the system and E_{cj} is the energy eigenvalues of microstate j in the combined state, c . Another way to write the combined helmholtz energy, based on these equations, is:

$$F_c = \sum p_i F_i - T S_{SCE} \quad (2.27)$$

where p_i is:

$$p_i = \frac{Z_i}{Z_c} \quad (2.28)$$

S_{SCE} is the state configurational entropy. The S_{SCE} is what stabilizes the phase phase formation and is predicted by:

$$S_{SCE} = -k_B \sum p_i \ln p_i \quad (2.29)$$

This entropy takes into account the statistical competition between the states making the combined helmholz energy more accurate.

2.1.6 First-principles calculation error

The error between the previous results (experimental or calculation) and present results was calculated using:

$$\sqrt{\frac{\sum [(A_{calc} - A_{ref})]^2}{\kappa}} = Difference \quad (2.30)$$