



Integrating atomistic simulations and machine learning to design multi-principal element alloys with superior elastic modulus

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Multi-principal element, high entropy alloys (HEAs) are an emerging class of materials that have found applications across the board. Owing to the multitude of possible candidate alloys, exploration and compositional design of HEAs for targeted applications is challenging since it necessitates a rational approach to identify compositions exhibiting enriched performance. Here, we report an innovative framework that integrates molecular dynamics and machine learning to explore a large chemicalconfigurational space for evaluating elastic modulus of equiatomic and non-equiatomic HEAs along primary crystallographic directions. Vital thermodynamic properties and machine learning features have been incorporated to establish fundamental relationships correlating Young's modulus with Gibbs free energy, valence electron concentration, and atomic size difference. In HEAs, as the number of elements increases, interactions between the elastic modulus values and features become increasingly nested, but tractable as long as non-linearity is accounted. Basic design principles are offered to predict HEAs with enhanced mechanical attributes.

Introduction

Multi-principal element, high entropy alloys (HEAs) are an emerging class of materials defined as containing multiple principal elements in equiatomic or near-equiatomic ratios ranging from 5 to 35 at.% that exhibit wide-ranging applications [1–5]. HEAs are highly stable at elevated temperatures with promising hardness, tensile strength, and corrosion resistance for potential structural applications in extreme environments. Since first being proposed by Yeh et al. in 2004 [6], the research interest in these alloyed systems has garnered significant attention due to their propensity to exhibit enhanced physical properties [7–10]. Expected as a leading contributor towards the successful synthesis of solid-phase solution face-centered cubic (FCC) or body-centered cubic (BCC) crystalline structures is the high configurational entropy. Other factors credited for stability and

enhanced physical properties are enthalpy of mixing, valence electron concentration (VEC), sluggish diffusion, lattice distortion, and cocktail effects [9, 11–15].

Extensive experimental and computational efforts have focused on investigating differing compositions. Yeh et al. first studied FeNiCrCoCuAl_x alloys (where x=0.5%, 1.0%, 2.0%) [6]. They found that the FeNiCrCoCuAl_{0.5} yield strength remained the same from room temperature up to 800 °C. Varying amounts of HEAs have exhibited exceptionally high hardness and resistance to softening at higher temperatures. Cantor et al. attempted to investigate a 16 and 20 element HEAs [16]. They found both alloys to be multiphase and brittle. To their surprise, a single-phase FCC structure rich in Cr, Mn, Fe, Co, and Ni was present within the alloys. Since this discovery, FeNiCrCoMn FCC HEAs and derivatives have received considerable attention [7, 8, 10, 14, 17–19].



Most literature has predominantly focused on the original definition of equiatomic or near-equiatomic alloys. This is due to the idea that a high entropy of mixing term lowers Gibbs free energy:

$$\Delta G_{\rm mix} = \Delta H_{\rm mix} - T \Delta S_{\rm mix},\tag{1}$$

where ΔG_{mix} is Gibbs free energy, ΔH_{mix} is the enthalpy of mixing, ΔS_{mix} is the entropy of mixing, and *T* is the temperature. A minimized ΔG_{mix} leads to the most thermodynamically stable phase. Little is known about the varying atomic weight percent of alloyed systems. This is due to the hypothesis stated previously; entropy drives single-phase stability. By ensuring the HEA is equiatomic, the entropy of mixing is maximized as

$$\Delta S_{\rm mix} = -R \sum_{i=1}^{n} c_i \ln c_i, \qquad (2)$$

where *R* is the ideal gas constant, and c_i is the mole fraction of component *i*.

Failure to generate a single-phase 20 element HEA was reported by Cantor et al. [16], which could be attributed to the high energetic barrier towards the desired atomic randomization resulting in a multiphase formation. However, for HEAs with fewer elements (4 or 5 or 6 elements) such as CoCrFeMnNi, Otto et al. [12] have reported the formation of ideal solid solution. In addition, Sarker et al. [20] predicted high-entropy highhardness metal carbides using entropy descriptors and reported the formation of single-phase solid solution for MoNbTaVWC5. It has also been shown by Rost et al. [21] that configurational entropy can stabilize oxides. Explored by Guo et al., the effects of enthalpy of mixing, atomic size difference, VEC, the entropy of mixing, and electronegativity differences have been reported [13]. Moreover, Guo et al. [13], as well as Zhang et al. [11] concluded that the three main driving factors of solid solution phase stability are the enthalpy of mixing, the entropy of mixing, and the atomic size difference among the atoms. Furthermore, Guo et al. reported that the effect of the VEC term was associated with the determination of whether the alloy would favor an FCC or BCC phase. Sarker et al. predicted high-entropy highhardness metal carbides using entropy descriptors.

Among the various groups that have experimentally explored non-equiatomic HEAs, the criteria for a stable solid solution HEA are more lenient than previously reported. Yao et al. synthesized a novel $Fe_{40}Mn_{27}Ni_{26}Co_5Cr_2$ stable FCC alloy and tested its tensile ductility [22]. In the same year, Tasan et al. synthesized the same HEA and confirmed a stable FCC lattice structure [23]. Tasan et al. reported that the four-component equiatomic CoCrFeMn alloy forms a multiphase material, whereas non-equiatomic $Fe_{37}Mn_{45}Co_9Cr_9$ forms a single FCC phase [23]. Zhang et al. synthesized a stable non-equiatomic alloy consisting of $Fe_{27.5}Ni_{16.5}Co_{10}Al_{2.2}Ta_{0.04}B$, which exemplified

the idea that non-equiatomic HEAs can compete with equiatomic HEAs in terms of mechanical properties [24]. Feng et al. found that short-range order has a significant impact on the magnetic and mechanical properties of HEAs [25]. Short-range order can arise in HEAs if the differences of the binary mixing enthalpies are greater between certain alloying pairs than others. This ordering decreases the entropy of mixing but is offset by an overall reduction in the potential energy of the system, giving rise to improved mechanical properties.

Classical molecular dynamics simulations have emerged as an effective discovery and screening method for studying the underlying mechanisms of atomic-scale deformation as well as strengthening mechanisms for HEAs via solute addition [19, 26, 27]. Micro-structures and stress on the atomic level are essential to study the mechanical behavior of different materials [6]. However, the tradeoff is high computational cost, limited to small systems (typically several thousand atoms) and limitations on the granularity of the principal component composition [8]. Hence, we cannot simulate all possible combinations of alloys as the number of principal components increases.

Machine and deep learning approaches to predict properties can accelerate the screening and discovery process as the models train over the simulations, and can explore a much larger chemical-configurational space than is practically possible using DFT or exhaustive molecular dynamics simulations [7]. Furthermore, the trained models must be robust in prediction with a finite number of samples due to overall computational cost of building the model based on synthetic atomistic simulations. Trained models are expected to uncover correlations between materials-aware features (e.g., configurational entropy, valence electron count, and element type) and target properties, which may not be apparent among numerous alloys [9].

Machine learning [28-33] and materials informatics [34, 35] provide an innovative means of screening for and predicting the properties of HEAs through the use of data-rich computational and experimental sources [36]. Machine learning is a robust method of automating analytical model building and evaluation. Using algorithms that iteratively learn from data allows computers to find insights without being explicitly programmed. Combining materials science with machine learning in the future is actively building on the use of computational and experimental data through quantifiable means of connecting properties through structure-property and performance activity relationships [37-39]. Machine learning has also been used effectively to predict phases of HEAs [40-43]. Several published attempts and works have been made to approach the problems of applying knowledge using various symbolic methods, and later, classification methods such as neural network and perceptrons. Additional integration with integrated computational materials engineering (ICME) and other models now provides an additional means of not only predicting the properties of these exciting



class of alloys but a byway for further exploring and sharing new compositional sets and relationships [44]. In exploring the vast compositional and phase space associated with HEAs sharing the evolving generation of materials descriptors that connect composition, structure, phase, and orientation with micro and macroscale mechanical and thermal phenomenon are of keen interest [45]. A very good review of DFT-based studies and mechanical property predictions of HEAs is offered in a recent review article [46].

The screening for HEAs through computational material science is an established field of materials, with several prior works including the use of ICME, imaging, techniques, and advanced manufacturing [47-49]. Several prior works focused on HEAs are not only scientifically important but also identify unresolved bottlenecks and challenges that suggest the use of repetitive studies of varying composition, structure, and morphology to clarify assumptions made on activity relationships with mechanical properties, including ultimate tensile strength, compression, and elastic or bulk modulus [45]. Unfortunately, finding trends that ultimately connect composition and microstructure demand repetitive experimental and theoretical studies that are often time-consuming, especially given the focus on capturing trends pertaining to mechanical, thermal, and electronic properties. Recent work focusing on combining atomistic simulations with machine learning to design HEAs has also been reported in the literature [50-52]. As listed above, although several state-of-the-art machine learning models have focused on designing HEAs, studies dedicated to understanding the mechanical properties of non-equiatomic HEAs are still scarce.

With high configurational entropy shown as not the only driving metric for single-phase solid solutions of HEAs, and with evidence that stable solid-phase solutions can be synthesized, the opportunity to study the physical properties of nonequiatomic systems presents itself. In this study, to assess the mechanical properties of HEAs, Young's modulus of FeNiCr, FeNiCrCo, and FeNiCrCoCu multicomponent alloys were investigated via a coupled framework of molecular dynamics simulations and machine learning. The Young's modulus of each equiatomic alloy was tested as a baseline metric. From then, each element was taken to be the dominant species in the alloy (i.e., $Fe_{64}Ni_{12}Cr_{12}Co_{12}$), and the testing repeated. This was accomplished along crystallographic directions [100], [110], and [111]. In this work, we establish mechanical property trends of non-equiatomic HEAs in comparison to their equiatomic counterparts. Overall, this work offers fundamental design principles to predict alloys with enhanced equilibrated mechanical attributes.

Results

Atomistic simulations of ternary, quaternary, and quinary alloys

Figure 1 displays characteristic crystal directions and respective nearest neighbor atomic environment along the [100], [110], and [111] directions. Due to altered bonding, HEAs are expected to demonstrate varied response to the applied stress along these three crystal directions. Figure 2 depicts Young's modulus as a function of varying temperatures for various compositions of the ternary (FeNiCr), quaternary (FeNiCrCu), and quinary (FeNiCrCuCo) alloys, respectively. In Fig. 2, a common notation scheme is used. For instance, for ternary alloys, equiatomic alloys indicate that all three elements are 33.33% weight fraction; Fe70NiCr alloys indicate that Fe is 70% and Ni and Cr at 15% each. Similar scheme is used for guaternary and guinary alloys. To facilitate clear comparison in these figures, only extremal values for Young's modulus along [100] and [111] directions are given in the same plot, whereas those along [110] are not included, which fall somewhere in the middle of the pack. However, in Supplementary Information Tables S1, S2, and S3, the numerical values of Young's modulus for each composition



Figure 1: Atomic arrangement in quinary (FeNiCrCuCo) alloys as viewed along crystal directions (a) [100], (b) [110], and (c) [111]. View is normal to the respective direction.





Figure 2: Chart of Young's modulus (GPa) plotted against the temperature (K) for various compositions in the (a) ternary alloys, (b) quaternary alloys, and (c) quinary alloys. Straight lines correspond to the [111] direction, whereas dashed lines correspond to the [100] direction.



corresponding to Fig. 2 are presented, which includes values along the [110] direction.

As compared to deformation along the [100] direction, the Young's modulus and overall stability to temperature change consistently improved in all alloys when deformation was along the [111] direction. For all HEAs except FeNi70Cr, Young's modulus (E) along different crystallographic directions reveal a common trend, i.e., $E_{111} > E_{110} > E_{100}$, which is also observed in reported trends for metals [53], alloys [54], ceramics [55], and aluminum nitride [56]. Although the particular reasons for departure from this observed trend are not obvious, there are some basic effects that could potentially explain this anomaly. In ternary FeNiCr alloys, Ni is the smallest element. In addition, the planar packing density of [110] is the lowest among the three crystallographic directions considered. A combination of these two effects lead to significant relaxation in the FeNi70Cr alloys, which could possibly influence the Young's modulus since similar effect of atomic relaxation on elastic properties has been reported in Ni nanofilms [57].

In ternary alloys [Fig. 2(a)], along the [100] direction, equiatomic and nickel-dominant alloys show somewhat enhanced modulus at lower temperatures, whereas at higher temperatures, high-concentration iron alloys show superior modulus. This is due to the high-concentration iron alloys having the greatest stability of the ternary alloys. High-concentration chromium alloys have a reduced modulus when compared to the other ternary alloys along the [100] direction.

When the deformation was applied in the [111] direction, iron-rich alloys again displayed the greatest modulus, while nickel-rich alloys shifted to having the lowest values. The reduction in rank for nickel is offset by its superb resistance to temperature changes. When comparing the Young's modulus at 100 K to that at 1000 K, the modulus remains relatively unchanged, with a difference of 2.28% from maximum modulus to the lowest modulus. The equiatomic and high-concentration iron alloys both demonstrate an enhancement in stability when compared to deformation along the [100] direction. The equiatomic alloys exhibit an increase in stability of roughly 21% and high-concentration iron alloys demonstrate an increase of 8%. The high-concentration chromium alloys displayed similar performance as the equiatomic alloys from room temperature through roughly 700 K, after which, the modulus decreased at a faster rate than the equiatomic alloys.

In quaternary alloys [Fig. 2(b)], along the [100] direction, the modulus for each alloy decreases with the addition of cobalt into the system. High-concentration iron alloys demonstrate enhanced modulus at lower temperatures, while this enhancement transitions to high-concentration nickel alloys at temperature 300 K and above. High-concentration nickel alloys show the greatest stability of any alloys along the [100] direction, changing 14.9% over the 1000 K temperature range. The iron-dominant quaternary alloys experience a massive reduction in stability, changing by roughly 40% over the same temperature range. Cobalt- and chromium-dominant alloys showed the lowest modulus of quaternary alloys in the [100] direction.

Along the [111] direction, high-concentration nickel alloys reveal superior modulus across the entire temperature range as well as an increase in stability when compared to its [100] deformation (14.9–14.1%). Iron-dominant quaternary alloys along the [111] direction exhibit a massive reduction in modulus when compared to its ternary counterpart, but displayed superior stability, changing only 4.6% when increasing the temperature from 100 to 1000 K. From the ternary nickel-dominant alloy and the quaternary iron dominant system, a trade off in modulus and stability is present along the [111] direction. Cobalt-rich alloys demonstrate a relatively consistent modulus from 100 K through 400 K, but were not stable to elevated temperatures as both moduli depreciated considerably after 500 K.

As shown in Fig. 2(c), addition of copper into the alloys allowed the testing of quinary systems. Along the [100] direction, the high-concentration nickel alloys showed the greatest modulus. It also experienced a reduction in stability (roughly an 8% decrease) when compared to the ternary and quaternary nickel-dominant alloys. Each alloy demonstrated a further decrease in Young's modulus when compared to its respective ternary and quaternary counterpart.

When deformation was applied along the [111] direction, high-concentration nickel alloys exhibited superior stability, changing 3.5% over the tested temperature range. When compared to its quaternary counterpart, the quinary iron-dominant alloys had an increase in stability of 1.1%. The nickel-based alloys again outperformed all other quinary alloys in terms of greatest overall elastic modulus across all temperatures changing only 12.8% along the temperature gradient. The biggest improvement observed was with the high-concentration cobalt alloys, which displayed the second greatest modulus over all temperatures behind only the nickel-dominant alloys. However, its stability was somewhat lackluster (20.1%). Copper-dominant quinary alloys had the lowest modulus across all temperatures as well as a similar stability to the cobalt-dominant alloys (19.1%).

Physical attributes vs Young's modulus

Utilizing enthalpy of mixing, entropy of mixing, atomic size difference (ASD), and valence electron configuration (VEC), the change in elastic modulus and thermal stability can be explained. Using calculated values from Takeuchi et al. for enthalpy of mixing for atomic pairs [58] and the equations outlined by Guo et al. [13], these four values were calculated for the alloys studied. Enthalpy and entropy of mixing are used to satisfy the Gibbs free energy equation outlined in the introduction. Table 1 reports the alloys tested, and their calculated thermodynamic values



as well as reiterating the maximum Young's modulus for each crystallographic directions.

To uncover the trends in results presented in Table 1, we attempted to understand the dependence of Young's modulus on VEC, ASD, and Gibbs free energy. Finding trends concerning E_{100} was first attempted, and as shown in Supplementary Information Fig. S1, an insignificant correlation between ASD, VEC, and Gibbs free energy and Young's modulus was found. When switching from [100] crystallographic direction to [111] in the FCC solid, a reduction in surface energy is anticipated, reducing the overall energy, giving a more stable structure [59]. With a reduction in surface energy, enthalpy and entropy will play a larger role and thus the Gibbs free energy, will have a larger effect on Young's modulus of the alloy. When deformation along the [111] direction was measured, clear trends were observed. VEC and ASD become more actively correlated with the Young's modulus, whereas the correlation with Gibbs free energy greatly increases, which is evident in Fig. 3.

To further test the reduction of external energy contributions and their effects on the Young's modulus, further testing was done at 0.15 K. An increase in the correlation of VEC, ASD, and Gibbs free energy was observed (Supplementary Information Fig. S2). These differing correlations show that the Young's modulus is connected to various physical attributes of the alloys. Each attribute contributes to the overall modulus, wherein the contribution from Gibbs free energy is the greatest, followed by ASD and VEC.

The prominence of Gibbs free energy as a descriptor validates that the stabilities of HEA are primarily governed by the interplay between the enthalpy and entropy of mixing. Figure 4(a) shows the Young's modulus along the [111] face as a function of entropy. An inverse relationship emerges between entropy and the Young's modulus, which contradicts the fundamental idea of entropy being a dominant driving force for stability. When comparing the equiatomic (increased entropy) alloys to their non-equiatomic counterparts, a near average Young's modulus is observed. This demonstrates that the entropy of mixing could offset the Young's modulus if one were interested in utilizing a metal with a lower Young's modulus that offers some other enhanced feature. Figure 4(b) depicts the inverse relationship of the Young's modulus as a function of enthalpy. This graph correlates strongly to the Gibbs free energy trend and shows that the enthalpy of mixing is a prominent factor in determining the stability of multicomponent alloys. Given the discrete nature of the entropies in the systems studied, we took a deeper look into the Young's modulus as a function of enthalpy for the quinary system while holding entropy constant. The results indicate that given similar entropies, the Young's modulus of the quinary alloy is strongly governed by its enthalpy of mixing (Supplementary Information Fig. S3). Overall, these results reveal that enthalpy is a vital thermodynamic quantity that plays a somewhat greater role than entropy in governing the stability of multicomponent alloys considered in this work.

Machine learning

Starting with the prediction of the [111] direction, the set of elementary features are pruned for accuracy and bias through backwards stepwise selection. This results in a total of four selected features: atomic radius (AR), VEC, cohesive energy (CE), and shear modulus (SM). Note that mean AR feature differs from the previously discussed ASD, but describes similar behavior since the ASD is just the ratio of the atomic radius variance and mean. The addition of CE and SM shows how the variance in the Young's modulus is well represented by atomic strain. The automatic tuning of the smoothing functions shows that only CE exhibits a significant non-linear effect with an effective

 TABLE 1:
 VEC, ASD, entropy (J/K), enthalpy (kJ), and maximum

 Young's modulus ($E_{111}, E_{110}, \text{ or } E_{100}$) for each crystallographic direction for all alloys studied.

Composition	VEC	ASD	Entropy (J/K)	Enthalpy (kJ)	<i>E</i> ₁₁₁ (GPa)	E ₁₁₀ (GPa)	<i>E</i> ₁₀₀ (GPa)
FeNiCr	7.92	1.04	9.13	-4.36	350	306	167
Fe ₇₀ NiCr	8	0.25	6.81	- 1.89	385	259	153
FeNi ₇₀ Cr	9.1	0.18	6.81	- 3.87	283	323	173
FeNiCr ₇₀	6.9	0.23	6.81	-3.54	351	271	121
FeNiCrCo	8.25	0.3	11.53	- 3.75	344	287	125
Fe ₆₄ NiCrCo	8.12	0.31	8.72	- 1.86	283	277	155
FeNi ₆₄ CrCo	9.16	0.21	8.72	-3.11	375	308	137
FeNiCr ₆₄ Co	7.08	0.23	8.72	-3.84	343	263	104
FeNiCrCo ₆₄	8.64	0.27	8.72	-2.12	366	265	85
FeNiCrCoCu	8.8	1.03	13.38	3.20	300	256	121
Fe ₆₀ NiCrCoCu	8.6	0.88	10.21	2.60	261	239	131
FeNi ₆₀ CrCoCu	9.4	0.78	10.21	-0.20	350	293	141
FeNiCr ₆₀ CoCu	7.4	0.78	10.21	0.80	321	253	108
FeNiCrCo ₆₀ Cu	8.9	0.74	10.21	1.00	345	255	89
FeNiCrCoCu ₆₀	9.9	1.22	10.21	7.8	234	196	92





Figure 3: The maximum Young's modulus (E₁₁₁) along the temperature gradient as a function of (a) VEC, (b) ASD, and (c) Gibbs free energy.

degrees of freedom of 2.9. The importance of the linear features result in the following equation:

$$y = -19077 + 184AR - 175VEC - 30SM,$$
 (3)

resulting in 97.3% of the variation being explained when combined with the CE non-linearities. For example, for every one increase in the mean atomic radius, the Young's modulus will increase by 184. As such, these relationships provide direct and interpretable prediction of the Young's modulus. When the model is applied to the validation data, the Root Mean Square Error (RMSE) results in a value of 0.19 with an R^2 of 0.96 for the observed vs predicted Young's modulus.

As the GAM model only takes the elemental features into account, rather than the weight fraction and composition, further analysis is performed on the residuals to verify the physics are being captured. First, the residuals are subset by the existence of each element in a sample. An Analysis of Variance (ANOVA) is applied to the subset MSE to reveal that the samples including Fe have a significant reduction in error compared to the other elements. More specifically, the Fe sample MSE is 0.025 compared to the overall MSE of 0.036. Next, each sample is a subset by the total number of elements within the composition. Again, ANOVA is applied to determine if there is a significant difference in errors between composition, but the test fails to reject the null hypothesis. Finally, when an OLS is applied to the residuals where the predictors are the individual element weight fractions, the regression coefficients indicate that the Fe and Cu samples tend to be underpredicted, the Cr and Ni are overpredicted, while Co does not significantly impact the model. This indicates that the overpredicted samples in the observed vs predicted of the validation data shown in Fig. 5 could be potentially due to not capturing all of the Cr and Ni effects. However, with an overall R^2 of 0.05 in the residual OLS model, the GAM model captures most of the variance (97.3%) with respect to the Young's modulus while considering the elemental and compositional influence.

To verify the accuracy of the GAM model across all planes, GAM is compared to several other common machine learning models. More specifically, these models include Multi-layer Perceptron (MLP) with four nodes and the tan*h* activation function, random forest (RF) with 500 trees, and support vector machine (SVM). As reported in Table 2, each respective model is trained using the same original set of features and compared against the ability to predict on the validation data set. As a result, the GAM model outperforms the other models with respect to the RMSE and R^2 of the validation data. The MLP does well in training the limited number of samples, but overfits on the validation data. Furthermore, when examining the residuals within the training





Figure 4: (a) Young's modulus as a function of entropy. The red squares represent the ternary alloys, green triangles represent quaternary alloys, and blue circles represent quinary alloys. (b) Young's modulus as a function of enthalpy.

data set, RF and SVM exhibit poor prediction of the Young's modulus as it deviates from the mean similar to that found in machine learning model development ('Methods' section).

Discussion

One of the most crucial concerns while building a machine learning model is the availability of sufficient quantities of training data that span the entire design space. If the training data are clustered around one or several specific regions of the design space, machine learning might suffer from lower accuracy in other parts of the design space. Selective to specific alloys and compositions, clustered experimental data also represent the current state among the physical metallurgy community. This situation can be overcome by employing atomistic simulations such as molecular dynamics to generate training data [9, 10]. By initially simulating the HEAs with extreme variances in the atomic weight percent for each system using molecular dynamics, it allowed for the machine learning data set to investigate a wide range of alloys. This is a powerful approach for rational materials design since machine learning reduces the time and computational resources needed to test a large number of systems as compared to using molecular dynamics.



Figure 5: The observed vs predicted plot of the validation data Young's modulus colored by the total number of elements in a sample composition. Subfigures (a–d) correspond to binary, ternary, quaternary, and quinary systems, respectively. The overall R^2 for the [111] direction is 0.96 and a validation RMSE of 0.19. In this case, "observed" indicates values obtained using molecular dynamics simulations.

TABLE 2: The comparison of multiple machine learning algorithms in predicting the held- out validation data	Plane	Training RMSE		Validation RMSE		Validation R ²				
		100	110	111	100	110	111	100	110	111
out validation data.	MLP	0.11	0.05	0.07	0.48	0.39	0.49	0.71	0.84	0.75
	RF	0.28	0.20	0.26	0.73	0.45	0.50	0.32	0.79	0.74
	SVM	0.59	0.30	0.01	0.77	0.46	0.44	0.25	0.78	0.80
	GAM	0.50	0.37	0.16	0.47	0.36	0.19	0.72	0.86	0.96

Each property shows moderate correlations towards the overall physical property. This is promising for alloy development because it shows that each metric plays a role in the structure–property relationship of a material allowing for greater refinement when tuning a material to the desired properties. The most correlated relationship is the Gibbs free energy with the Young's modulus. The Young's modulus of the alloys drop when increasing the energy of the system. Increasing the energy of the system imparts more kinetic energy into the individual atoms, increasing equilibrium bond length and reducing the barrier of stress needed to overcome the bond dissociation energies. This causes alloys with an elevated Gibbs free energy contribution to more easily deform to an applied stress.

ASD is the next most correlated metric when concerning the Young's modulus. When lattice distortion was first discussed as playing a major role in enhancing the physical properties of HEAs, ASD was thought to be a leading contributor. Nonetheless, studies have shown that ASD itself is not [10]. ASD only



considers atomic radius and not the desired bond lengths of the nearest neighbor and the next nearest neighbors. Repulsion of atoms can occur if the lattice constant is lower than the expected metallic bond length [60]. This in turn will cause an attraction towards the next nearest neighbor as the lattice constant will be greater than the expected bond length. These electrostatic forces will cause an atomic-level pressure on the atom observed. Though the net force will be zero, the individual stresses further cause distortion to the lattice as the individual elements deviate from their Bravais lattice sites. These local atomic-level pressures are correlated to the charge transfer among the neighboring atoms and thus the electronegativity differences between the adjacent atoms also play a role [61]. The localized stresses will increase the resistance to the motion of the lattice dislocations during deformation. During the elastic deformation regime, the bonding between atoms impacts the Young's modulus. If increasing the ASD increases the local atomic stress, this bonding will be slightly disrupted and the Young's modulus will decrease, supporting the inverse relationships found in Supplementary Information Figs. S1(b) and S2(b).

The role of VEC in phase stability is the determination of whether an FCC, BCC, or mixed structure will dominate in the solid-phase solution. A VEC value greater than or equal to 8.0 favors an FCC lattice structure while a VEC value less than or equal to 6.7 favors BCC. Values in between represent an intermixed lattice structure comprised of both FCC and BCC components [13]. While every alloy tested is above the 6.7 threshold, a few alloys were tested that may have BCC character if not forced to fit a FCC lattice (i.e., high-concentration chromium alloys). Supplementary Information Figure S4 depicts the same trends outlined in the 'Results' section, showing that the inclusion of these lower VEC alloys has no significant impact on the overall results. The VEC contribution towards Young's modulus increases, while the Gibbs free energy and ASD slightly decreases when considering FCC-dominated structures.

Previous work has shown that tuning the VEC can directly effect physical properties of the alloys. Chen et al. explored the effects of decreasing the VEC of an FCC HEA by increasing the concentration of nickel in an AlCoCrFeNi alloy and increasing the VEC in a BCC structure through addition of molybdenum in a CoCrFeNiCu system [62]. They found that decreasing the VEC in an FCC favoring alloy and increasing it in a BCC alloy promotes dual phasic systems that enhanced the compressive fracture strain of the respective materials while exhibiting suitable ductility. Jin et al. designed eutectic HEAs that showed improved stability by tuning VEC and enthalpy of mixing [63]. Li et al. found that the Young's modulus of polycrystalline CrMnFeCoNi HEAs decreases while increasing the VEC [64]. Our results support the findings of Li et al. and provide further evidence of the role of VEC in influencing the Young's modulus of HEAs.

By merging the machine learning approaches with highthroughput molecular dynamics simulations, we have developed a quantitatively accurate model to predict elastic moduli of HEA according to the physical descriptors based on the composition. The generated training set using molecular dynamics simulations homogenously sample elastic properties of a part of the constituent ternary, quaternary, and quinary systems. Explainable machine learning models have offered useful insights into the underlying physics that govern the behavior, which can guide us for adjusting the compositions and features while designing new HEAs with the desired target properties. From the regression coefficients, we can ascertain the most important features which are related to the elastic modulus values-the mean radii, VEC, shear modulus, and cohesive energy. From molecular dynamics simulations, we observe that high weight percentage values of Fe and Ni content lead to higher elastic modulus values which concur with the machine learning efforts.

From the results, we infer that as the number of elements increases, the interactions between the elastic modulus values and features become increasingly nested, but tractable as long as non-linearity is accounted. Hence, we require non-linear models to capture and represent these interactions. In the future, more physical and chemical features can be included to make the models more robust and accurate. The proposed and discussed method of predicting material property can help enable quicker deployment and wider-scale use of trained models in the design of new high-temperature materials.

Conclusions

By varying equiatomic and non-equiatomic FCC HEAs, general trends for future alloy development have been reported. The diverse alloys were minimized to an FCC lattice structure and subject to uniaxial compression. Through the utilization of thermodynamic properties, fundamental relationships were established between the elastic modulus and varying known alloy metrics. It was shown that by measuring the Young's modulus along the [111] crystallographic direction the overall energy of the alloy will reduce allowing one to observe the trends about structure-property relationships. By further reducing external energy contributions via lowering the simulation temperature to near absolute zero, Gibbs free energy, enthalpy, entropy, VEC, and ASD exhibit an inverse relationship with the Young's modulus; an increase in any of these metrics would decrease the Young's modulus. Four features, namely atomic radius, VEC, cohesive energy, and shear modulus play a vital role in predicting elastic modulus of HEAs. These findings further demonstrate that multi-principle element systems garner their interesting properties from a number of different sources. We reveal in general, compositions that exhibit higher Young's modulus. Fe-dominant ternary alloys show superior elastic



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modulus, whereas Ni-dominant quaternary and quinary alloys demonstrate enhanced elastic modulus. By utilizing the connection established in this work between mechanical properties and differing metrics, future alloy development can be more easily tuned to achieve the desired properties.

Methods

Atomistic simulations

Atomistic simulations with 3D periodic boundary conditions (PBC) were conducted within the framework of Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) [65]. The interatomic potentials governing minimization and physical property testing of each structure were based off the embedded-atom method (EAM) [66, 67]. The total energy (E_{tot}) of the system takes the form

$$E_{\text{tot}} = F_{\alpha}(\sum_{j \neq i} \rho_{\beta}(R_{i,j})) + \frac{1}{2} \sum_{j \neq i} \varnothing_{\alpha,\beta}(R_{i,j}), \qquad (4)$$

where *F* represents the embedding energy, ρ_{β} represents the atomic electron density, $\emptyset_{\alpha,\beta}$ is the short-range pair potential interaction and α , and β are the element types of atoms *i* and *j*, respectively. The interatomic potentials implemented in this work to simulate material systems FeNiCr, FeNiCrCo, and FeNi-CrCoCu were taken from Farkas et al. [68]

Surface energy varies with orientation as do other mechanical properties [69]. In cubic materials, the main elastic anisotropy axes are the crystallographic [100], [110], and [111] directions. To establish the structure–property relationships, it was imperative to perform the calculations for each composition along these three directions. Fundamentally, among these three directions, [100] and [111] are where the Young's modulus are at extremal values [70]. As discussed earlier, this trend is also observed in the present work. As a result, while we have computed Young's modulus along these three axes, for brevity and clarity in figures, we focus on data along the [100] and [111] directions.

To study various HEA compositions, large supercells were implemented, which resulted in 4000 atoms, 8000 atoms, and 16,000 atoms in supercells for [100], [110], and [111] oriented structures, respectively. The atoms were randomly placed on an FCC lattice structure with the desired atomic fraction for each element. Each structure was minimized via the same process, wherein energy minimization was performed using a stopping tolerance of 10^{-6} eV for the energy differential between each timestep. The system was annealed, wherein it was brought to 1000 K at a rate of 0.001 K fs⁻¹ and subsequently cooled to 10^{-5} K using the Langevin function with a microcanonical (NVE) ensemble at a rate of 0.0002 K fs⁻¹. Finally, the structure was equilibrated using an isobaric-isothermal (NPT) ensemble,

followed by an NVE ensemble over 1 ps each. Each minimized structure was visually assessed using Visual Molecular Dynamics software prior to additional testing [71].

It is imperative to note that the EAM interatomic potential used in this work was developed to model FCC near-equiatomic mixtures of FeNiCrCoCu [68]. Since we have used this potential to mimic non-equiatomic compositions, all simulated compositions were tested to ensure that they are stable FCC structures as explained later using the radial distribution function (RDF) analysis. In addition, as explained earlier, a robust scheme of annealing was implemented for each composition to test the adequacy of the EAM potential.

We further performed RDF analysis [Fig. 6] to ensure the structural stability and verify if the structures were significantly disordered from the FCC lattice. RDF provides a numerical description of the arrangement of atoms relative to an arbitrary central atom. In crystalline materials, the general shape of an RDF plot can be used to identify the crystal system due to the ordered nature of individual unit cells. An FCC unit cell is more densely packed than a BCC unit cell, producing more peaks of high probability within a certain range of radii. As a result, molecular dynamics simulations can be structurally tracked and verified by comparing the RDF plots of HEAs to standard plots of an FCC element (e.g., nickel) and a BCC element (e.g., iron). Since we have simulated FCC-based HEAs, closer analysis of the widths and heights of the RDF peaks gives an indication of how well the structure compares to an ideal FCC lattice. A narrower peak is a result of a better FCC fit, while a broader peak implies more atoms are displacing from their ideal FCC locations.

The RDF calculations for the ternary, quaternary, and quinary structures show convincing patterns. Perhaps, the most important realization is that none of the generated structures deviate from their FCC behavior at any temperature, ranging from 0 to 1000 K. As shown in Fig. 6, the plots of g(r)—the calculated value of the total RDF at a given radius-versus r all show five distinct peaks within a range of 7 Å. Respective coordination numbers are also given for all cases and elemental RDF is shown for one of the cases. Analysis of a pure Ni and a pure Fe shows that five peaks are representative of an FCC structure, whereas three peaks imply a BCC lattice. However, an increase in temperature is accompanied by a decrease in peak sharpness and the occurrence of overlap in adjacent probability peaks. While at 0 K [Fig. 6(a, c)], the space between peaks has a zero percent chance of being occupied by an atom, the outer peaks lack this forbidden region at higher temperatures. Additionally, the graphs at 800 K [Fig. 6(b)] and 400 K [Fig. 6(d)] display significantly shorter and broader peaks than those at 0 K of the same composition, a trend explained by the tendency for atoms to vibrate as their internal energy increases and phonon loses coherence with temperature. As a particle vibrates more vigorously, there is





Figure 6: Total RDF and coordination number plots for (a) Cr-dominant tertiary HEA at 0 K, (b) Ni-dominant quaternary alloy at 800 K, (c) Cu-dominant quinary alloy at 0 K, and (d) elemental RDF and coordination number for Fe-dominant quinary alloy at 400 K.

a lower chance it will be found in the ideal FCC position. In general, as evident in Fig. 6, the RDF plots show that the various equiatomic and non-equiatomic compositions used in the present work are stable FCC structures. When comparing different compositions to each other, the same trends take shape at each temperature. For each of the three categories of compounds-ternary, quaternary, and quinary-the Ni-dominant structures exhibit the narrowest RDF peaks, indicative of a very stable FCC structure. In contrast, the Fe-dominant structures as well as the equiatomic alloys (for example, equiatomic FeNiCr) consistently have the broadest and shortest peaks, by a significant margin. One exception to this trend is the Cu-dominant guinary alloy, wherein RDF plot shows abnormally short peaks. In general, the RDF analysis indicates that Ni-dominant alloys are most stable in an FCC arrangement, followed by Co, Cr, Fe, and Cu.

Each alloy was fully minimized, and later subjected to a compressive Young's modulus simulation at temperatures ranging from 100 to 1000 K. Thermal effects were mitigated using the NPT ensemble for each structure. The simulation box was deformed along the *x*-axis at an engineering strain rate of 0.001% fs⁻¹, and the change of pressure (stress) was monitored in order to simulate the elastic deformation range of the alloy. From the change in pressure and change in the size of the simulation box, Young's modulus was calculated from the stress vs. strain curve according to the classical mechanics' definition:

$$E = \frac{\sigma}{\varepsilon},\tag{5}$$

where σ is the stress due to the deformation, and ε is the strain present in the system. An example for calculation of Young's modulus using the stress vs. strain curve is given in Supplementary Information Fig. S5. Final results reported herein were obtained by using three different initial configurations for each data point have discrepancies, which are shown by the use of error bars.

Machine learning and design of experiments Data set

We consider alloys that belong to the $Fe_xNi_yCr_zCo_uCu_w$ systems, where the weight percentage values of each element specified by x, y, z, u, and w are constrained by x + y + z + u + w = 100% at 5% weight increments. The compositions span each ternary, quaternary, and quinary system resulting in a total of 877 samples. The weighted compositions were incorporated as initial, starting randomly arranged cell structures on a per-atom basis. A rigorous assessment was performed to ensure randomization was truly random, and sensitivity analysis was considered. The randomization seed value was altered for every single atomic structure, which ensured that the starting configurations were indeed



different. This aspect of randomization was also visualized for several structures, wherein the variation in starting configurations was evident as well as the random placement of atoms on the FCC lattice was verified. Importantly, the presence of error bars in the results is evidence that the starting configurations generated via this process were dissimilar. In addition, elemental coordination number analysis [Fig. 6(d)] was also performed for all alloys, which reveal that the atomic fractions are aptly accommodated. Pressure displacement values are converted to stress–strain based on their elastic modulus values, described in detail by Plimpton et al. [72]. Results of the simulations which were performed at room temperature (300 K) were considered for building the machine learning models.

Machine learning model

An important goal of implementing machine learning is to predict the Young's modulus with a minimal number of training samples while using data that can be collected with little effort. As such, the features used in building the predictor matrix will be collected from summary statistics or transformations of elemental properties based on the weight fraction. More specifically, Wen et al. [39] introduced a 20 feature set for predicting hardness values of HEAs, 11 of which described the formation of different phases (i.e., phase parameters), and the other nine were related to the mechanical properties (i.e., mechanical parameters). Note that the features generated will use many of the same elemental properties but transformed in a different manner. As such, the features must be reduced in the final machine learning model to account for any multicollinearity. The Young's modulus is the response variable that will be predicted and the only material property that will be calculated from molecular dynamics.

To ensure the robustness of the machine learning model to predict multiple compositions, a random subset 90% of the total number of synthetic samples are used as a training data set while the remaining samples are held-out as a validation. Each feature in both sets of data are normalized by their respective mean and standard deviation from the training data set. This ensures there



Figure 7: Plot of the residuals fitting Young's modulus using an ordinary least squares model over all simulated compositions. Subfigures (a–d) correspond to binary, ternary, quaternary, and quinary systems, respectively. The residuals clearly show a non-linear trend within the data.



is no information in the validation data set that could influence the training data. Choice of the machine learning model must be able to account for potential non-linear behavior in the Young's modulus. This negates many of the standard machine learning approaches due to the assumption of linearity. For example, in Fig. 7, when Ordinary Least Squares (OLS) regression is applied to the prediction of the [111] direction Young's modulus, there are clear signs of non-linearity.

Furthermore, the model must be interpretable, i.e., a welldefined relationship to the Young's modulus may be understood with a small subset of features. Due to these constraints, the algorithm chosen in this application is the Generalized Additive Model (GAM) [73]. The GAM is an extension of the OLS model, but includes non-linear effects, i.e.,

$$\boldsymbol{E} = \sum_{i=1}^{n} s_i(X_i),\tag{6}$$

where E is the Young's modulus, s_i is a smoothing spline, and X_i is a feature. In this paper, each smoothing spline is optimized using a combination penalized smoothing regression and the restricted maximum likelihood [74]. The interpretability of the model is addressed through the combination of the linear regression coefficients, the statistical significance of a spline smoother, and the selection of the features through backward stepwise selection [75]. Note that this process does require interaction with the user to finalize the model, but ensures interpretable results.

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Data availability

Majority of the data generated and analyzed in this work are included in this published article and associated Supplementary Information. Additional data and scripts that support the findings of this work are available from the corresponding author upon reasonable request. The machine learning code developed for this work will be released in Supplementary Information pending review by the U.S. Department of Energy.

Declarations

Conflict of interest The authors declare no conflict of interest.

Supplementary Information

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