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Application of Machine Learning in Exploration of Uncharted Territory of Multi-Principal Element Alloys

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Abstract:- Multi-principal element alloys (MPEAs) present an enormous (~ 10^8 distinct types of compositions), largely uncharted compositional landscape, of which only a small fraction has been discovered to date. The critical task for materials scientists and metallurgists is to identify potential compositions with tailored properties for specific application while minimizing the reliance on laborious and energy-intensive experiments. In this study, we developed a robust machine learning framework utilizing nature-inspired optimization method to navigate potential MPEA candidates. Cuckoo Search Optimization (CSO), a metaheuristic algorithm based on the brood parasitism behavior of cuckoo birds, was utilized to generate novel MPEAs with targeted mechanical properties (yield strength, ultimate tensile strength and elongation). A dataset of 700 instances, sourced from experimental literature on MPEAs, was utilized. CSO explored the search space to generate novel MPEAs with good combinations of mechanical properties (yield strength (YS), ultimate tensile strength (UTS) and elongation ($\boldsymbol{\varepsilon}$), by iteratively replacing inferior solutions with better ones, converging towards optimal or near-optimal solutions. Pareto front solutions were then identified using Pareto dominance, ensuring that no individual objective among the multi-objective criteria (YS, UTS and $\boldsymbol{\varepsilon}$) could be improved without compromising another. Thus, this research leverages the potential of machine learning in accelerating the discovery of high-performance MPEAs, paving the way for future innovations in materials design and engineering.

Keywords*: Cuckoo Search Optimization, Machine learning, Mechanical properties, Multi-Principal Element Alloys.*

1. Introduction

Recently emerged new strategy of developing alloys that involves mixing of multiple principal elements in equimolar or near-equimolar proportion to form multi-principal element alloys (MPEAs) also known as high-entropy alloys (HEAs) or complex concentrated alloys (CCAs) has revolutionized the field of materials science. MPEAs present a promising frontier in addressing real-world challenges, offering exceptional properties and performance compared to conventional alloys along with its vast unearthed compositional space. Since the inception of the MPEAs concept, numerous compositions with exceptional properties have been discovered and developed. Notable examples include CoCrFeMnNi [1], Al_x(CrFeCoNiCu) with varying Al concentration (x=0 to 3) [2], NbMoTaW [3], $Nb_{40}Ti_{25}Al_{15}V_{10}Ta_5Hf_3W_2$ [4], MoNbTaVW [5], HfNbTaZr [5], $Re_{0.1}Hf_{0.25}NbTaW_{0.4}$ [6] $Al_{10.3}Co_{17}Cr_{7.5}Fe_9Ni_{48.6}Ti_{5.8}Ta_{0.6}Mo_{0.8}W_{0.4}$ [7], $Al_{10.2}Co_{16.9}Cr_{7.4}Fe_{8.9}Ni_{47.9}Ti_{5.8}Mo_{0.9}Nb_{1.2}W_{0.4}C_{0.4}$ [8], $CoCrFeNiTa_{0.4}$ [9] and $CrFeCoNiNb_{0.5}$ [10]. Despite these advances, only a minuscule fraction of the potential compositions within the vast compositional space (approximately 10^8 distinct types of compositions) have been discovered so far. The conventional trial-and-error approach of discovering and developing new compositions is

both laborious and time-intensive. In response to these challenges, machine learning (ML) has emerged as a powerful tool for navigating the uncharted territories of MPEAs.

Several studies have adopted various machine learning approach in predicting mechanical properties of HEAs. For instance, Bundela et al. [11] utilized various regression models, including linear regression, lasso, ridge, random forest, XGBoost, and bagging regressor, to predict the microhardness of HEAs. Bhandari et al. [12] employed a random forest regressor to predict the yield strength of MoNbTaTiW and HfMoNbTaTiZr across different temperatures. Furthermore, Zhou et al. [13] developed three regressor models to predict Young's modulus and hardness of novel high entropy ceramics by integrating data from previous density functional theory (DFT) calculations and experimental results. Wen et al. [14] advanced a property-oriented materials design strategy by combining machine learning with design of experiments (DOE). Their approach successfully identified alloys with high hardness within the Al-Co-Cr-Cu-Fe-Ni HEA systems, resulting in the discovery of 35 alloys with hardness values exceeding the best in their training dataset. Moreover, Poonia et al. [15] employed differential evolution, a metaheuristic optimization technique, to optimize the composition of an alloy with chosen set of elements, aiming to maximize the hardness of the former.

While numerous studies have focused on discovering novel compositions by targeting individual properties, such as hardness or yield strength, a significant gap remains in the literature regarding the simultaneous optimization of multiple mechanical properties. This study aims to address this gap by employing machine learning techniques to explore the vast, uncharted territory of MPEAs with the objective of identifying compositions that offer an optimal combination of key mechanical properties: yield strength (YS), ultimate tensile strength (UTS), and elongation ($\mathcal{E}$). To explore the vast compositional space, the Cuckoo Search Optimization (CSO) algorithm was employed, while the Non-Dominated Sorting Genetic Algorithm II (NSGA-II) was used to navigate Pareto optimal compositions that offer the optimal combination of mechanical properties.

2. Methodoly

2.1 Data collection

A comprehensive dataset comprising 700 instances of multi-principal element alloys (MPEAs) was meticulously curated from experimental literature. This dataset encompasses detailed compositional and mechanical property data, with the compositional information spanning 13 elements: Al, Co, Cr, Cu, Fe, Ni, Mn, Ti, V, C, Nb, Zr, and Mo. The mechanical properties recorded in the dataset include Yield Strength (YS), Ultimate Tensile Strength (UTS), and elongation ($\mathcal{E}$).

To identify the best alloy composition that exhibits the most favorable combination of mechanical properties within this dataset, we employed the Non-Dominated Sorting Genetic Algorithm II (NSGA-II). A detailed discussion about NSGA-II has been provided in the subsequent section. Through this analysis, the composition $AlCoCrFeNiZr_{0.008}$, characterized by YS, UTS, and elongation values of 1572 MPa, 3517 MPa, and 29.7%, respectively, was identified as the best composition within the database. The results are visually represented in a 3D plot, with YS, UTS, and elongation depicted on the three axes, providing a clear illustration of the relationship between these critical mechanical properties within the database, as shown in Fig. 1. The best combination of mechanical properties is distinctly highlighted within a red square.

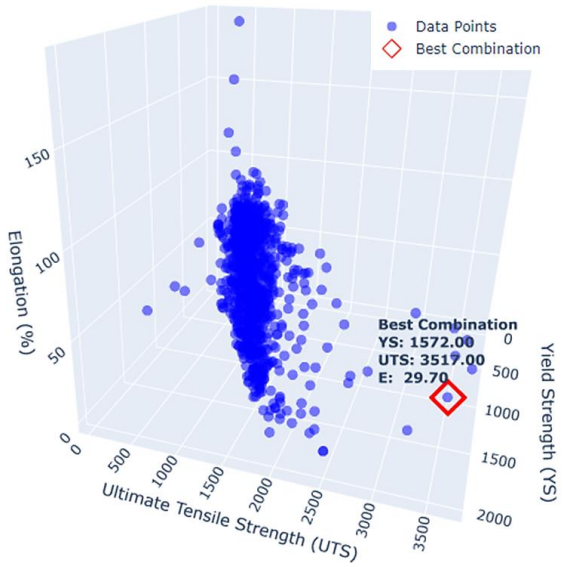

Fig. 1. 3D plot illustrating the distribution of Yield Strength (YS), Ultimate Tensile Strength (UTS), and elongation (%) values for each data point in the dataset. The data point encircled within a red square represents the composition identified as having the optimal combination of these mechanical properties, as determined by the NSGA-II.

2.2 Regression Model training and testing

To predict the mechanical properties of MPEAs, we employed several ML algorithms, including Neural Network (NN), Random Forest Regressor (RFR), Extra Trees Regressor (ETR), CatBoost Regressor (CBR), and XGBoost Regressor (XGB). The dataset was divided into an 80:20 ratio for training and testing the models. Standard scaling was applied to normalize the data. The performance of these models is depicted in Fig. 2. Among all the regression models, the CatBoost Regressor (CBR) demonstrated superior performance, achieving the highest R^2 score of 0.91, along with the lowest Mean Squared Error (MSE) of 0.07 and Mean Absolute Error (MAE) of 0.16. Due to its exceptional performance, CBR was selected for further optimization of the alloy compositions.

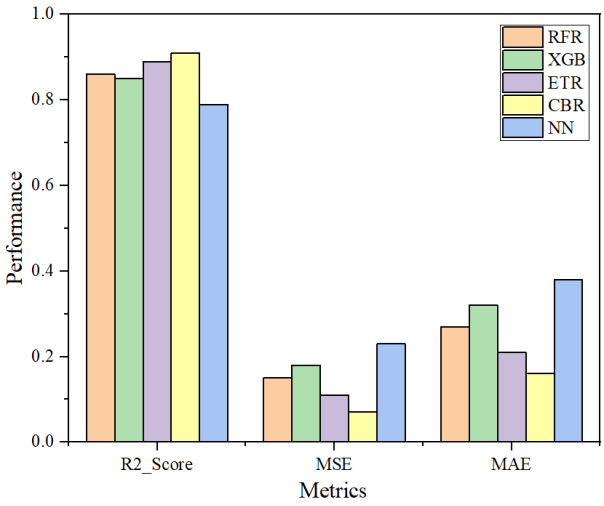

Fig. 2. Performance (R2_score, Mean Squared Error, Mean Absolute Error) of each algorithm.

2.3 Exploration and navigation of novel alloys

Our objective is to discover new alloy compositions that not only match but potentially surpass the mechanical properties of the benchmark composition $AlCoCrFeNiZr_{0.008}$ identified within the database. To achieve this goal,

we implemented Cuckoo Search Optimization (CSO) within the CBR model, integrated into a multi-output regressor framework. CSO is based on the brood parasitism behavior of cuckoo birds, which strategically lay their eggs in the nests of other host birds. If the host bird detects the presence of alien eggs, it may either discard the foreign eggs or abandon the nest to build a new one. This natural behavior forms the basis of the CSO algorithm, which adeptly balances exploration and exploitation to solve complex optimization problems. The detailed workflow of the CSO process is illustrated in Fig. 3.

CSO operates by mimicking Lévy flights, a random walk characterized by a series of long jumps, to explore the search space extensively. Simultaneously, it replaces the worst solutions with potentially better ones, thereby refining the solution set. This dual approach enables the CSO to navigate the expansive search space of MPEAs efficiently. The compositional space was defined across 13 dimensions, with each dimension representing a distinct elemental component. To efficiently explore this high-dimensional space, we configured key parameters within the CSO algorithm, including n (population size), α (step size of the Lévy flights), β (Lévy flight distribution), Pa (discovery rate of alien eggs), number of iterations and search space boundary. The choice of CSO is particularly advantageous in this context due to its robustness and effectiveness in handling high-dimensional and nonlinear optimization problems.

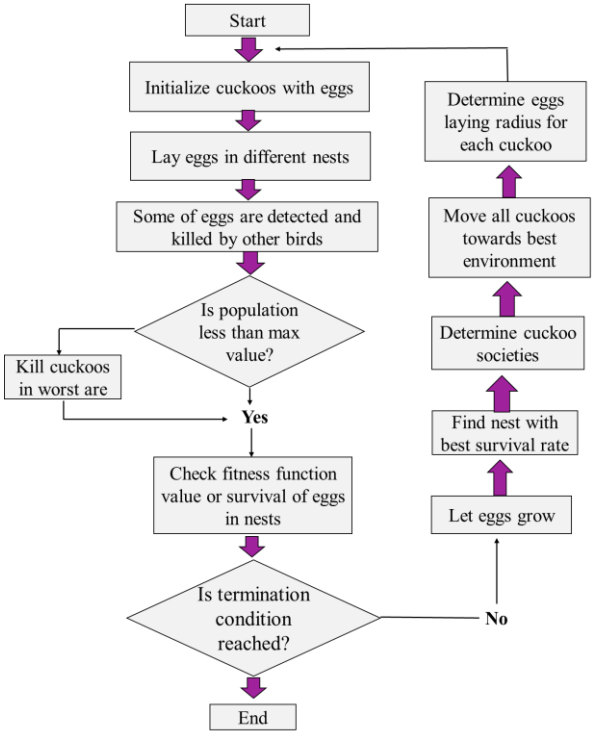

Fig. 3. Workflow of cuckoo search optimization.

To navigate alloy compositions that offer the optimal combination of mechanical properties, we employed the Non-Dominated Sorting Genetic Algorithm (NSGA-II), a highly regarded technique in the field of multi-objective optimization. The NSGA-II is particularly effective for problems where multiple objectives must be optimized simultaneously, and it is widely used due to its ability to identify a diverse set of Pareto-optimal solutions. In the context of multi-objective optimization, the term "non-dominated" refers to solutions that are Pareto-optimal, meaning that no other solution in the search space is superior when all objectives are considered together. These solutions form what is known as the Pareto front, representing the best trade-offs between competing objectives.

3. Result and discussion

In an effort to discover new MPEA compositions that could potentially surpass the benchmark composition ($AlCoCrFeNiZr_{0.008}$), we employed the Cuckoo Search Optimization (CSO) algorithm, incorporating Pareto

dominance within our multi-objective optimization framework to identify non-dominated solutions. A list of novel compositions representing these non-dominated solutions is presented in Table I.

Among the newly generated compositions, $Al_{0.84675}Co_{0.84675}Cr_1Fe_{0.9544}Ni_{0.84675}Zr_{0.00677}$ exhibits mechanical properties that closely match those of the benchmark composition in the database, ($AlCoCrFeNiZr_{0.008}$). Furthermore, two compositions—MoNbTiVZr and MoNbTiV0.25Zr—were identified that surpass the benchmark, demonstrating mechanical properties of [YS = 1748 MPa, UTS = 3800 MPa, ε = 23%] and [YS = 1740 MPa, UTS = 3876 MPa, ε = 27%], respectively. Notably, these compositions have been previously reported in the literature but were not included in our original database. Their experimentally derived YS, UTS, and elongation values are highlighted in red. A comparison of the predicted mechanical properties with the reported experimental values reveals a strong correlation, thereby affirming the reliability of our optimization process. Consequently, the mechanical properties predicted by our methodology exhibit close alignment with experimental results, as validated by the compositions [MoNbTiVZr and MoNbTiV0.25Zr], underscoring the accuracy and effectiveness of our approach.

Table: I Novel MPEA compositions optimized using CSO and Pareto front solutions.

S. No.	Alloy Composition	YS (MPa)	UTS (MPa)	ε (%)
1.	$Al_{0.7262}Co_{0.7785}Cr_{0.8063}Fe_{0.7027}Ni_1Mo_{0.0934}$	1209.976	3629.1116	19.9018
2.	$Al_{0.8225}Cr_1Ni_{0.8225}Zr_{0.4528}$	1229.609	3254.284	29.3906
3.	$AlCr_{0.9583}Fe_{0.675}Ni_{0.9264}Zr_{0.0058}$	1130.77	3158.11	25.85
4.	$Al_{0.84675}Co_{0.84675}Cr_1Fe_{0.9544}Ni_{0.84675}Zr_{0.00677}$	1572	3506.71	29.7
5.	$Al_1Cr_1Cu_{0.00937}Ni_{0.9409}Ti_{0.1028}V_{0.3879}$	1150.3729	2889.0830	27.8307
6.	$Al_1Cr_{0.556}Fe_{0.556}Ni_{0.5215}Ti_{0.2384}$	1226.918	2126.552	23.589
7.	$Al_{0.8247}Fe_{0.9693}Ni_1Zr_{0.0074}$	1017.25	3507.13	28.87
8.	★ **$Mo_1Nb_1Ti_1V_1Zr_1$ [16]**	**1748 (1786)**	**3800 [3828]**	**23 [26]**
9.	$Al_{0.9756}Cr_1Fe_{0.8236}Ni_1Zr_{0.00747}$	1110.755	3821.219	29.175
10.	$Al_{0.9328}Co_1Cr_{0.9827}Fe_{0.8769}Zr_{0.0062}$	1043.51	3487.12	29.0895
11.	$Al_{0.9256}Cr_{0.752}Cu_{0.0987}Fe_{0.4703}Ni_1Mn_{0.0567}Ti_{0.1488}Nb_{0.1302}Zr_{0.0071}Mo_{0.0567}$	1087.4859	2693.45	17.719
12.	$Al_{0.9039}Co_1Cr_{0.9363}Fe_{0.9624}Zr_{0.00693}$	1655.31	3378.25	21.688
13.	★ **$Mo_1Nb_1Ti_1V0.25Zr_1$ [17]**	**1740 [1776]**	**3876 [3893]**	**27 [30]**
14.	$Al_1Cr_1Fe_{0.7356}Ni_1$	1146.430	3400.346	26.564
15.	$Al_1Co_{0.587}Cr_{0.18028}Ni_{0.767}Mn_{0.1734}V_{0.1802}C_{0.01469}$	1290.797	2580.7718	22.9337

4. Conclusion

This study successfully employed advanced machine learning techniques, including the CatBoost Regressor (CBR) and Cuckoo Search Optimization (CSO), to predict and optimize the mechanical properties of multi-principal element alloys (MPEAs). Among the 700 alloy compositions analyzed, $AlCoCrFeNiZr_{0.008}$ was identified as the optimal composition within the database, offering the best combination of yield strength, ultimate tensile strength, and elongation. Subsequently, we employed CSO, in conjunction with the Non-Dominated Sorting Genetic Algorithm II (NSGA-II) to navigate the vast compositional space defined across 13 dimensions to find Pareto-optimal solutions. This approach led to the discovery of various novel compositions, including $Al_{0.84675}Co_{0.84675}Cr_1Fe_{0.9544}Ni_{0.84675}Zr_{0.00677}$, which closely matched the mechanical properties of the benchmark alloy ($AlCoCrFeNiZr_{0.008}$). Moreover, the compositions MoNbTiVZr and MoNbTiV0.25Zr were found to surpass the benchmark in key mechanical properties, with predicted values in strong agreement with those reported in the literature. This research underscores the potential of integrating machine learning models with metaheuristic optimization techniques to navigate the vast compositional space of MPEAs, providing a reliable method for

discovering alloys with superior mechanical properties. These findings not only demonstrate the effectiveness of our approach but also lay the groundwork for future experimental validation.

References

[1] Cantor, B., et al., *Microstructural development in equiatomic multicomponent alloys.* Materials Science and Engineering: A, 2004. **375**: p. 213-218.
[2] Yeh, J.W., et al., *Nanostructured high-entropy alloys with multiple principal elements: novel alloy design concepts and outcomes.* Advanced engineering materials, 2004. **6**(5): p. 299-303.
[3] Han, Z., et al., *Microstructures and mechanical properties of TixNbMoTaW refractory high-entropy alloys.* Materials Science and Engineering: A, 2018. **712**: p. 380-385.
[4] Pang, J., et al., *A ductile Nb40Ti25Al15V10Ta5Hf3W2 refractory high entropy alloy with high specific strength for high-temperature applications.* Materials Science and Engineering: A, 2022. **831**: p. 142290.
[5] Dixit, S., et al., *Refractory High-Entropy Alloy Coatings for High-Temperature Aerospace and Energy Applications.* Journal of Thermal Spray Technology, 2022. **31**(4): p. 1021-1031.
[6] He, H., et al., *Carbide-reinforced Re0. 1Hf0. 25NbTaW0. 4 refractory high-entropy alloy with excellent room and elevated temperature mechanical properties.* International Journal of Refractory Metals and Hard Materials, 2023. **116**: p. 106349.
[7] Tsao, T.K., et al., *On the superior high temperature hardness of precipitation strengthened high entropy Ni-based alloys.* Advanced Engineering Materials, 2017. **19**(1): p. 1600475.
[8] Tsao, T.-K., et al., *The high temperature tensile and creep behaviors of high entropy superalloy.* Scientific reports, 2017. **7**(1): p. 12658.
[9] Jiang, H., et al., *Effects of Ta addition on the microstructures and mechanical properties of CoCrFeNi high entropy alloy.* Materials Chemistry and Physics, 2018. **210**: p. 43-48.
[10] He, F., et al., *Designing eutectic high entropy alloys of CoCrFeNiNbx.* Journal of Alloys and Compounds, 2016. **656**: p. 284-289.
[11] Bundela, A.S. and M. Rahul, *Machine learning-enabled framework for the prediction of mechanical properties in new high entropy alloys.* Journal of Alloys and Compounds, 2022. **908**: p. 164578.
[12] Bhandari, U., et al., *Yield strength prediction of high-entropy alloys using machine learning.* Materials Today Communications, 2021. **26**: p. 101871.
[13] Zhou, Q., et al., *Machine learning-assisted mechanical property prediction and descriptor-property correlation analysis of high-entropy ceramics.* Ceramics International, 2023. **49**(4): p. 5760-5769.
[14] Wen, C., et al., *Machine learning assisted design of high entropy alloys with desired property.* Acta Materialia, 2019. **170**: p. 109-117.
[15] Poonia, A., M. Kishor, and K.P.R. Ayyagari, *Designing of high entropy alloys with high hardness: a metaheuristic approach.* Scientific Reports, 2024. **14**(1): p. 7692.
[16] Wu, Y., et al., *Phase composition and solid solution strengthening effect in TiZrNbMoV high-entropy alloys.* Materials & Design, 2015. **83**: p. 651-660.
[17] Zhang, Y., X. Yang, and P. Liaw, *Alloy design and properties optimization of high-entropy alloys.* Jom, 2012. **64**: p. 830-838.