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Electronegativity Difference and Hardness in High-entropy Alloys

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Abstract: The vast range of possible combinations of High-entropy alloys (HEAs) requires a reliable prediction process to minimize the cost with experiments. Therefore, we perform an investigation of the correlation between the electronegativity difference and the hardness of thirty HEAs. Their composition and hardness were obtained from the literature, the electronegativity of each element was extracted from the periodic table, and the electronegativity difference was calculated in DIAMOY 1.0 software. The results were expressed in terms of a plot involving both variables, and the coefficient of correlation. The moderate correlation coefficient and the behavior of the dispersion of the points in the plot of both variables showed that hardness depends moderately on the electronegativity difference.

Keywords: Electronegativity difference; Hardness; High-entropy alloys.

Diferença de Eletronegatividade e Dureza em Ligas de Alta Entropia

Resumo: A vasta gama de combinações possíveis de ligas de alta entropia (HEAs) exige um processo de previsão confiável para minimizar o custo dos experimentos. Portanto, uma investigação da correlação entre a diferença de eletronegatividade e a dureza de trinta HEAs foi realizada. Sua composição e dureza foram obtidas da literatura, a eletronegatividade de cada elemento foi extraída da tabela periódica e a diferença de eletronegatividade foi calculada no software Diamoy 1.0. Os resultados foram expressos em um gráfico envolvendo ambas as variáveis e o coeficiente de correlação. O coeficiente de correlação moderado e o comportamento da dispersão dos pontos no gráfico de ambas as variáveis mostraram que a dureza depende moderadamente da diferença de eletronegatividade.

Palavras-chave: Diferença de eletronegatividade; Dureza; Ligas de alta entropia.

Introduction

The elements that compose the material, their quantity, the manufacturing route selected, etc., straightly affect the mechanical properties of the materials. In the context of metallic materials, pure metals present very strict utilization due to their limited range of properties. In view of this, the conventional alloys (with two main elements) expanded the range of desired properties. In this direction, posteriorly, the multicomponent alloys, combining three

or more metals and non-metals, partially fulfilled the gap left by the conventional alloys (limited range of properties) [1]. However, the main disadvantage is relatively low knowledge about their properties due to the vast design space compared to the fact that this research field is yet in the beginning. The major categories of multicomponent alloys refer to their configurational entropy, which can be: (a) low-entropy alloys (LEAs); (b) medium-entropy alloys (MEAs); and (c) high-entropy alloys (HEAs) [1].

Objective

The main objective of this work is to obtain the relationship between the electronegativity difference and the hardness of high-entropy alloys.

Material and Methods

The methodology follows the 4-step workflow addressed in the following paragraphs.

(a) First step: refers to the selection of available data from the literature: the hardness values of 30 distinct HEAs were extracted from the references [2-3].

(b) Second step: performs calculation of electronegativity difference ($\Delta\chi$), in percent, which is the standard deviation of the electronegativities related to the average electronegativity (see Equation 1). The nomenclature addresses f_i, molar fraction of the i -th alloy element; $\chi_i$is the electronegativity of the$i$ -th element;$\chi_m$is the mean of the electronegativities of the alloying elements. This parameter impacts the phase separation and new phase formation [4]. As far as this research could reach, in general, intermetallic compounds can be minimized for relatively small values of this parameter [4]. The calculation is performed in Diamoy 1.0 software [5].

$$\Delta\chi = \sqrt{\sum_{i=1}^{n} f_i \ (\chi_i - \chi_m)^2} \qquad (1)$$

(c) Third step: verification and analysis of a possible mathematical relation between electronegativity difference and hardness: establishment of the behavior of electronegativity difference (in percent) and hardness (in Vickers).

(d) Fourth step: if there is a mathematical relationship between both electronegativity difference and hardness, then there is the formulation of an equation between both parameters. The possible equation intends to help in the prediction of hardness as a function of the electronegativity difference.

Table 1 shows the 30 HEAs selected to compose the database of this paper, their codification in this work, and their corresponding Vickers hardness values. They are codified in the alphabetical order of the elements.

Table 1. HEAs by code, composition, and their hardnesses [2-3].

Code	HEA	Hardness (HV)	Code	HEA	Hardness (HV)
1	AlCoCrCuFe	407	16	CoCrFeMnNi	130.2
2	AlCoCrCuFeNi	472	17	CoCrFeMnNiV	636
3	AlCoCrFeMnNi	684	18	CoCuFeMnNi	208
4	AlCoCrFeNi	484	19	CoFeMoNiV	625
5	AlCoCuFeNi	536	20	CoCuFeMnNi	296
6	AlCoCuFeNiTi	626	21	CrCuFeMoNi	263
7	AlCoCuFeNiZr	472	22	CrMoNbTaVW	1072
8	AlCrCuFeNi	495	23	CrNbTiVZr	482
9	AlCrFeMoNbTaTiVW	838.2	24	CuFeNiTiVZr	590
10	AlCrFeMoNi	905	25	FeMoNiTiVZr	740
11	AlCuFeNiTi	516	26	HfMoNbTaTiZr	505
12	AlFeNiTiVZr	800	27	HfMoTaTiZr	542
13	AlHfNbTaTiZr	441	28	HfNbTaTiZr	420
14	AlMoNbTiV	537	29	MoNbTaTiW	507.5
15	CoCrCuFeNi	286	30	MoNbTaVW	536

Results

The plot of the 30 points addressing the HEAs listed in Table 1 is shown in blue dots in Figure 1, while the line of best fit is in red.

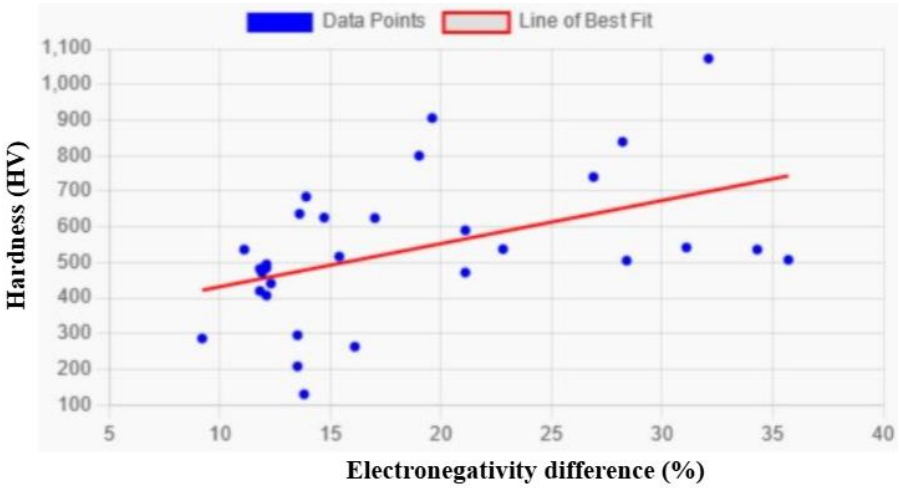


Figure 1. Vickers hardness values in the domain of the electronegativity difference.

In the universe of these 30 HEAs, although the behavior of the dispersion of the points by itself already denotes that the plot does not allow the formulation of a reliable linear equation to calculate hardness as a function of electronegativity difference, a linear regression calculation proposes Equation 2, in which H is the hardness in Vickers. This drawback refers to the moderate correlation coefficient obtained, which is 0.47. Therefore, based on the selected data, the moderated interdependence between hardness and electronegativity difference can be addressed.

$$H(\Delta\chi) = 12.11\,\Delta\chi + 310.50 \quad (2)$$

Discussion

The calculated results of electronegativity difference were obtained for the 30 HEAs collected from the literature, showing that the correlation between both parameters was not low enough to be discarded, although not sufficiently high to be reliable. The corresponding values can be found in Table 2.

Table 2. Hardness and electronegativity difference for the 30 HEAs selected in this paper.

Code	Electronegativity difference (%)	Hardness (HV)	Code	Electronegativity difference (%)	Hardness (HV)
1	12.1	407.00	16	13.8	130.2
2	11.9	472.00	17	13.6	636.0
3	13.9	684.00	18	13.5	208.0
4	12.1	484.00	19	17.0	625.0
5	11.1	536.00	20	13.5	296.0
6	14.7	626.00	21	16.1	263.0

7	21.1	472.00	22	21.1	590.0
8	12.1	495.00	23	32.1	1072.0
9	28.2	838.2	24	11.8	482.0
10	19.6	905.00	25	26.9	740.0
11	15.4	516.00	26	28.4	505.0
12	19.0	800.00	27	31.1	542.0
13	12.3	441.00	28	11.8	420.0
14	22.8	537.00	29	35.7	507.5
15	9.2	286.00	30	34.3	536.0

For example, HEA 1 has lower hardness than HEA 2, even dotted with higher electronegativity difference. However, HEA 3 has hardness higher than those of HEAs 1 and 2. According to Figure 1, the reliability of the results and the trustworthiness of the model increase for values of electronegativity difference at around 12%. Conversely, the sparsity of the data related to the line of best fit starts to increase from values of 14% of electronegativity difference. For values lower than 12% and higher than 33%, scarcity of data influences the results of the prediction. Therefore, it is necessary to add more data to these ranges of electronegativity differences.

Conclusions

In this paper, we investigated the relationship between Vickers hardness and electronegativity difference within a set of 30 HEAs. The hardness values were extracted from the literature, whereas the electronegativity differences were calculated from the information about the electronegativities of the elements involved in the formulation of the HEAs. The study related to both variables demonstrated that they are moderately correlated to the point of obtaining a non-reliable linear equation to perform hardness prediction as a function of the electronegativity difference. Therefore, in the context of the selected data, electronegativity difference is just moderately relevant in the hardness prediction of these 30 HEAs.

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References

1. Nonato RBP, Restivo TAG, Machado Junior JC. Uncertainty Quantification in Masses of Alloy Components and Atomic Radii Modification in High-Entropy Alloys Design: Thermophysical Parameters Calculation Approach. Materials Research 2025;28:e20240350.
2. Borg CKH, Frey C, Moh J, Pollock TM, Gorsse S, Miracle DB, Senkov ON, Meredig B, Saal JE. Expanded dataset of mechanical properties and observed phases of multi-principal element alloys, Sci Data. 2020;7:430.
3. Restivo TAG, Restivo GMG. Development of ultra-hard multi-component alloys, Journal of Materials Research. 2021;36:1316-27.
4. Fang S, Xiao X, Xia L, Li W, Dong Y. Relationship between the widths of supercooled liquid regions and bond parameters of Mg-based bulk metallic glasses. Journal of Non-crystalline Solids. 2013;321:120-5.
5. Nonato RBP, Restivo TAG. A multiparametric and multicriteria software to evaluate the design of ultra-hard metal alloys. In: Reinaldo Cardoso; João Batista Quintela. (Org.). Open Science Research X. 1ed.Guarujá: Científica Digital, 2023;1:1464-83.