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Balancing phases for obtaining the same hardness in Al-Cu-Zn alloys with different compositions

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ABSTRACT: Five Al-Cu-Zn alloy samples were developed using different chemical compositions. All samples have the same hardness of 67 RB, which was obtained by a methodology that predicts the hardness of alloys. Each alloy sample was analyzed using X-ray diffraction, scanning electron microscope, and hardness measurements. This analysis permitted to know the change in chemical composition for maintaining the same hardness in each sample. The samples used in equilibrium have of three phases, η, α and τ', however, in these samples there are other phases present, such as ε, β and θ. As is well known, if the chemical percentage is changed in the alloys, the phase percentage changes, and therefore the mechanical properties. This work results show that even when the chemical percentages drastically change, the hardness remains the same. The principal cause for maintaining the same hardness is using the contact area in the white zone composed by η and ε with respect to the other phases. If the perimeter of contact of the white zone is kept within a specific range, the hardness of all samples will be the same with a margin of error that is less than 3%. This is important to the development to new alloys.

KEYWORDS: Balance diagram; Hardness; Phases; Ternary alloy

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RESUMEN: *Obtención de la misma dureza por balanceo de fases en las aleaciones Al-Cu-Zn*. Se prepararon 5 muestras de las aleaciones Al-Cu-Zn con diferentes porcentajes químicos, utilizando una metodología que pronostica la dureza en este tipo de aleaciones con la finalidad de que todas las muestras tengan la misma dureza (67 RB). Cada una de estas muestras fueron caracterizadas por DRX, Microscopia Electrónica de Barrido (MEB) y un durómetro, se observó el cambio en la estructura. Las muestras utilizadas en condición de equilibrio constan de tres fases que son η, α y τ', sin embargo, en estas muestras se presentan otras fases como son las fases ε, β y θ. Este trabajo demuestra que es posible aun con los cambios en los porcentajes químicos que son drásticos, es posible tener la misma dureza. La principal causa que se encontró en este trabajo para mantener la misma dureza es el área de contacto de una zona blanca (compuesta por las fases η y ε) con respecto a las otras fases. Si el perímetro de contacto de la zona blanca está dentro de un cierto rango, la dureza de todas las muestras será la misma con un margen de error menor al 3%, esto es importante para desarrollo de nuevas aleaciones con las características mecánicas deseadas.

PALABRAS CLAVE: Aleación ternaria; Diagrama de equilibrio; Dureza; Fases

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1. INTRODUCTION

The Al-Cu-Zn alloy system is important for several reasons. In the region of the ternary diagram, it has a shape memory (Lovely and Torra, 1999; Iacovello *et al.*, 2018), in another section, they present superplasticity (Hsu and Wang, 1996) and in another, the transformation of the four phases that causes a contraction of 4% of its volume (Kovachera *et al.*, 1993; Klopotov *et al.*, 2016).

We have developed a series of investigations in which we have determined the mechanical properties of an Al-Cu-Zn alloy modifying the percentage of Zn, Al or Cu in order to determine the increase or decrease of one of the mechanical properties, for example hardness (Adeosun *et al.*, 2011; Yan *et al.*, 2014; Saravanan and Sellamuthu, 2014). These investigations are important because deducing or predicting what will happen when one of the elements is varied will allow for the design of alloys that fulfill required needs.

Therefore, many of the questions that researchers have proposed in material investigation have to do with which proportion of elements should be mixed in order to obtain materials with the desired mechanical, physical and chemical properties (Suárez *et al.*, 2011; Tiryakioğlu, 2015; Guler *et al.*, 2018).

Many studies have been performed on the phases in Al-Cu-Zn alloys in which the changes in mechanical properties, related to the presence of certain phases or the absence of the same have been explained, as shown in various investigations (Saravanan and Sellamuthu, 2014; Alaneme *et al.*, 2017).

In previous works it has been theorized, but not shown, that there can be different alloys with the same hardness that have chemical percentages and phases that are very different from Al-Cu-Zn ternary alloys (Villegas-Cárdenas *et al.*, 2011: Villegas-Cárdenas *et al.*, 2014).

Understanding that the type of interaction between phases affects the mechanical properties of an alloy has been an important topic for many years. In other words, a key question in material investigation has been how elements can be combined in such a way as to produce a solid with specific properties (Rohrer, 2014). The objective of this work is to obtain a series of different alloys with different chemical compositions but with the same hardness, only by manipulating the percentages of the phases.

2. MATERIALS AND METHODS

2.1. Obtaining the percentages of alloys

According to the article "Prediction of the Hardness of Al-Cu-Zn Alloys in Casting and Cooling" (Villegas-Cárdenas *et al.*, 2014), it is possible to determine the hardness of the Al-Cu-Zn alloys when they are within a zone made up by phases α, η and τ. In Fig. 1, in isothermal range 25°C, we can see two series of dotted lines, in which the first goes from M1 to M8 and the second from M9 to M16. Each of these is an Al-Cu-Zn alloy with a different chemical percentage.

From each line shown in Fig. 1 we obtain an equation that represents a chemical percentage; Eqs. (1) and (2).

$$X_{zn} = -1.9438X_{cu} + 0.50334 \quad (1)$$

$$X_{zn} = -2.9823X_{cu} + 0.97337 \quad (2)$$

The Eq. (1) is the straight line that represents the points that go from M1 to M8 in Fig. 1. The Eq.

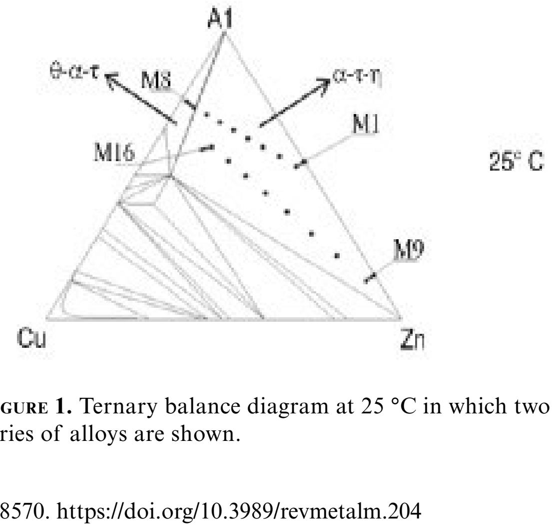

Figure 1. Ternary balance diagram at 25 °C in which two series of alloys are shown.

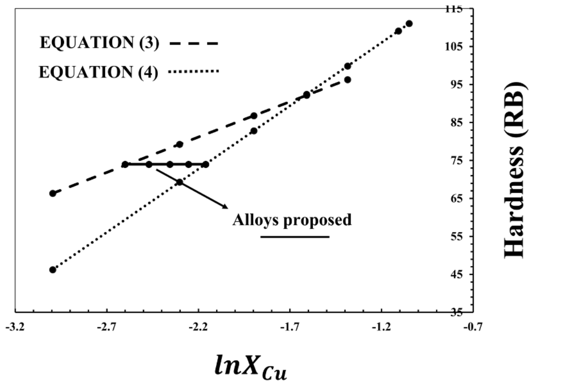

Figure 2. Representation of equations 3 and 4 and of the test samples used to obtain a hardness of 74 RB in all samples.

(2) represents points M9 to M16. Each of the points shown in Fig. 1 represents an alloy, when the hardness of these alloys is obtained after the homogenizing process, the hardness of these alloys shows a lineal tendency as observed in Fig. 2. The graph of equations 3 and 4 are shown in Fig. 2,

The equations from this lineal regression are:

$$HB = 18.617 lnX_{cu} + 122.12 \qquad (3)$$

$$HB = 33.326 lnX_{cu} + 146.05 \qquad (4)$$

In Fig. 2 we show the samples used in this work. As can be seen, there is a horizontal line set at 74RB so that all samples present this same hardness. Each of these alloys has a certain percentage of Cu, but the percentages of Zn and Al are unknown. Therefore, the slope formed between each of the alloys proposed with respect to the point of intersection of equation 3 and 4 must be known. The point of intersection of the two lines is 19.65% Cu with a hardness of 91.83 RB, with the previous point and the points of each of the samples, the slope of the hardness of each alloy as shown in Table 1 is obtained.

The slope of hardness have a direct relationship with the slopes of atomic percentages therefore we used an interpolation between the slopes of Eqs. (1-4) and the slopes of Table 1.

Table 1. Obtaining the slope of hardness as per the hardness of each alloy classified from S1 to S5

Sample	%Cu	Teórica Hardness (RB)	Slope of Hardness
S1	0.074274	74	18.3249
S2	0.084512	74	21.1290
S3	0.094751	74	24.4408
S4	0.104990	74	28.4406
S5	0.115229	74	33.3973

Table 2. Relationship between slopes for interpolation

Independent Variable (slope of hardness)	Dependent Variable (slope atomic fraction)
18.617	-1.943800
P_{Hd}	P_{at}
33.326	-2.982300

Table 2 shows interpolation, where P_{Hd} is the hardness slope obtained in Table 1 while P_{at} is the slope formed by the atomic fraction which is unknown and therefore is a dependent variable in interpolation, and:

$$P_{at} = \left[\frac{-2.9823 - (-1.9438)}{33.326 - 18.617}\right] [P_{Hd} - 18.617] - 1.9438 \qquad (5)$$

It is also necessary to determine the intercept with X_{Zn} for which the point of intersection of Eqs. (1) and (2) is needed, and which is a virtual point that is found at 0.452605 at. Cu and -0.376433 at. Zn. The equation of the ordinate at origin B is:

$$B = -0.376433 - P_{at}(0.452605) \qquad (6)$$

Table 3. Obtaining percentages of Al, Cu, Zn for obtaining a hardness of 74RB in each of the samples

Sample	P_{at}	B	Zn (fraction at.)	Al (fraction at.)	Cu (fraction at.)
S1	-1.9232	0.4940	0.3512	0.5746	0.0743
S2	-2.1212	0.5836	0.4043	0.5111	0.0845
S3	-2.3550	0.6894	0.4663	0.4389	0.0948
S4	-2.6374	0.8173	0.5404	0.3547	0.1050
S5	-2.9873	0.9756	0.6314	0.2534	0.1152

With the slopes from Table 1 and equations 5 and 6 we obtain Table 3 that shows the percentage of Zn and Al needed to obtain a hardness of 74 RB. Each of the simples in theory, as per our methodology, should have a hardness of 74 RB.

2.2. Experimental methodology and results

The samples were developed using conventional casting and the percentages shown in Table 3, after which a 180 h homogenizing process was performed at 350 °C. Each of the samples was characterized using DRX and electronic scan microscopy (SEM). The hardness was also measured in each of the samples in Rockwell B (RB).

The hardness obtained for each of the samples was not 74 RB, the results that in theory should have been obtained, however, the average was 68.41 RB, showing an error of only 7.56% which is an acceptable error. Table 4 shows the hardness of each sample and the error in each.

2.3. DRX Analysis

The results obtained in the diffractometer are shown in Fig. 3. All samples have the phases η, ε, α and τ', but samples S2, S3 and S4 show the phase β and samples S1 and S2 show the phase θ. Sample S2 shows a greater number of phases, 6, and as per the results shown in Table 4, we can see that this sample has greater hardness (only 2RB). Also, seen in Fig. 3 is the intensity in the phases η and α which decreases as the amounts of Zn and Cu increase. At the same time the intensity of phase ε increases in sample S2, in which there is almost no phase ε, phases β and θ are present, especially the latter, which is present in greater intensity.

Table 4. Real and predicted hardness for each sample, along with the error between the two

Sample	Hardness predicted	Hardness Real Average	Mistake (%)
S1	74	67.50	8.78
S2	74	69.57	5.99
S3	74	68.75	7.09
S4	74	68.00	8.11
S5	74	68.21	7.82

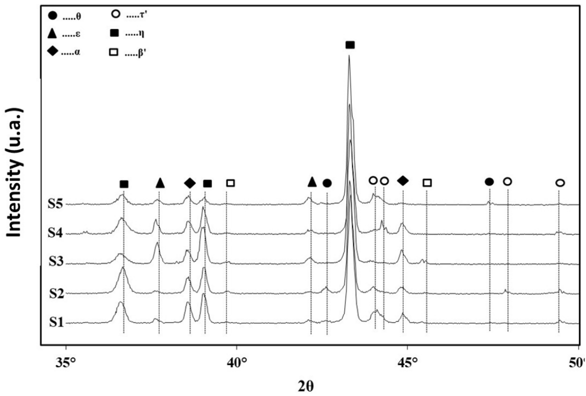

Figure 3. Diffractogram for each of the samples, in which the phases of each can be seen.

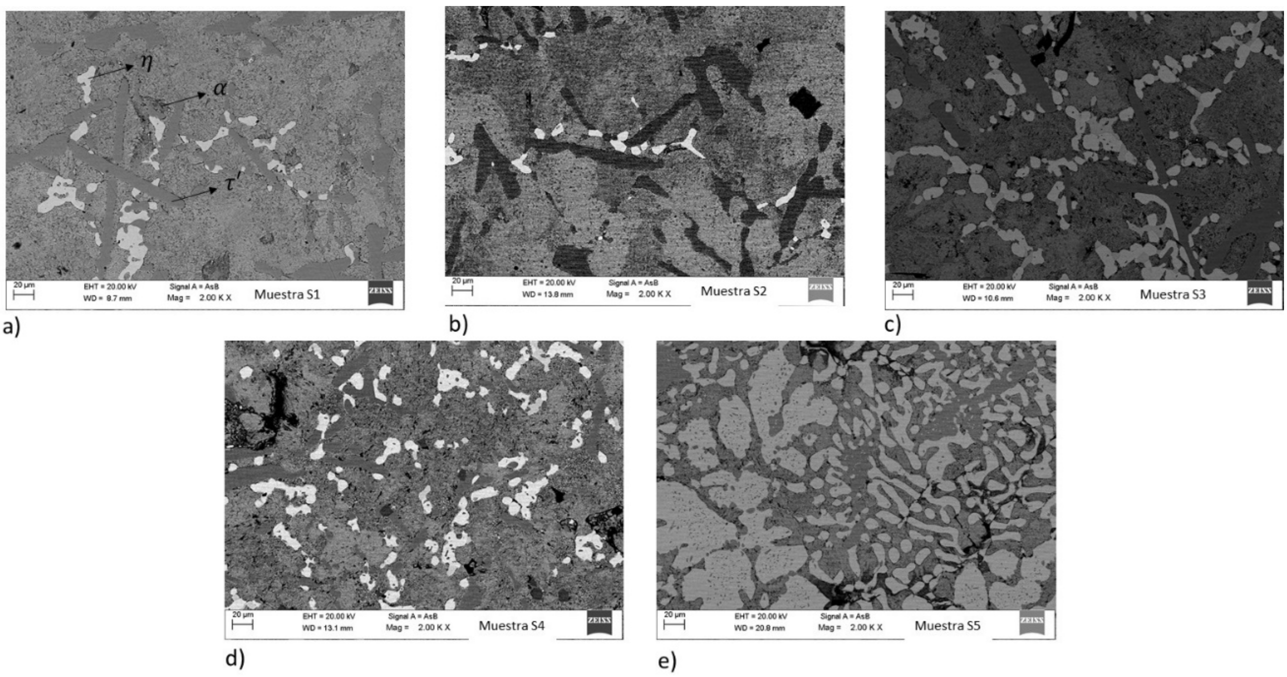

Figure 4. Metallography of each sample in backscatter mode in the electronic scan microscope.

2.4. Metallography

Figure 4 shows the different metallographic taken for each sample using an SEM in backscatter mode for phase identification:

In the metallographic shown in Fig. 4, we can see that there is a considerable increase in the white zone where the greater proportion in phase η and a lower proportion in phase ε are found. As the amount of Zn is increased, this white zone increases, which is logical because phase η is rich in Zn. The issue is that this increase does not occur in lineal form as corresponds to the amount of Zn, which does increase in linear form. We can see important differences in the metallography, for example, when a perimeter analysis, shown in the metallographic, has a white zone, compared to the rest of the phases, the graph shown in Fig. 5 is obtained. Analysis of the graph in Fig. 5 shows that all the samples behave in the same way, meaning that they all have a greater number of repetitions within the range of 25.09 to 35.61 micrometers. We must point out that the graph only shows 52% of all information obtained, however, the graph shows the densest part of the statistics for each sample used.

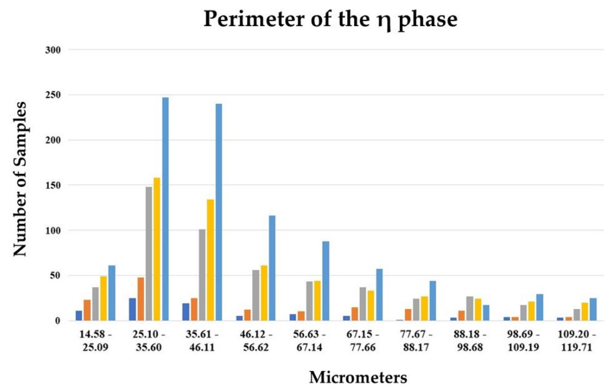

Figure 5. Bar chart of the perimeter of phase η for each of the samples from S1 to S5.

Table 5 shows the maximum and minimum measurements for each sample, along with the percentage that is represented in Fig. 5.

3. DISCUSSION OF RESULTS

Even with the error that exists between the theoretical and real results, we must mention that the hardness remained the same, in other words all samples basically have the same hardness. The difference between the highest and lowest hardness is only 2.07 RB and the standard deviation in the samples is 0.79011, achieving the goal of having 5 samples with the same hardness.

All samples have the phases η, ε, α and τ' and the intensity of these changes with the chemical percentage. This change in the intensity of the phases helps to have the same hardness even when exist a dramatic change in the chemical percentage, for example the percentage of cupper between the samples S1 to S5 are 50% and the same the Aluminium and Zn. Therefore, is possible to obtain materials with the same hardness but with different percentages chemical.

On the other hand, we must remember that the perimeter shown in the metallographic is just the proportional relationship of the area that is in contact with other phases. This is important because in the surface area there is a greater number of dislocations, and more dislocations will correspond to greater hardness, showing a relationship between dislocations and hardness.

The perimeter of each of the samples with respect to the white zone increases proportionally to the increase in the percentages in Zn and Cu. According to table 5 in each sample the 50% have a perimeter within a range of 14.58 µm to 46.12 µm and this is presented in the Fig. 5.

Therefore, we can say that this methodology can obtain different samples with different chemical percentages but with the same hardness. This is possible maybe by the perimeter that have the η phase

Table 5. Additional information from Fig. 5, in which standard deviation averages are presented, as well as the real percentages for the three main ranges shown in Fig. 1

Sample	# Measurement	Measurement		Average	Standard	# Dates by interval			Total	% Total
		Maximum	Minimum			14.58 a 25.01 (µm)	25.02 - 35.61 (µm)	35.62 - 46.12 (µm)		
S1	83.00	119.71	14.58	47.36	26.47	11.00	25.00	19.00	55.00	66.27
S2	178.00	253.13	15.47	58.92	41.95	23.00	48.00	25.00	96.00	53.93
S3	659.00	923.30	15.95	99.61	116.05	37.00	148.00	101.00	286.00	43.40
S4	646.00	506.35	17.20	67.99	63.45	49.00	158.00	134.00	341.00	52.79
S5	1132.00	4097.79	15.34	111.45	262.69	61.00	247.00	240.00	548.00	48.41

with respect another phase according to the metallographic.

4. CONCLUSIONS

According to what we have previously analyzed:

- The average hardness obtained in this work was 68.41. The difference between the minimum and maximum hardness obtained in samples S1 to S5 was 2.1 RB which is only 3% and we can effectively say that they have the same hardness and that the methodology shown in this work is correct.
- The diffractograms show that phases η, ε, α and τ' exist in all samples, even though some samples have other additional phases. Samples S3 and S4 have phase β, sample S1 has phase θ and sample S2 includes phases θ and β. Even with the existence of other phases like θ and β, they don't make much difference in the hardness of the samples. Therefore, we can conclude that the interaction between base phases is the reason that the hardness doesn't change.
- When a metallographic analysis is performed using the SEM in backscattering mode, we can see that there is an increase in the white zone made up by phases η, ε and in some cases by β as the amount of Zn increases and the amount of Al decreases. The difference between the highest and lowest percentage of aluminum is more than 50%, meaning that it is possible to obtain alloys with the same mechanical, physical and chemical properties but with different chemical percentages for the purpose of making a more economical product or improving the product's usefulness.
- Finally, as we know, most of the dislocations are found at the edge of the grain and the number of dislocations is directly related to hardness. When an analysis of the perimeter of the white zone is performed in accordance with metallographic obtained by backscattering using a SEM, we obtain a relation between the surface area in contact with the white zone and the other phases, which allows us to conclude that the samples have the same hardness, only if 80% of the white zone has a perimeter within the range that spans from 14.58 to 56.62 micrometers. When this condition exists, the sample will have the same hardness for these alloys.

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