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Effects of Dead Sea Water and Severe Plastic Deformation on the Microhardness and Corrosion Resistance of Copper-Alloyed Aluminum

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Abstract

Due to its lightweight, aluminum is frequently deployed in a variety of technical applications. Copper was added to aluminum at varying percentages, namely, 2, 4, and 6%, to improve its mechanical properties and resistance to corrosion. Following the copper additions and the upsetting process, microhardness was examined. Additionally, research was done on the effects of Dead Sea water on corrosion resistance and microhardness. It was found that the microhardness increased as the copper percentage increased in the as-cast process. A noticeable improvement in microhardness was achieved after the upsetting process, whereas a decrease in microhardness occurred after insertion in the Dead Sea water. Furthermore, microhardness rose as copper content increased. For example, a 2% Cu addition resulted in a 32% hardness enhancement, a 4% Cu addition resulted in a 160% hardness enhancement, and a 6% Cu addition yielded the most improvement, or about 242.7%, which resulted in corrosion resistance enhancement.

Keywords: Microhardness; Aluminum; Copper; Corrosion resistance; Dead Sea water.
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1. Introduction

Aluminum's diverse characteristics have made it the second most frequently used and produced metal (most widely in the non-ferrous category) in everyday applications.[1] Dada *et al*. reported that aluminum alloys can be optimized for next-generation applications by integrating developing technologies, designing alloys sustainably, and conducting future research in these areas.[2] Davis revealed that smooth metal has a high degree of electromagnetic spectrum reflection, spanning from radio waves to visible light and beyond into the infrared and thermal ranges.[3] Approximately 80% of visible light and 90% of radiant heat that strikes its surface are reflected by it.[3] Because aluminum is non-toxic and has good formability, as well as the ability to be recycled,

it is used in the food industry to make storage containers and thin foils. Aluminum (Al) structures are becoming increasingly used in the construction and building industries, as well as in container packaging and electrical transmission lines.

There are various reasons for aluminum corrosion. Al has a high acid solubility yet does not corrode in nitric acid.[1] Aluminum is also susceptible to pitting corrosion due to passivity loss, and corrosion occurs in the presence of a chloride (Cl^-) environment. However, the presence of an oxidant is required for corrosion to occur, chloride alone is insufficient. According to Callister, the strengthening process is based on the notion of reducing surface boundary area so that the grain boundary is less likely to slip, resulting in enhanced strength.[4] The addition of fine copper particles (4-10 wt.% Cu) to aluminum may improve strength, hardness, fatigue, creep resistance, and machinability. Copper is particularly soluble in aluminum. Therefore, Al-Cu alloys have the lowest corrosion potential of any aluminum alloy. Rana indicated that alloying elements are chosen based on their effect and suitability.[5] Dendrites of solid Al solution make up the microstructure of cast Al-Cu alloys, which form

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a brittle, nearly continuous multi-phase network of eutectics at the grain boundaries.[1] Wrought products are made up of an Al solid solution matrix with soluble and insoluble elements scattered within it.

Cu, as an alloying element, has a negative influence on the corrosion performance of Al in chloride-containing solutions. Thus, a sufficient amount of composition must be generated to improve mechanical qualities while maintaining corrosion resistance. According to Román *et al*., the concentration of Cu in the alloys was a critical determinant in the corrosion resistance of the Al-Cu alloys, controlling the proportion of phases as well as the morphological distribution of these phases.[6] When compared to pure Al, the presence of Cu offers cathodic sites that boost neighboring anodic activity and increase the corrosion susceptibility of the Al-Cu alloys. An increase in the Cu quantity is linked to an increase in the concentration of the Al_2Cu intermetallic or theta phase, resulting in a higher sensitivity to corrosion for the investigated alloys. The corrosion resistance of Al-Cu alloys is improved by a microstructural morphology characterized by a reduced region of contact between the α-phase and the Al_2Cu intermetallic phase. Because previous studies yielded contradictory results regarding the effect of Cu-alloying on the pitting potential of aluminum alloys, the current study aims to investigate the effect of Cu content in Al-Cu alloys and provide a thorough explanation of the relationship between pitting corrosion resistance and the various phases and microstructures present in the alloys. The corrosion-causing media must be brought to the surface, and the corrosion products must be removed. At room temperature and higher temperatures, Bergsma *et al*. studied the tensile properties of "air slip" cast aluminum alloy ingots 2618 and 2618 alloyed with extra copper (along with some additional manganese, iron, and nickel).[7] Seshagiri *et al*. showed that AA2219 is an outlier because of its higher copper content, which provides more eutectics to promote fracture healing.[8] According to Kaya's findings, increasing deformation hardening together with rising explosive ratios resulted in a lower impact toughness of composites.[9] Madhusudan *et al*. demonstrated the effect of particle composition by altering the copper concentration from 5 to 15 wt%.[10] In both cast and homogenized settings, hardness rose as particle content increased. When compared to the alloy, composites have a 13% decrease in strength and a 15% decrease in strain.[11] The X-ray diffraction (XRD) and energy-dispersive X-ray spectroscopy (EDS) investigations revealed that the intermetallic phases Al_2Cu and Al_4Cu_9 were present in all samples.

To forecast the formation of Al-Cu compounds at the solid-state boundary, the effective Gibbs free energy change of formation model was suggested, and the calculation, combined with kinetic factors, revealed that Al_2Cu (Al side) and Al_4Cu_9 (Cu side) occurred first. Lapovok found that when copper and aluminum powders were simultaneously compacted and strengthened, the mechanical strength properties of the 20 and 30 wt.% copper powder were improved by more than twice, and the electrical conductivity of the 30 wt.% copper powder was increased significantly.[12] Morales examined how acetylene and oxygen gas pressure affected the bronze-aluminum coatings' ability to withstand corrosion.[13] Corrosion resistance increases in the order Al-6% Cu, Al-6% Si. This was found by examining the corrosion behavior of pure aluminum, Al-6%Cu, and Al-6%Si alloys in Na_2SO_4 solutions in the presence and absence of NaCl, NaBr, and NaI. The strongest ions thus far have been chloride ions.[14] The effect of plastic deformation on the corrosion resistance of alloys has been discussed in previous studies.[15,16] Higher copper concentrations resulted in an enhanced conductivity, even though microarc oxidation (MAO) coatings typically improve the electrical insulation because of the alumina phases (*i.e.*, α-AlO_3 and γ-AlO_3).[17] The development of coherent Cu-containing "β" precursors during preaging was responsible for the notable improvement in the resulting alloy's strength.[18] Temperature and flow rate had an accelerated influence on fouling, and the corrosion inhibitor starch effectively reduced fouling in heat exchangers made from 6061 Al.[19] The maximum critical state for dynamic equilibrium between impedance matching and attenuation constants in the low-frequency band is raised as a result of equivalent dipoles produced by double electron-rich sites (containing Cu and Zn elements) acting as effective attenuation units.[20] An aluminum nanopillar metasurface filter was used as a narrowband filter for enhancing thermophotovoltaic (TPV) system performance.[21] The passivation solution mixed with Zn^{2+} showed better corrosion resistance than that mixed with Cu^{2+} and Ni^{2+}.[22] Oxide films formed on the surface of TiNiCu shape memory alloys (SMAs) are beneficial for use as heat engine actuators due to the following reasons.[23] Corrosion-resistant alloys (CRA) pipelines typically have higher initial costs than carbon steel pipelines, but they offer significant long-term savings by reducing corrosion maintenance, inspection, and repair costs.[24]

The main objective of this study is to determine how copper addition at different concentrations, specifically 2, 4, and 6%, influences the microhardness and mechanical properties of the resulting Al-Cu alloys. Additionally, the effect of Dead Sea water and the upsetting process on the Al-

Cu corrosion resistance will be investigated.

2. Materials, equipment, and experimental procedures

2.1 Materials

The present work made use of a variety of materials, including commercially pure Al (99.8 %), high-purity Cu, pure graphite crucibles, and pure graphite rods. In the current work, copper is utilized as an alloying element. It was accessible as a 99.8 % pure powder with a melting point of 1083 ºC and a density of 8.2 gm/cm^3 at 20 ºC, where the chemical composition of copper is 0.01% Zn, 0.02% Pb, 0.01% Sn, 0.003% P, 0.02% Fe, 0.02% Ni, 0.03% Co, 0.002% Al and 99.50 % latent Cu by wt.%. The chemical composition of commercial Al is 0.09% Fe, 0.05% Si, 0.005 % Cu, 0.004% Mg, 0.004% Ti, 0.008% V, 0.005% Zn, 0.0011% Mn, 0.005% Na. The chemical composition of Dead Sea water (prine) is shown in Table 1.

Table 1: Elemental composition of detritic minerals present in the Dead Sea mud.[14]

Mineral	Content (wt.%)
Silicon dioxide	20
Calcium oxide	15.5
Aluminum oxide	4.8
Magnesium oxide	4.5
Iron(III) oxide	2.8
Sodium oxide	1.7
Potassium oxide	1.3
Titanium(IV) oxide	0.5
Sulfur trioxide	0.4
Phosphorus pentoxide	0.3
Chloride	6.7
Bromide	0.2

The Al-Cu binary phase diagram is shown in Fig. 1. The maximum solubility of copper in aluminum matrix is about 6 % Cu at 548.2 °C eutectic temperature. This indicates that the possible amount of copper to be added is 6 %, extra percentage of copper will not dissolve in the mixture.

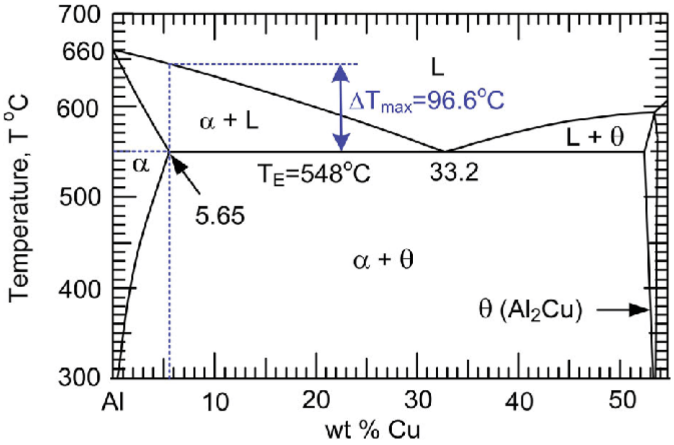

Fig. 1: Binary Al-Cu phase diagram.

2.2 Equipment and experimental procedures

The study made use of a variety of tools and machinery, one of which was an electric resistance furnace (Nabertherm) with a temperature of 1750 °C. In addition, an MT7530 Trinocular Metallurgical Microscope, an Instron machine with a 100 kN capacity (Quasar 100), and a Falcon 400 Microhardness tester were used for different analyses. Workpieces were prepared with an Excel-type computer numerical control (CNC) milling machine. Additionally, the average surface roughness (Ra) was measured using a surface roughness tester with a 0.8 mm cut-off distance, and the ISO 13565 (Rk) parameters were also taken into account during the evaluation process. These devices and tools were essential to carrying out thorough analyses and evaluations for the study.

2.2.1 Preparation of pure aluminum alloy and Al-Cu alloys

The aluminum pure was made by melting a pre-determined amount of high purity aluminum rods at 850 °C and then adding a predetermined amount of pure Cu to the melt in a graphite crucible. The melt was swirled for 2 minutes and returned to the furnace for 30 minutes before being put into a brass mold to solidify in the air. The aluminum pure alloy was synthesized as 14 mm diameter and 70 mm long cylindrical rods, from which test samples were machined. In the same manner, Al-Cu alloys were prepared at 2, 4, and 6 % Cu additions. The melting and casting of Al and Al-Cu alloys are shown in Fig. S1.

2.2.2 Digital microhardness tester

In materials, hardness is one of the most important characteristics to consider, and it is often used to give a broad notion of a material's strength and resistance to wear and scratching. A micro-Vickers hardness tester (Falcon 400, Netherlands) based on ASTM E140 that shown in Fig. S2 was

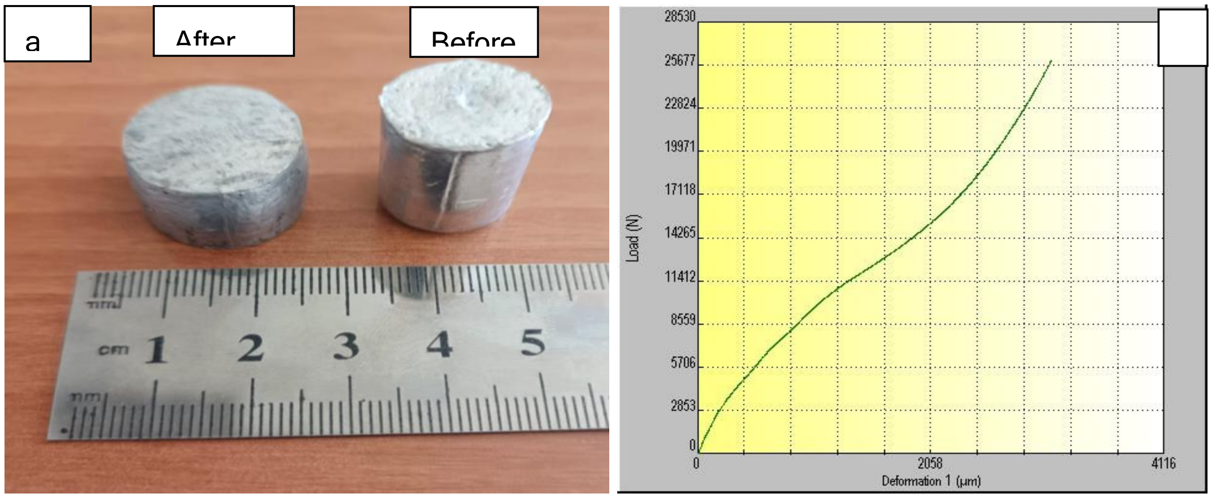

Fig. 2: (a) Workpiece before and after upsetting process and (b) load- deflection curve of Al-Cu alloy.

used to measure the samples' Vickers microhardness values in order to acquire the results. The measurements were made seven times with a 200 gf and a 10-second dwell duration. The average was then calculated.

2.2.3 Upsetting test

The mechanical properties of aluminum alloys as a whole are examined through the process of upsetting for the prepared small pieces, as shown in Fig. 2a. This procedure entails using the Instron machine to generate a load-deflection curve as shown in Fig. 2b. A true stress-true strain connection can be obtained from this curve. A thorough grasp of the material's mechanical characteristics and performance is made possible by this connection, which offers insightful information about how the material behaves under the applied loads.

2.2.4 Corrosion resistance procedure using Dead Sea water

After the upsetting process, all aluminum alloys that had been alloyed by copper in varying percentages such as 2, 4, and 6% were weighed, placed in Dead Sea solution for 21 days, and then taken out and weighed once more to measure the mass difference, or mass loss, which was then used as a gauge for corrosion resistance.

3. Results and discussion

3.1 Effect of Cu addition on the microstructure of aluminum

Fig. 3 demonstrates in detail how the structure of aluminum varies with varying copper concentrations. At first, pure

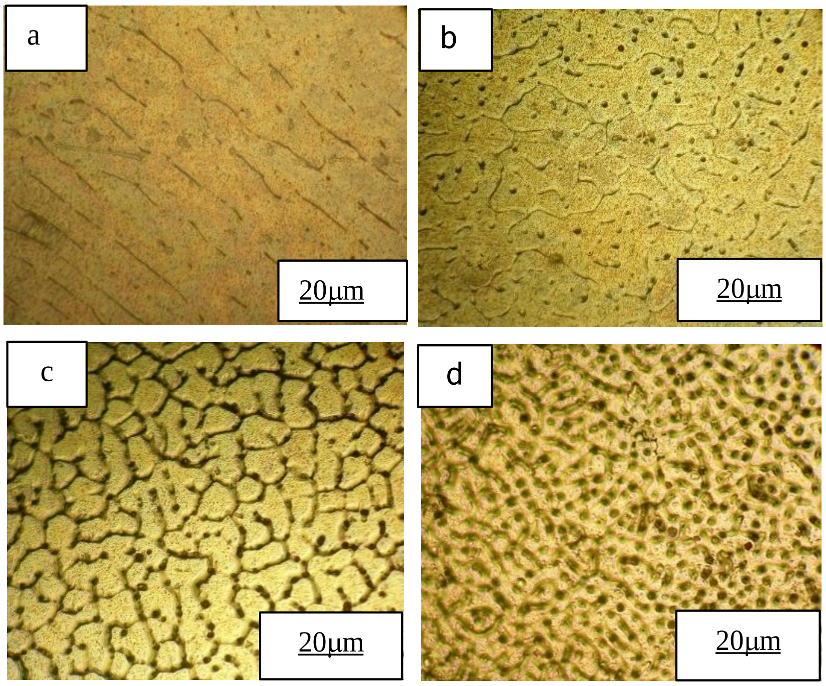

Fig. 3: Photomicrographs showing the general microstructure of Al and Al-Cu Alloys, (a) Al, (b) Al-2 % Cu, (c) Al-4 % Cu, and (d) Al-6% Cu at 500x.[25]

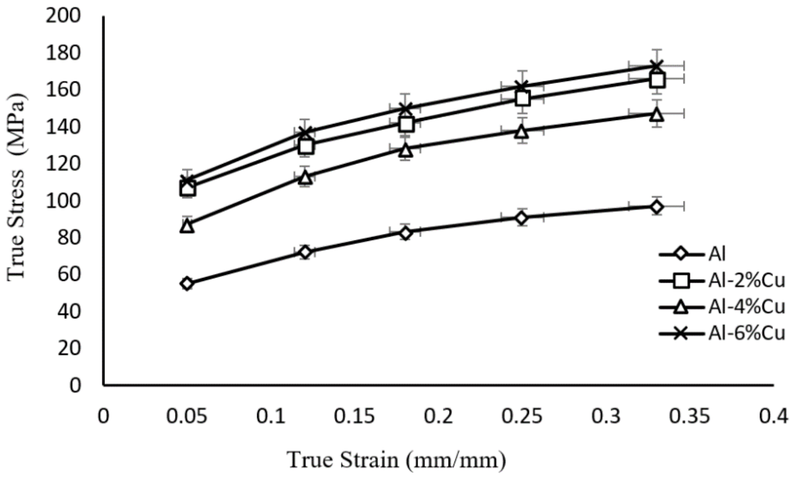

Fig. 4: Effect of Cu addition on the mechanical characteristics of Al.

aluminum has a columnar structure and poor mechanical properties, as seen in Fig. 3a. However, with adding 2% copper, as shown in Fig. 3b, the structure transforms into large equi-axial grains with relatively greater mechanical characteristics. The 4% copper addition, as seen in Fig. 3c, substantially enhances the mechanical properties by causing the grains to arrange into smaller equi-axial structures. As shown in Fig. 3d, the structure eventually transforms into fine grains with copper scattered throughout the grain at a 6% copper addition. Al-qawabah, suggests that this phenomenon could be explained by a rise in the percentage of copper, which leads to an increase in the amount of copper that appears inside the grain.[25]

3.2 Effect of Cu addition on the mechanical characteristics

Fig. 4 shows that the mechanical properties of the material and the addition of Cu are directly correlated. It is clear that as the amount of Cu increases, the mechanical properties noticeably improve. However, the improvement diminishes marginally with the addition of 4% Cu. This is explained by the fact that Cu dissolves in aluminum at a maximum of about 6%. Furthermore, the grain refinement obtained with 4 and 6% Cu additions is almost identical, resulting in a plateau in the mechanical characteristic enhancement above the 4% Cu addition.

3.3 Effect of Cu additions and after the upsetting process on the average microhardness of commercially pure Al

Fig. 5 illustrates how microhardness often rises as copper content increases. For example, a 2% Cu addition results in a 32% hardness enhancement, a 4% Cu addition results in a 160% hardness enhancement, and a 6% Cu addition yields the most improvement, or about 242.7%. This significant improvement can be attributed to the formation of fine grains as a result of the intermetallic compound formed (Al_2Cu) after the addition of Cu. These compounds give the material an overall strengthening effect, *i.e.*, it hinders the grain from growth and leads to the observed rise in microhardness, which can be observed obviously in Fig. 5.

On the other hand, it can be seen from Fig. 6 that there is a

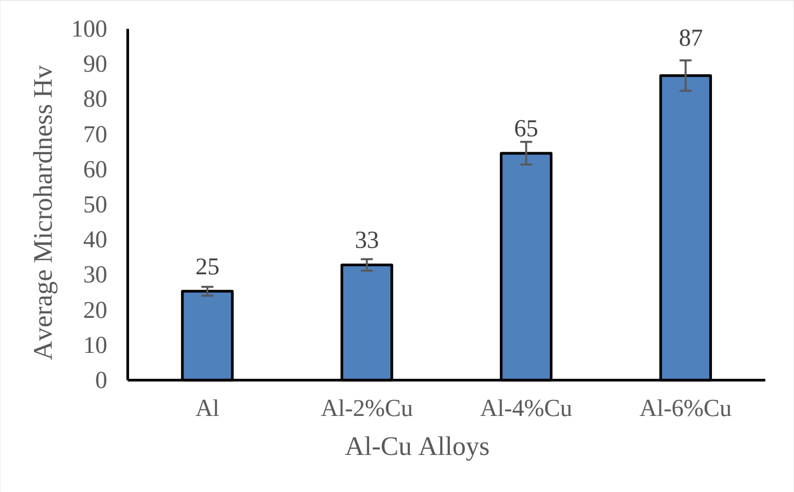

Fig. 5: Effect of Cu additions on the average microhardness of commercially pure Al.

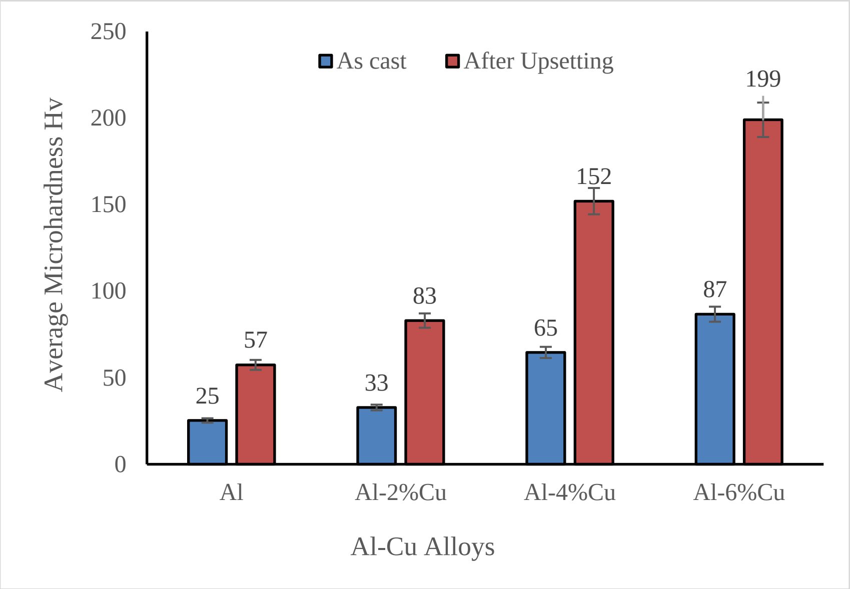

Fig. 6: Effect of Cu additions and after upsetting process on the average microhardness of commercially pure Al.

pronounced enhancement in microhardness after the upsetting process, with a maximum of 246.7% attained at 6% copper addition. This can be explained by the fact that after the upsetting process, there is a severe plastic deformation, which results in the refining of grains.

3.4 Effect of Cu additions, upsetting process, and after dead water insertion on the average microhardness of commercially pure Al

Fig. 7 clearly illustrates that the addition of 6 % copper resulted in a sharp increase in the microhardness, almost 3 times that of sample 1, as demonstrated by the data. The findings are congruent with our findings.[26,27] They discovered that the hardness of the alloys grew consistently as the copper concentration increased up to a maximum of 6 %. The copper concentration of the aluminum-rich phase affected the microhardness of the phase in a way comparable to that of the tensile strength of the phase. During the testing, it was discovered that as the copper concentration of the alloys increased, the wear loss dropped and reached a minimum of 2% Cu. According to Fig. 7, there is a sharp decrease in microhardness as a result of the as-cast test in comparison with upsetting. The microhardness of the 2% Al-Cu alloy was decreased by 60.50%. At the same time, the microhardness of the Al-Cu alloy containing 4 and 6% of Cu was decreased sharply by 57.56 and 56.40%, respectively. The decrease in Hv of the Al-Cu 6% alloys was from 199 in the upsetting method to 87 in the as-cast method.

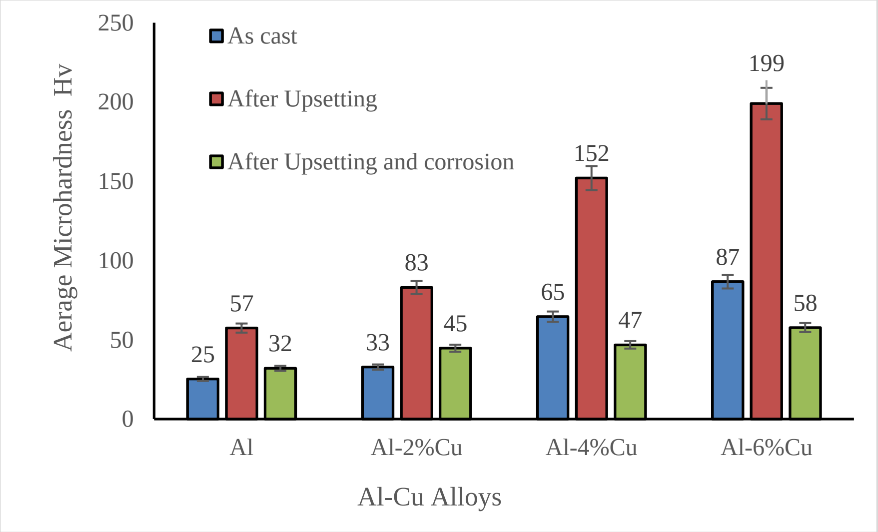

Fig. 7: Effect of Cu additions, upsetting process, and after Dead Sea water insertion on the average microhardness of commercially pure Al.

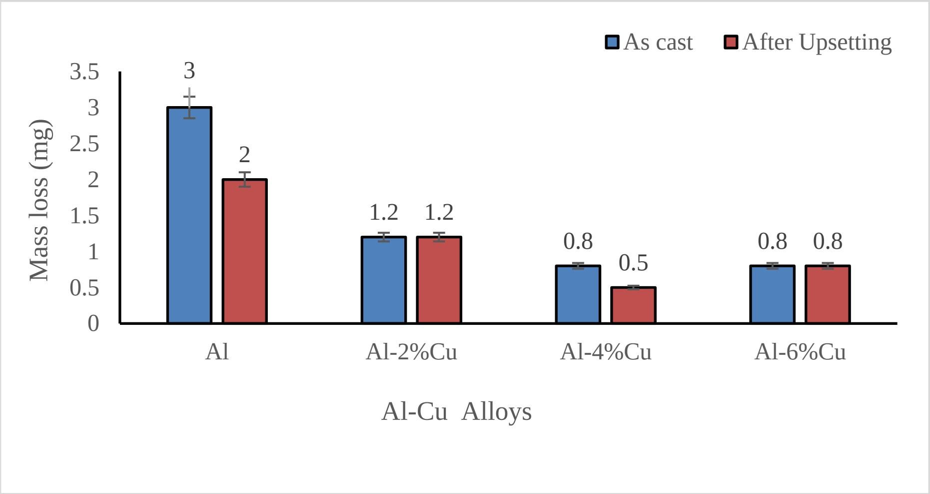

Fig. 8: Mass loss for Al-Cu alloys in the as-cast and after upsetting process conditions using Dead Sea water.

As we can see in Fig. 7, there is a change in microhardness Hv because of the upsetting after the corrosion test compared to the previous way. The microhardness of the Al-0%Cu and Al-2%Cu alloys were increased by 26.53 and 36.36%, respectively. However, there are decrease in the microhardness of the Al-4% Cu and Al-6% Cu by 27.56 and 48.39%, respectively.

3.5 Effect of copper additions and upsetting process on the average corrosion resistance using Dead Sea water

The inclusion of copper improves the corrosion resistance of Al. Similarly, increasing the copper content gave an enhancement in its corrosion resistance. This tendency can be explained by the beneficial effect of copper addition on grain size refining. It has been demonstrated that fine-grained materials have better corrosion resistance. During the as-cast method, the mass loss will decrease by increasing the percentage of copper in the alloy, as is clear in Fig. 8. Previous studies on corrosion in general attribute the enhancement in steel to Cu additions.[15,16] Furthermore, Salman *et al*. suggested Ta/Cr coating could be used for nuclear reactor core components in emergency situations for zirconium alloys,[28] where Song *et al*.[29] evaluated the service behavior of aluminum alloy in the marine atmospheric environment, the results showed that the wind speed and wind direction in the Xisha environment had a significant effect on the corrosion weight loss of 2A12 aluminum alloy. Fig. 8 shows the corrosion resistance after the upsetting process for Al-Cu alloys, which was recorded as a reference sample, and Al-Cu alloys with 6% Cu. In the same figure, the mass of Al-Cu 4% loss is 0.5 mg, which is the lowest amount compared to the other mixture alloys. This is because the Al-4%Cu alloy's hardness decreases by about 105 Hv after being inserted into Dead Sea water, whereas the Al-6%Cu alloy has 141 Hv hardness loss.

4. Conclusion

Extensive research has been done on the microhardness and corrosion resistance of Al-Cu alloys with different copper percentages. Microhardness increased with increasing copper% in the as-cast process, most likely as a result of the creation of new intermetallic compounds, Al-Cu. It's interesting to note that during the as-cast process, the microhardness dropped and the structure became softer in all samples with 2, 4, and 6% copper concentration. In particular, the softer alloy structure caused by the 4% Cu addition to the as-cast alloy decreased the press capacity during the forming procedures. This has led to additional research into these alloys' machinability, resistance to corrosion, and other properties. Notably, following upsetting, the alloy with 6% Cu concentration showed an enhanced resistance to corrosion and noble qualities. After the insertion of Al-Cu alloys, the microhardness of the Al-0%Cu and Al-2%Cu alloys was increased by 26.53 and 36.36%, respectively. However, there is a decrease in the microhardness of the Al-4% Cu and Al-6% Cu by 27.56 and 48.39%, respectively. Dead Sea water is a very corrosive medium, which explains this observation. As a result, the copper concentration of aluminum alloy is linked to the hardness reduction tendency.

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Conflict of Interest

There is no conflict of interest.

Supporting Information

Applicable.

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