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Chloride ion – corrosion activator and passivator

ABSTRACT

The presence of certain ions in the environment, particularly chloride ions (Cl^-), can significantly affect the corrosion behavior of brass alloy. Chloride ions are known for their ability to disrupt passive layers on the surface of metals and alloys, which can lead to localized corrosion such as pitting and dezincification—selective removal of zinc from the alloy—that can severely compromise its structural integrity. In certain cases, chloride ions may also act as passivators of corrosion processes, depending on their concentration, the composition of the material being tested, and the pH value of the solution. Therefore, understanding the mechanisms of brass corrosion in the presence of chloride ions is essential for its safe and long-term use, especially in water transport systems, sanitary equipment, and the marine industry, where these ions are commonly present. This study investigates the factors influencing the intensity and type of brass corrosion in chloride environments, with special emphasis on electrochemical testing methods, primarily polarization measurements with the determination of corrosion parameters (corrosion potentials, corrosion current densities, corrosion rates), the extent of passive and transpassive regions, and the morphology of corrosion products. The obtained results indicate a pronounced corrosive effect of both low and high chloride ion concentrations, as well as an inhibitory effect of certain chloride concentrations, for which numerous data can also be found in the literature.

Keywords: brass, corrosion, dezincification, chloride ions

1. INTRODUCTION

The corrosion behavior and dezincification process of cold-worked CuZn-42 brass were investigated in an acidic sulfate solution at pH 2 using the linear polarization method.

The measured corrosion potentials and corrosion current densities were observed as characteristics of the dezincification process and corrosion resistance of the tested cold-worked CuZn-42 brass samples. The corrosion behavior of brass has mainly been studied from the perspective of the dezincification mechanism and stress corrosion. Dezincification is a well-known (delegating) process that signifies the loss of brass and is an important physical-mechanical property leading to surface destruction [1–4]. It occurs in solutions containing specific chemical compounds.

Previous studies [5,6] have established that ions such as thiocyanates, bromides, and iodides do not participate in dezincification, while chlorides and sulfates, at certain concentrations, cause dezincification [7,8].

Two main theories explain the mechanism of brass dezincification: the first theory assumes selective dissolution of zinc, which leaves behind a porous residue rich in metallic copper, while the second theory assumes simultaneous dissolution of zinc and copper, with copper being redeposited to some extent. There are still conflicting opinions regarding copper redeposition, as well as the form and nature of brass corrosion products that separate on the sample surface during the selective dissolution of the alloy [9,10].

The corrosion behavior and dezincification process of cold-worked CuZn-42 brass were investigated in an acidic sulfate solution at pH 2, with the addition of chloride ions, using the linear polarization method and DC techniques (including Tafel extrapolation, potentiodynamic polarization, and linear polarization). The measured corrosion potentials and corrosion current densities were observed as characteristics of the dezincification process and corrosion resistance of the tested cold-worked CuZn-42 brass samples.

According to previous research on the dissolution of copper in chloride medium, the anodic reaction is reversible mainly due to the strong, thermodynamically more favorable complexation of copper ions with chloride ions

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[11,12,13]. The cathodic response is dominated by oxygen reduction, which is considered to be irreversible. Copper with chloride ions can form several complexes: CuCl , CuCl_2^- , CuCl_3^{2-} or CuCl_4^{3-} . The formation of the CuCl layer takes place according to the reaction: $\text{Cu} + 2\text{Cl}^- \rightarrow \text{CuCl}_2^-$ [14].

CuCl is poorly soluble in NaCl solution, resulting in the formation of a ion CuCl_2^- complex: $\text{CuCl} + 2\text{Cl}^- \rightarrow \text{CuCl}_2^- + \text{e}^-$. During hydrolysis CuCl_2^- ions in NaCl solution can cause precipitation of copper (I) oxide: $2\text{CuCl}_2^- + \text{H}_2\text{O} \rightarrow \text{Cu}_2\text{O} + 2\text{H}^+ + 4\text{Cl}^-$.

It is generally accepted that the anodic dissolution of Cu depends on the concentration of Cl^- ions and does not depend on the pH of the solution. At concentrations of Cl^- ions greater than $1\text{mol}\cdot\text{dm}^{-3}$, it is possible to form more complex complexes such as CuCl_3^{2-} and CuCl_4^{3-} [14-16].

When a passive film is created on a metal (e.g., Cu_2O) that does not have good protective properties, pitting corrosion will occur [17] in the presence of aggressive ions which is very dangerous because it quickly penetrates deep into the metal mass and can lead to cracking of the structure under stress whereby the Cu_2O film formed in this way can provide excellent protection during high-temperature treatment of copper and its potential application in nuclear facilities [18]. Pitting corrosion most often occurs during the transition from active to the passive state. Cu_2O stability depends on the concentration of chloride ions. Concerning dezincification of α and $\alpha + \beta'$ brasses, two mechanisms were proposed selective Zn dissolution [19] and a more widely accepted selective dissolution model consisting of the initial dissolution of both Zn and Cu , followed by Cu redeposition.

The corrosion rate depends on the environment: in 5% HCl it is $1.2\text{ mm}\cdot\text{year}^{-1}$ [20], and in 10–35% HCl , it can reach up to $7.1\text{ mm}\cdot\text{year}^{-1}$. In HCl solutions with oxygen, intense dissolution of zinc and copper occurs along with oxidation and copper redeposition reactions [21]. In seawater, corrosion is influenced by chloride concentration, temperature, and copper content.

2. EXPERIMENTAL PART

The electrochemical investigation of the corrosion and electrochemical behavior of various types of brass was conducted using the linear polarization method, which included determining the values of steady-state (corrosion) potentials, current densities, and corrosion current densities. The measured corrosion potentials and corrosion current densities were observed as characteristics of the dezincification process and corrosion resistance of the tested cold-worked CuZn-42 brass samples.

Apparatus

For polarization measurements, AMEL equipment was used, including: potentiostat model 553, programmable function generator model 568, interface model 560/A/log, integrator model 731, and digital x/y recorder model 863.

All measurements were performed using a classical three-electrode electrochemical cell made of glass, with space to accommodate the electrodes and a thermometer.

The tested CuZn-42 brass samples, produced in the laboratory, had the following chemical composition: Cu –57.95% (copper purity 99.997%), Zn –41.91% (zinc purity 99.85%), and others – 0.14%.

The samples were deformed to the following degrees of deformation: 0%, 20%, 40%, 60%, and 80%, and then embedded in a cold-polymerizing acrylate. The deformations were performed under laboratory conditions by cold rolling the samples between two parallel rollers, with the deformation extending throughout the entire height of the sample in all directions. The degree of deformation was determined based on the surface area of the deformed sample, where all samples had the same surface area, $A=0.38\text{cm}^2$.

A non-deformed copper electrode was used as a comparative sample.

Immediately before each polarization measurement, the surface of the working electrode was mechanically polished with #1000 grit sandpaper, then with alumina, rinsed with distilled water and ethyl alcohol, and air-dried. A saturated calomel electrode and a platinum wire (surface area= 2.0cm^2) were used as the reference and counter electrodes, respectively. All potential values reported in this work are given relative to the saturated calomel electrode (SCE). The solution stirring rate was 300 min^{-1} .

Reagents Used

During the polarization measurements, a $1\cdot 10^{-1}\text{M}$ Na_2SO_4 solution was used as the base electrolyte, into which chloride ion test solutions were added to achieve the following chloride concentrations in the base electrolyte: $5\cdot 10^{-4}\text{M}$ NaCl , $5\cdot 10^{-3}\text{M}$ NaCl , $5\cdot 10^{-2}\text{M}$ NaCl , $1\cdot 10^{-1}\text{M}$ NaCl , $5\cdot 10^{-1}\text{M}$ NaCl , and 1.0M NaCl .

Experimental Technique

The measurement methods used in all experiments were as follows:

1. Measurement of the potential-time dependence, $E=f(\tau)$, and determination of corrosion potentials (E_{corr}).
2. Potentiostatic method, $I=f(E)$; determination of the current density as a function of potential,

which includes recording the current density dependence on potential variation.

3. Determination of corrosion current densities, j_{corr} .
The values of corrosion current densities were determined by extrapolating the obtained Tafel curve to the corrosion potential, in the system: $\log j=f(E)$.

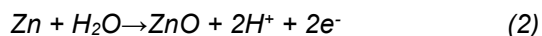
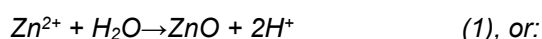
3. RESULTS, ANALYSIS, AND DISCUSSION

Chloride ions can play either a stimulating or inhibiting role in the corrosion process of brass [22]. A stimulating effect is observed when CuCl and Cu₂O films are present under the given conditions, or when the Cu₂O film is thermodynamically stable, but the presence of a critical amount of chloride ions promotes the disruption of its structure [23]. According to literature data [24–26], a chloride ion concentration of $8 \cdot 10^{-3}\text{M}$ exhibits an inhibiting effect on the corrosion of brass in a neutral ammoniacal solution. For a $1 \cdot 10^{-1}\text{M}$ Na₂SO₄ solution with added Cu²⁺ and Cl[−] ions, studies have shown that the highest corrosion of 63Cu–37Zn brass occurs at a Cl[−] concentration of $5 \cdot 10^{-3}\text{N}$, while inhibitory effects are observed at higher chloride concentrations.

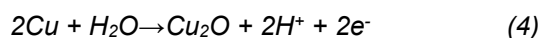
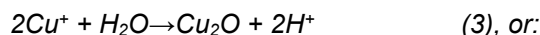
In strongly acidic solutions (pH values 1.5–3.5), when the chloride ion concentration is below $1 \cdot 10^{-2}\text{N}$, the inhibitory effect of chloride ions is not significant; however, as the chloride concentration increases, polarization curves indicate the presence of anodic inhibition.

The kinetics and mechanism of the anodic dissolution of brass and the formation of a film in a chloride solution can be explained by the following equations [27,28,29,30]:

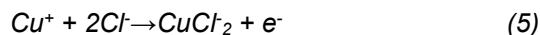
In the initial stage of corrosion, ZnO is formed as a result of the following reactions:



and Cu₂O oxide is formed as a result of the following reactions:



After the surface of the brass becomes covered with ZnO and Cu₂O compounds, a CuCl film forms on the surface through the following reaction:



This film may undergo a disproportionation reaction:



or the dissolution process of the formed CuCl₂[−] complex:



This paper presents the results of electrochemical investigations of brass behavior in chloride ion solutions. Based on the obtained results, conclusions were drawn regarding the corrosive and inhibitive behavior of brass in the tested solutions at pH 2.

The obtained values for corrosion potentials at chloride ion concentrations of $5 \cdot 10^{-2}\text{M}$, $1 \cdot 10^{-1}\text{M}$, and 1.0M fully correspond with the results presented in reference [31]. In a 0.6M NaCl solution, an increase in zinc content in brass leads to a shift of the corrosion potentials toward more negative values, which is in agreement with the results obtained in this study [32–34].

For the polarization curves shown in Figure 1, it is characteristic that the Cu electrode exhibits the lowest corrosion current density values (Table 1). As the degree of deformation increases, the corrosion current density values of brass also increase, with the highest values observed for CuZn-42 brass with 60% deformation. Among the tested brass electrodes, the lowest corrosion current density (j_{corr}) was recorded for the CuZn-42(80%) brass electrode [35,36].

Table 1. Corrosion current density values for CuZn-42 brass in chloride ion solutions with concentrations of $5 \cdot 10^{-4}\text{M}$ and $5 \cdot 10^{-3}\text{M}$ Cl[−] ions

$1 \cdot 10^{-1}\text{MNa}_2\text{SO}_4 + 5 \cdot 10^{-4}\text{MCl}^-$							
Sample		Cu	CuZn-42 (0%)	CuZn-42 (20%)	CuZn-42 (40%)	CuZn-42 (60%)	CuZn-42 (80%)
pH=2	j_{corr} (mA/cm ²)	0.727	1.04	1.108	1.166	1.42	1.035
$1 \cdot 10^{-1}\text{MNa}_2\text{SO}_4 + 5 \cdot 10^{-3}\text{MCl}^-$							
Sample		Cu	CuZn-42 (0%)	CuZn-42 (20%)	CuZn-42 (40%)	CuZn-42 (60%)	CuZn-42 (80%)
pH=2	j_{corr} (mA/cm ²)	0.988	1.298	1.585	1.688	1.721	1.278

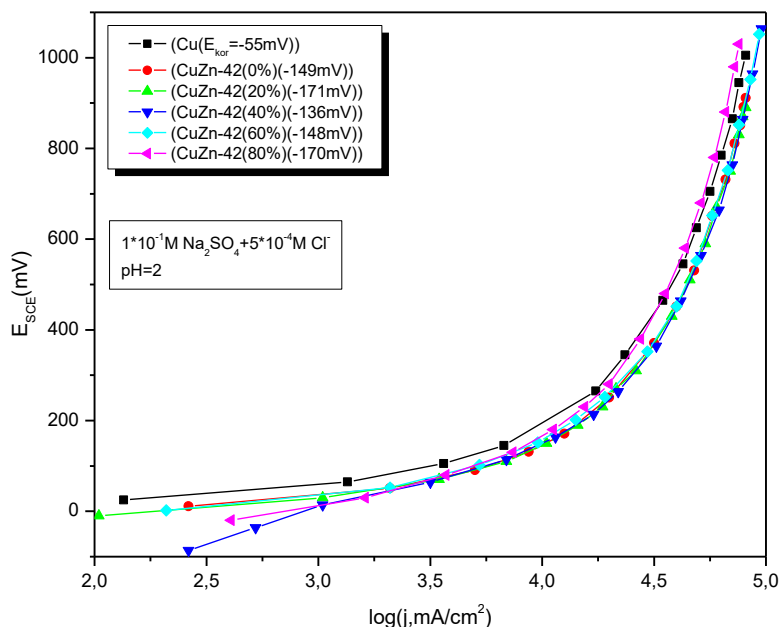


Figure 1. Polarization curves for deformed brass CuZn-42 in a $5 \times 10^{-4} \text{ M Cl}^-$ ion solution at pH 2

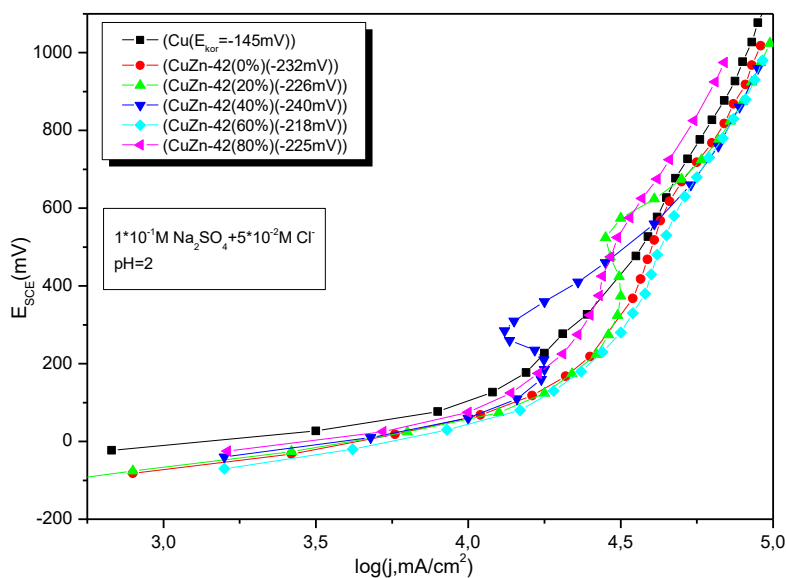


Figure 2. Polarization curves for deformed brass CuZn-42 in a $5 \times 10^{-2} \text{ M Cl}^-$ ion solution at pH 2

The lowest corrosion current density value (Table 2) is observed for the Cu electrode, while for the brass electrodes, the j_{corr} values increase with increasing deformation up to 60%. The lowest

j_{corr} value among the brass samples is found for CuZn-42(80%), which is explained by the inhibitory effect of the increased degree of deformation [35,37,38].

Table 2. Corrosion current density values for CuZn-42 brass in a $5 \times 10^{-2} \text{ M Cl}^-$ ion solution at pH value 2

1·10 ⁻¹ M Na ₂ SO ₄ + 5·10 ⁻² M Cl ⁻							
Sample		Cu	CuZn-42 (0%)	CuZn-42 (20%)	CuZn-42 (40%)	CuZn-42 (60%)	CuZn-42 (80%)
pH=2	j _{corr} (mA/cm ²)	0.678	1.083	1.122	1.182	1.197	1.008

At a chloride ion concentration of $5 \cdot 10^{-2} \text{M}$, the corrosion current density values are lower than those at lower chloride concentrations ($5 \cdot 10^{-4} \text{M}$ and $5 \cdot 10^{-3} \text{M}$) (Table 1). This is due to the inhibitory effect of higher chloride ion concentrations on the brass corrosion process [25,39,40].

During the investigation of the corrosion behavior of the tested brass samples in a solution with a chloride ion concentration of $5 \cdot 10^{-2} \text{M}$, the presence of current peaks was observed on the anodic polarization curves (Figure 2). The most pronounced current peak appeared for the CuZn-42(40%) electrode, while for the other tested electrodes, the current peaks were more "spread out" and had a plateau-like shape [41]. A chloride ion concentration of $5 \cdot 10^{-2} \text{M}$ combined with a low pH value represents critical conditions for the appearance of current peaks, corresponding to the formation of copper oxide and chloro-complex films on the surfaces of the tested samples: CuO, Cu₂O, CuCl, CuOCl [23]. At the end of polarization, a white precipitate was observed on all electrodes, indicating the dezincification process [7,10].

Previous studies indicate that chloride ions generally increase the corrosion current density of

60Cu-40Zn brass, especially when moving from low to higher chloride concentrations (e.g., in the presence of benzotriazole, anodic current densities are significantly smaller even with added chlorides [42]. Other work [43] on dezincification shows that alloys with ~60%Cu suffer increased corrosion in higher chloride environments, unless inhibitors or other alloying elements are present (effects of pH, chloride, bicarbonate, and phosphate on brass dezincification). Although these reports do not specifically compare $1 \cdot 10^{-4} \text{M}$ vs $1 \cdot 10^{-6} \text{M Cl}^-$, they support the trend that low chloride concentrations lessen corrosion, and that at some threshold concentration (which in your study appears to be $\sim 1 \cdot 10^{-4} \text{M}$), an inhibitory or plateau effect might occur.

Further increase of the chloride ion concentration to $1 \cdot 10^{-1} \text{M}$, $5 \cdot 10^{-1} \text{M}$, and 1.0M (Figures 3, 4, 5) leads to an increase in current density values compared to lower concentrations ($5 \cdot 10^{-4} \text{M}$, $5 \cdot 10^{-2} \text{M}$). The current peaks on the anodic polarization curves become more pronounced, which is caused by the formation of copper oxide and copper-chloride complex films on the surface of the tested samples.

Table 3. Corrosion current density values for CuZn-42 brass in a $1 \cdot 10^{-1} \text{M Cl}^-$ ion solution

$1 \cdot 10^{-1} \text{M Na}_2\text{SO}_4 + 1 \cdot 10^{-1} \text{M Cl}^-$							
Sample		Cu	CuZn-42 (0%)	CuZn-42 (20%)	CuZn-42 (40%)	CuZn-42 (60%)	CuZn-42 (80%)
pH=2	$j_{\text{corr}}(\text{mA/cm}^2)$	0.921	1.485	1.56	1.604	1.662	1.475

Table 4. Corrosion current density values for CuZn-42 brass in a $5 \cdot 10^{-1} \text{M Cl}^-$ ion solution

$1 \cdot 10^{-1} \text{M Na}_2\text{SO}_4 + 5 \cdot 10^{-1} \text{M Cl}^-$							
Sample		Cu	CuZn-42 (0%)	CuZn-42 (20%)	CuZn-42 (40%)	CuZn-42 (60%)	CuZn-42 (80%)
pH=2	$j_{\text{corr}}(\text{mA/cm}^2)$	1.083	1.553	1.598	1.878	2.269	1.488

Table 5. Corrosion current density values for CuZn-42 brass in a 1.0M Cl^- ion solution

$1 \cdot 10^{-1} \text{M Na}_2\text{SO}_4 + 1.0 \text{M Cl}^-$							
Sample		Cu	CuZn-42 (0%)	CuZn-42 (20%)	CuZn-42 (40%)	CuZn-42 (60%)	CuZn-42 (80%)
pH=2	$j_{\text{corr}}(\text{mA/cm}^2)$	1.442	3.158	3.49	3.752	4.11	3.005

In general, an increase in the degree of brass deformation leads to a rise in corrosion current density values, which is particularly pronounced in the presence of chloride ions, where both higher Cl^- concentrations and higher deformation levels result in increased j_{corr} values. However, in certain cases (e.g., CuZn-42 deformed to 80%), a deviation from this trend is observed, as the lowest

j_{corr} value among the brass samples was recorded. This effect can be explained by the inhibitory influence of extensive deformation, which induces structural changes in the material (grain densification, phase stabilization, or the formation of protective surface films), thereby mitigating the aggressive action of chloride ions. Thus, although increased deformation and chloride concentration

are generally associated with enhanced corrosion activity, extreme deformation levels may locally

exhibit an opposite, inhibitory effect (Tables 3, 4, and 5).

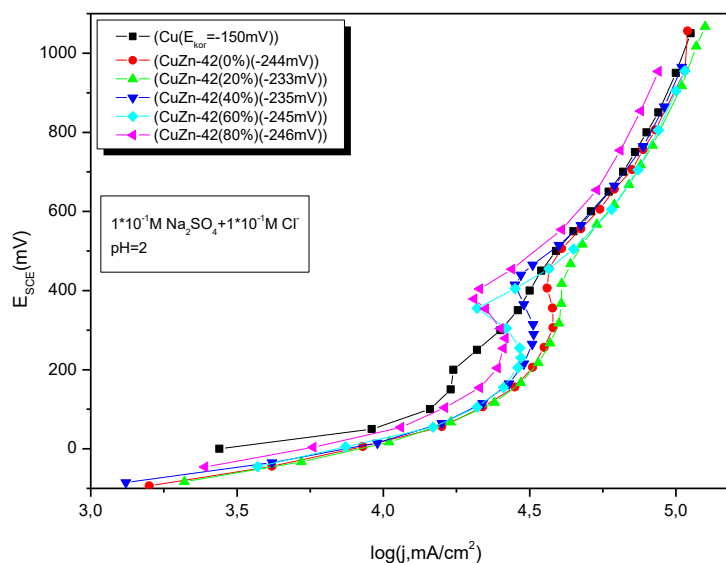


Figure 3. Polarization curves for deformed brass CuZn-42 in a $1 \times 10^{-1} \text{ M Na}_2\text{SO}_4$ solution with the addition of $1 \times 10^{-1} \text{ M Cl}^-$ ions at pH 2

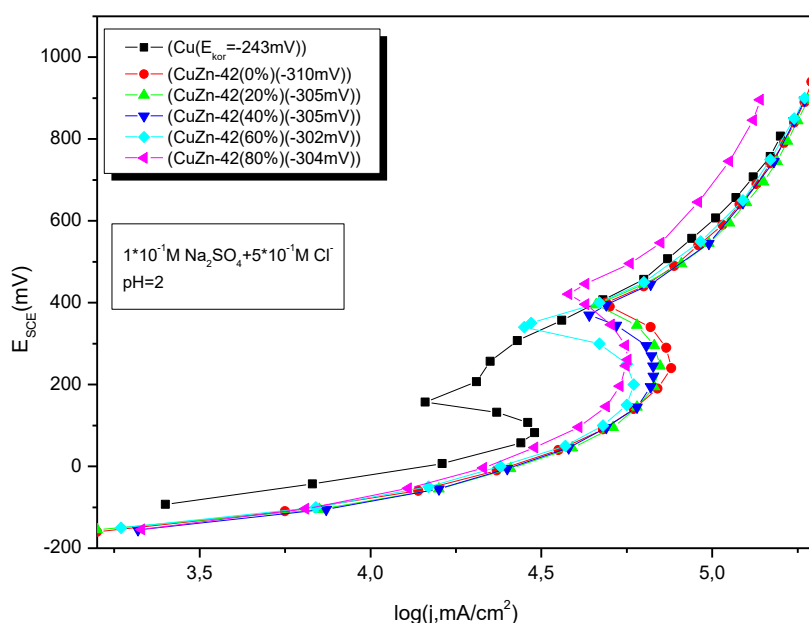


Figure 4. Polarization curves for deformed brass CuZn-42 in a $1 \times 10^{-1} \text{ M Na}_2\text{SO}_4$ solution with the addition of $5 \times 10^{-1} \text{ M Cl}^-$ ions at pH 2

The highest corrosion current density value is observed for brass with a 60% degree of deformation at all tested chloride ion concentrations. The lowest corrosion current density values are found for brass with an 80% degree of deformation and the copper electrode.

Compared to the solution with a chloride ion concentration of $5 \cdot 10^{-3} \text{ M}$ (Table 1), in the solution

with a chloride ion concentration of $1 \cdot 10^{-1} \text{ M}$ (Table 3), higher j_{corr} values are observed for the following CuZn-42 brass electrodes: 0% and 80%, while lower j_{corr} values are found for the copper electrode and the CuZn-42 brass electrodes with 20%, 40%, and 60% deformation. This means that these four electrodes are inhibited even at a chloride ion concentration of $1 \cdot 10^{-1} \text{ M}$.

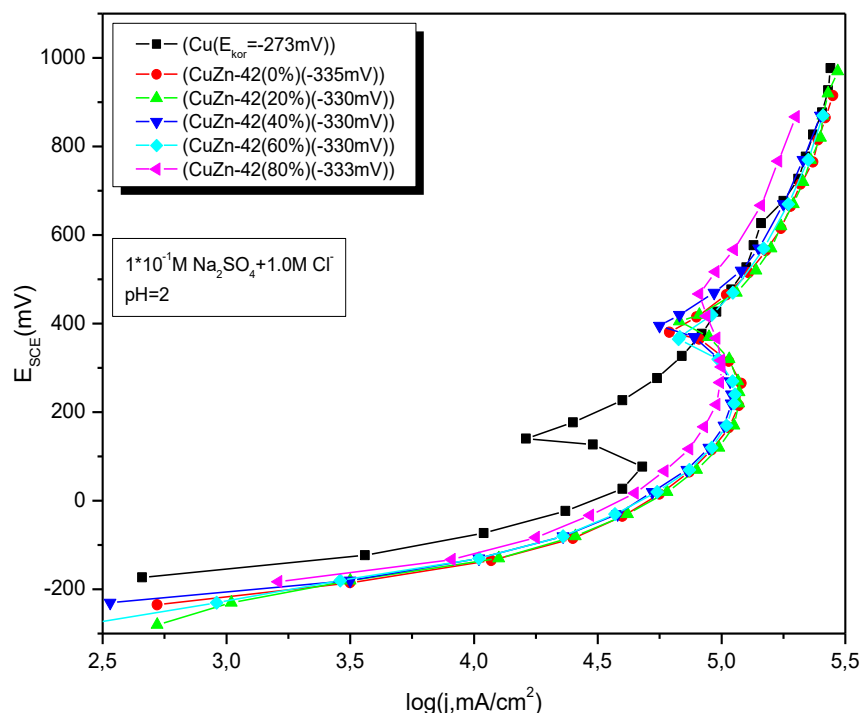


Figure 5. Polarization curves for deformed brass CuZn-42 in a $1 \times 10^{-1} \text{M Na}_2\text{SO}_4$ solution with the addition of 1.0M Cl^- ions at pH 2

4. CONCLUSION

Dezincification was observed at all tested chloride ion concentrations. The presence of a white precipitate (ZnO) and dark discolorations on the brass surface clearly indicate this process. Anodic polarization curves show that brass dissolution starts primarily with zinc and is followed by the simultaneous dissolution of zinc and copper, consistent with earlier findings that dezincification proceeds by selective removal of the more reactive component.

For CuZn-42 brass with 80% deformation, the lowest corrosion current density values were recorded, pointing to the inhibitory effect of high deformation. In contrast, the highest j_{corr} values were obtained at 60% deformation, confirming that deformation up to a certain level enhances corrosion activity.

The influence of chloride ions showed a dual effect. At lower concentrations, they promoted corrosion and dezincification, while at higher concentrations ($5 \cdot 10^{-2} \text{M}$ and $1 \cdot 10^{-1} \text{M}$) signs of inhibition were observed, likely due to changes in the stability of the Cu_2O film. Under these conditions, a complex surface layer composed of copper oxide, copper chloride, and zinc oxide was formed on the brass surface.

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IZVOD

HLORIDNI JON - AKTIVATOR I PASIVIZATOR KOROZIJE

Mesing, legura bakra i cinka, široko se koristi u različitim industrijama zbog svojih povoljnih mehaničkih svojstava, dobre obradivosti i otpornosti na koroziju u mnogim sredinama. Međutim, prisustvo određenih jona u sredini, naročito hloridnih jona (Cl^-), može značajno uticati na koroziono ponašanje ove legure. Hloridni joni su poznati po svojoj sposobnosti da narušavaju pasivne slojeve na površini metala i legura, što može dovesti do lokalizovane korozije kao što su tačkasta korozija i decinkacija – selektivno uklanjanje cinka iz legure, što može ozbiljno ugroziti njenu strukturalnu stabilnost. Takođe, u pojedinim slučajevima hloridni jon može odigrati i ulogu pasivatora korozionih procesa, što zavisi od njegove koncentracije, sastava materijala koji se ispituje, kao i pH-vrednosti rastvora. Iz navedenog, razumevanje mehanizama korozije mesinga u prisustvu hloridnih jona od suštinskog je značaja za njegovu sigurnu i dugotrajnu primenu, posebno u sistemima za transport vode, sanitarnoj opremi i pomorskoj industriji, gde su ovi joni često prisutni. Ovaj rad se bavi ispitivanjem faktora koji utiču na intenzitet i vrstu korozije mesinga u hloridnim sredinama, sa posebnim osvrtom na elektrohemijske metode ispitivanja i morfologiju korozionih proizvoda.

Ključne reči: mesing, korozija, decinkacija, hloridni joni

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