EFFECT OF SILICON ADDITION ON DEZINCIFICATION OF L72 BRASS IN CHLORIDE MEDIA

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Abstract

Brass alloys (Cu-Zn) are susceptible to dezincification, especially in chloride-rich environments. This study investigates the effect of adding 0.5% silicon to L72 brass on its corrosion behavior and resistance to localized corrosion induced by microcracks. Samples with and without silicon were immersed in a 3.5% NaCl solution and analyzed using electrochemical testing, EDX techniques, and dezincification coefficient (z) measurements. The results show that silicon addition improves corrosion resistance by increasing the corrosion potential, reducing current density, and minimizing zinc loss. OM and EDX analyses confirmed that the silicon-containing alloy exhibited a smoother surface and higher copper content. However, microstructural observations revealed that silicon can alter grain boundaries and promote microcrack formation, which may initiate localized corrosion. These findings suggest that while silicon enhances overall dezincification resistance, it may also introduce structural vulnerabilities, highlighting the need to optimize silicon-alloyed brass compositions to improve corrosion resistance while minimizing microcrack-induced degradation in aggressive environments.

Keywords: *Brass alloy; silicon-alloyed brass; dezincification; corrosion resistance*.

1. Introduction

Brass alloys, primarily composed of copper and zinc, are widely used in plumbing [1], marine [2], and industrial [3] applications due to their excellent machinability, corrosion resistance, and mechanical strength. However, in certain environments, brass can undergo dezincification, an issue where zinc is leached from the alloy, which results in a weakened and porous copper structure [4], [5]. This phenomenon significantly diminishes the strength and lifespan of brass components, especially in harsh conditions involving chloride ions, acids, or stagnant water [6]-[8]. Traditionally, strategies to mitigate this issue have focused on altering the alloy composition, such as adding tin, arsenic, or antimony to inhibit the dezincification process [9], [10]. More recently, silicon has been incorporated as an alloying element in brass to improve casting properties, reduce lead content in compliance with environmental regulations, and enhance strength [11]-[13]. Silicon-alloyed brasses are considered more

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environmentally friendly and are increasingly being used as alternatives to leaded brasses. Despite their growing popularity, the corrosion behavior of silicon-containing brass alloys is still not fully understood, particularly regarding localized forms of corrosion like dezincification. Some reports indicate that silicon-alloyed brasses may be more resistant to uniform corrosion, but they might suffer from localized degradation due to microstructural heterogeneities that arise during solidification or service.

One critical yet underexplored factor is the presence of dezincification within the alloy matrix [14]. These dezincifications can form due to mechanical fatigue, casting shrinkage, or thermal stress during operation. They serve as preferential sites for corrosion initiation, allowing aggressive species like chloride ions to penetrate the material, bypassing protective oxide films and accelerating localized corrosion. Once initiated, dezincification can propagate along these cracks, undermining the structural integrity of the component. In this study, we investigate the dezincification corrosion in a silicon-alloyed brass system. By examining the metal structure, testing electrical properties, and analyzing the surface, we explore how microcracking influences the initiation and progression of dezincification. The findings of this study aim to deepen the understanding of failure mechanisms in silicon-alloyed brass components and provide guidance for improving the corrosion resistance of brass alloys under real-world service conditions.

2. Experiments

2.1. Materials and sample preparation

In this study, L72 brass alloy was chosen as the base material to investigate the influence of silicon on dezincification corrosion. L72 is a commercial brass alloy containing approximately 72 wt.% copper and 28 wt.% zinc, known for its good corrosion resistance and mechanical formability. To study the effect of silicon content, silicon was incrementally added to the L72 alloy at concentrations of 0%, 0.1%, 0.2%, 0.3%, 0.4%, 0.5%, and 0.6% by weight.

The alloys were prepared by melting old Russian L72 artillery shell (grade 5) and Cu-16%Si intermediate alloy (self-made) at $T = 1020^{o}C$ for 1 hour in an induction furnace under ambient atmosphere (Use charcoal and flux to prevent oxidation and remove impurities). The molten alloy was then cast into cylindrical steel molds (20 mm in diameter and 100 mm in length) at $T = 1120^{o}C$ and allowed to cool in air. All ingots were subsequently homogenized at 650°C for 4 hours to ensure uniform distribution of elements and to reduce casting stress.

Disc-shaped specimens (5 mm thick) were sectioned from the ingots using a precision diamond saw. The samples were ground sequentially with silicon carbide papers (grades 400 to 2000) and polished with 1 µm alumina slurry until a mirror-like finish was achieved. Finally, the samples were ultrasonically cleaned in ethanol and rinsed with deionized water before testing.

2.2. Chemical composition and Microstructural Characterization

The chemical composition of the brass alloys was determined by emission spectroscopy (Spectrotest TXC 03) according to the method BS EN 150790-15 (Average value of 3 different locations under the same survey conditions, with error ±0.0001).

The microstructure of the silicon-modified L72 alloys was analyzed using optical microscopy (OM) and X-ray diffraction (XRD) analysis was performed using Cu Kα radiation ($\lambda = 1.5406$ Å) over a scanning range of 20° - 90° to identify phases and detect any new intermetallic compounds or secondary phases caused by the addition of silicon. Energy-dispersive spectroscopy (EDS) was used to determine the elemental composition present on the sample surface after immersion in a 3.5 wt.% NaCl for 24 hours (The experiment was performed at 3 different locations, taking the average value to limit the error).

2.3. Electrochemical testing

The electrochemical behavior was studied using a three-electrode setup: the brass sample served as the working electrode (with a 1 cm^2 exposed area), a platinum electrode served as the counter electrode, and a saturated calomel electrode (SCE) was used as the reference electrode. The electrolyte was a freshly prepared 3.5 wt.% NaCl aqueous solution.

The samples were stabilized in the electrolyte for 60 minutes at open circuit potential (OCP). Potentiodynamic polarization was conducted at a scan rate of 1 mV/s, ranging from -0.8 V to +0.3 V versus OCP. Electrochemical impedance spectroscopy (EIS) was performed at OCP over a frequency range of 10^5 Hz to 10^{-2} Hz using a 10 mV amplitude sine wave (Each experiment was repeated 3 times, taking the average value to limit the error).

2.4. Dezincification testing

To assess dezincification resistance, all samples were subjected to accelerated corrosion testing in accordance with ISO 6509-2:2017 (Corrosion of metals and alloys - Determination of dezincification resistance of copper alloys with zinc). Each specimen was immersed in a 1% $CuCl_2$ solution at 75 ± 2°C for 24 hours. After exposure, the samples were cross-sectioned and analyzed using OM and EDS to determine the depth of dezincification zones, quantify zinc loss, and observe any localized attack associated with microcracks (Each experiment was repeated 3 times, taking the average value to limit the error). Dezincification coefficient is evaluated through the following formula (1):

$$Z = \frac{\left(\dfrac{m_{Zn^{2+}}}{m_{Cu^{2+}}}\right)_{solution}}{\left(\dfrac{m_{Zn}}{m_{Cu}}\right)_{alloy}} \qquad (1)$$

3. Results and discussion

3.1. Chemical composition and hardness

The chemical composition and hardness data of L72 brass and its silicon-alloyed variants (LK72-x) reveal that the addition of silicon from 0.1% to 0.6% has minimal effect on the overall copper and zinc content, which remain relatively stable around 71.5-71.8 wt.% Cu and 27.5-27.9 wt.% Zn (Tab. 1). Even with the small increase in silicon, the Brinell hardness (average of three measurements) of the alloys stays about the same, going from 73 HB in the basic L72 alloy to 71 HB in the samples with 0.5-0.6% Si. This suggests that silicon additions in the studied range do not markedly affect the bulk hardness of the alloy. However, changes in the material's structure-like smaller grain sizes and uneven distribution of elements-seen in samples with silicon might affect other properties that hardness does not measure, such as how likely the material is to crack or corrode in specific areas. The "Remain" fraction increases slightly with higher Si content, likely due to undissolved silicon-rich phases or inclusions forming during casting. Even though the mechanical hardness stays the same, silicon can change the structure and behavior of the material, which might help resist dezincification but could also lead to microcracks if there is too much segregation between the grains. Thus, although Si alloying in this range appears mechanically neutral in terms of hardness, it likely plays a more significant role in corrosion resistance.

Tab. 1. Chemical composition (Wt%) and hardness of brass alloys alloyed with Si

Sample	**Cu**	**Zn**	**Si**	**Fe**	**Mn**	**Ni**	**Remain**	**Hardness, HB**
L72	71.7993	27.7006	-	0.0251	-	-	0.4750	73
LK72-0.1	71.6977	27.9015	0.1008	0.0190	0.0040	0.0005	0.2765	73
LK72-0.2	71.6996	27.7989	0.2015	0.0169	0.0031	0.0004	0.2796	73
LK72-0.3	71.5980	27.6995	0.3025	0.0161	0.0029	0.0004	0.3806	72
LK72-0.4	71.5008	27.7998	0.3991	0.0132	0.0021	0.0003	0.2847	72
LK72-0.5	71.5971	27.5018	0.5010	0.0151	0.0020	0.0002	0.3828	71
LK72-0.6	71.4895	27.5997	0.6105	0.0142	0.0011	0.0001	0.2849	71

3.2. Microstructural characterization

Figure 1 displays optical microscopy images of L72 brass samples, L72 (a) and LK72-0.5 (b), revealing distinct differences in their microstructures due to the presence of silicon. In Fig. 1a, the microstructure of the L72 brass without silicon exhibits larger, more uniform grains with smooth and well-defined grain boundaries. This type of microstructure is typical for heated α-brass alloys, indicating that it can bend easily but may also be more susceptible to zinc loss at the edges of the grains when exposed to corrosive environments. In contrast, Fig. 1b shows the microstructure of LK72-0.5 alloy, where the addition of silicon results in smaller grains and more irregular, jagged grain boundaries. The grain structure appears uneven in certain areas, with dark spots likely caused by silicon-rich particles observed along the edges of the grains. These characteristics suggest that silicon influences the material's cooling and solidification processes, potentially leading to uneven cooling or reduces grain growth during crystallization. The smaller grains and increased space between the grains in the silicon-alloyed sample may enhance its strength by improving the performance of the grain boundaries. However, the uneven distribution of silicon can create stress concentration points and potential differences between the grains and the grain boundaries, making the material more susceptible to cracking or dezincification, especially when exposed to corrosive environments over extended periods (inducing intergranular corrosion through a galvanic corrosion mechanism).

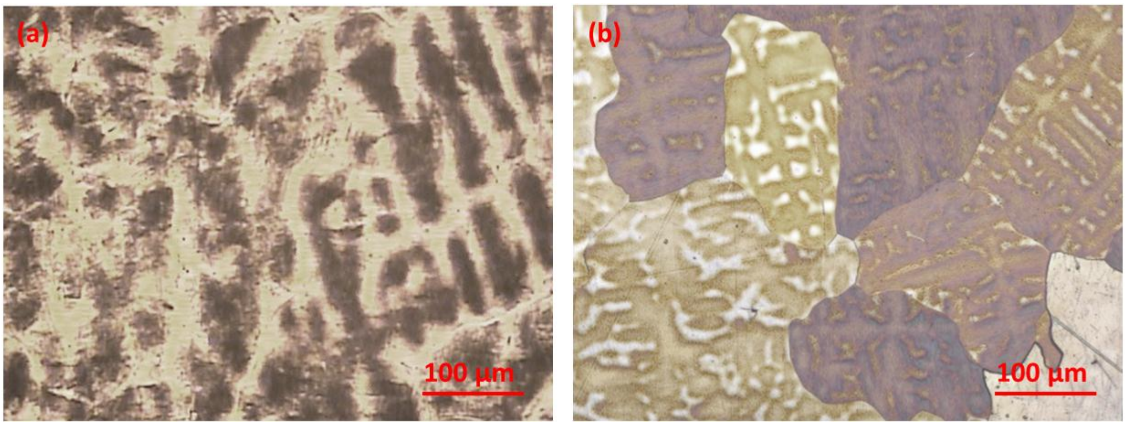

Fig. 1. Optical microscopy images of L72 brass without (a) and LK72-0.5 (b).

The X-ray diffraction patterns, it can be observed that both L72 and LK72-0.5 samples exhibit a face-centered cubic (FCC) lattice structure characteristic of α-brass alloys. The addition of 0.5% Si does not generate a new phase but induces lattice distortion, leading to a reduction in peak intensity and a broadening of the diffraction peaks at d_1 = 2.1245 Å, (111) plane, and d_2 = 1.8420 Å, (200) plane and compared with the JCPDS, copper file No. 04-0836 and ASTM 03-1005- face-centered cubic copper phase - standard diffraction card. This indicates that Si acts as a solid solution alloying element, contributing to grain refinement and potentially enhancing certain mechanical properties, such as strength.

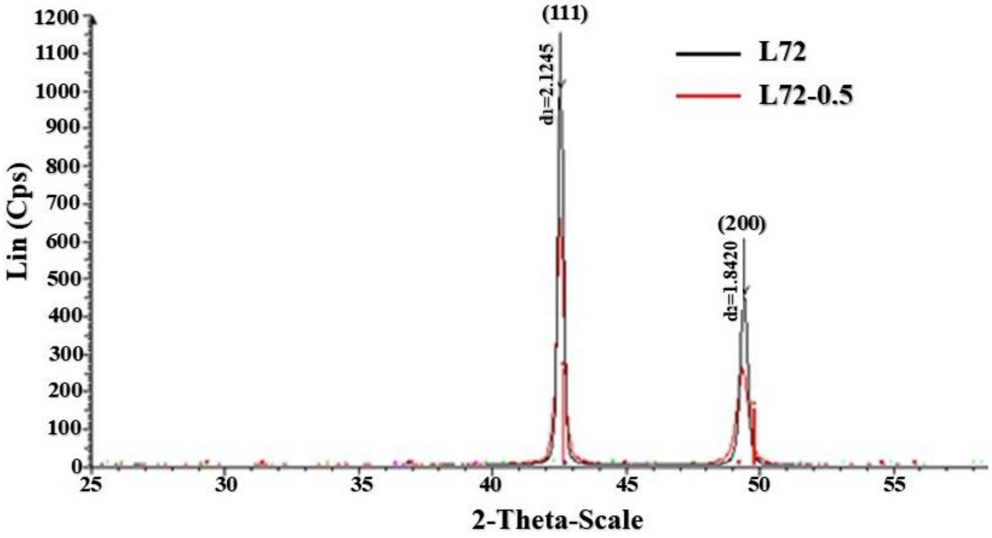

Fig. 2. X-ray diffraction patterns of samples L72 and LK72-0.5.

3.3. Electrochemical testing

The polarization curves in Fig. 3 show that brass without silicon (dash curve) has a higher corrosion current density (j_{corr}) and lower corrosion potential (E_{corr}), indicating greater susceptibility to corrosion than the silicon-containing alloy.

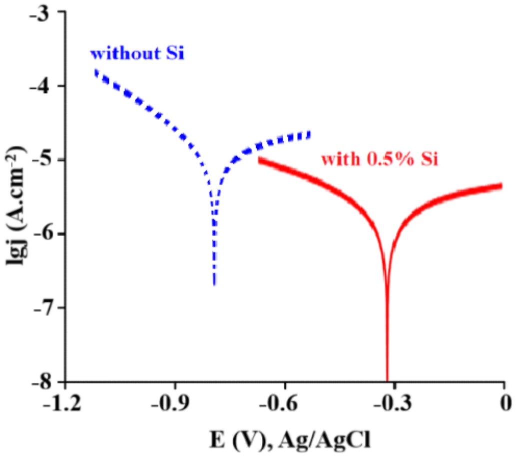

Fig. 3. Polarization curves of L72 (dash curve) and LK72-0.5 (continuous curve).

In contrast, the continuous curve, which represents LK72-0.5 brass, indicates a shift in E_{corr} to higher values and a significant decrease in j_{corr}. This data suggests that the addition of silicon enhances the corrosion resistance of the brass. Furthermore, the continuous curve displays a more stable region where the current increases gradually at higher positive voltages. This stability is likely due to the formation of a protective silicon-rich layer, which helps prevent further metal dissolution. Overall, the polarization behavior demonstrates that the inclusion of silicon in brass makes it more resistant to corrosion by reducing the rate at which it deteriorates and maintaining a stable surface against metal loss.

The electrochemical parameters and corrosion rates indicate that adding 0.5% silicon significantly enhances the corrosion resistance of brass. The sample L72 shows a more negative corrosion potential (E_{corr} = -0.801 V), a higher corrosion current density (j_{corr} = 1.340 × 10^{-5} A·cm^{-2}), and a higher corrosion rate (v_{corr} = 1.729 × 10^{-4} mm/year), which reflects a greater tendency for electrochemical dissolution. In contrast, the sample LK72-0.5 exhibits a more positive corrosion potential (E_{corr} = -0.324 V), suggesting improved thermodynamic stability against corrosion. Its corrosion current density decreases significantly to 5.566 × 10^{-6} A·cm^{-2}, and the corrosion rate drops to 7.180 × 10^{-5} mm/year, representing a nearly 59% reduction compared to L72. This considerable decrease in jcorr and vcorr indicates that the addition of silicon effectively suppresses the corrosion process, likely by forming a more protective surface film or modifying the structure of the passive layer. The positive shift in E_{corr} further supports the conclusion that silicon improves the electrochemical stability of brass in corrosive environments. Overall, the data demonstrate that LK72-0.5 alloy significantly enhances its corrosion resistance by reducing the corrosion current, shifting the potential toward more noble values, and decreasing the degradation rate of the material. This makes the brass more suitable for applications that require enhanced durability in aggressive conditions.

Tab. 2. Electrochemical parameters and corrosion rates of brass without and containing 0.5% Si

Sample	**E_{corr}, V**	**j_{corr}, A.cm^{-2}**	**v_{corr}, mm/year**
L72	-0.801	1.340 × 10^{-5}	1.729 × 10^{-4}
LK72-0.5	-0.324	5.566 × 10^{-6}	7.180 × 10^{-5}

3.4. Dezincification testing

In Fig. 4, the OM images samples after being in a 3.5% NaCl solution for 24 hours show different surface appearances for the sample L72 (a) and LK72-0.5 (b), which shows how they corrode differently.

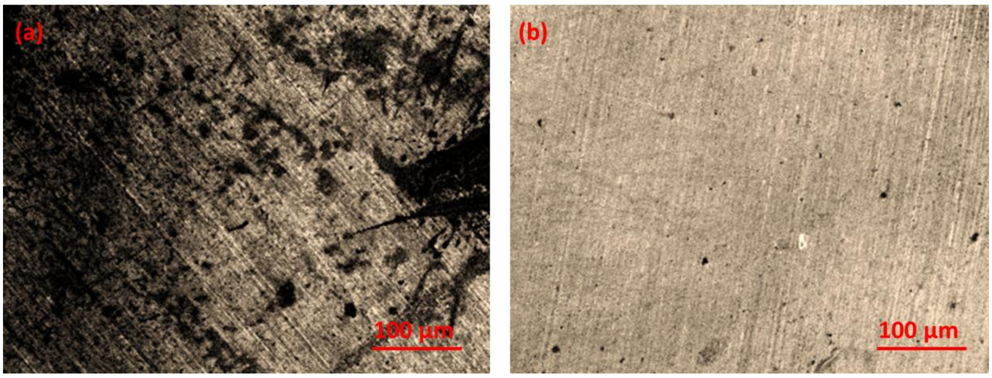

Fig. 4. OM images of L72 (a) and L72-0.5 (b) in NaCl 3.5% solution after 24 hours.

In OM image (a), the surface of L72 alloy looks very damaged, showing many pits, cracks, and loose corrosion materials, which means it has been heavily attacked by chloride ions. The rough and uneven texture suggests that the passive film, if formed, is unstable and readily broken down in the saline environment. In image (b), the surface of LK72-0.5 looks much smoother and more solid, with fewer corrosion pits and a better overall shape. The presence of silicon appears to enhance the stability and protective quality of the surface layer, reducing its susceptibility to pitting and uniform corrosion. The less damaged surface backs up the previous electrochemical data, which showed that the Si-containing brass had a lower corrosion current density and rate. This indicates that silicon helps create a stronger and more protective layer on the surface or changes the structure of the alloy in a way that makes it harder for chloride ions to get through. Overall, the analysis shows that adding 0.5% Si to brass helps reduce surface damage in environments with a lot of chloride, making it more resistant to both specific and overall corrosion.

The EDX spectra of brass samples in a 3.5% NaCl solution after 24 hours show noticeable differences in surface makeup between the sample L72 and LK72-0.5, suggesting they corrode differently. In the L72 spectrum, there are strong signals for copper (Cu), zinc (Zn), oxygen (O), and chlorine (Cl), with particularly high amounts of Cl and O showing that corrosion products and chloride-containing oxides have formed.

In the EDX spectrum of LK72-0.5, there are also signals for Cu, Zn, O, and Cl, but the Cl signal is not as strong, and there is a lot of silicon (Si), showing that Si has been added to the alloy successfully. The weaker Cl signal and less strong O peak in LK72-0.5 mean there are fewer or less harmful corrosion products, suggesting better protection against chloride damage. The Si peak indicates that silicon might help create a stronger and protective oxide layer, which keeps Cl^- from getting in and lowers corrosion. Furthermore, the OM image of LK72-0.5 supports this, showing a relatively smoother and more compact surface morphology compared to L72. After 24 hours in a 3.5% NaCl solution, the chemical makeup of brass samples shows clear differences between the L72 and LK72-0.5 in Tab. 3. L72 has more zinc (48.89%) and less copper (24.56%), which means zinc is being washed away, and it also has more oxygen (20.21%) and chlorine (6.34%), indicating that zinc-rich compounds are forming. In comparison, LK72-0.5 has a lot more copper (46.28%), less zinc (30.16%), lower oxygen (14.15%), and more chlorine (9.25%), plus a small amount of silicon (0.16%). These results suggest that adding silicon helps to decrease the loss of zinc and encourages the creation of a more stable, copper-rich protective layer, which improves resistance to corrosion in salty environments. Overall, the EDX analysis shows that adding 0.5% Si helps reduce corrosion by changing the surface chemistry and preventing the creation of harmful chloride-related damage, which matches the better electrochemical performance seen in the polarization and corrosion rate data.

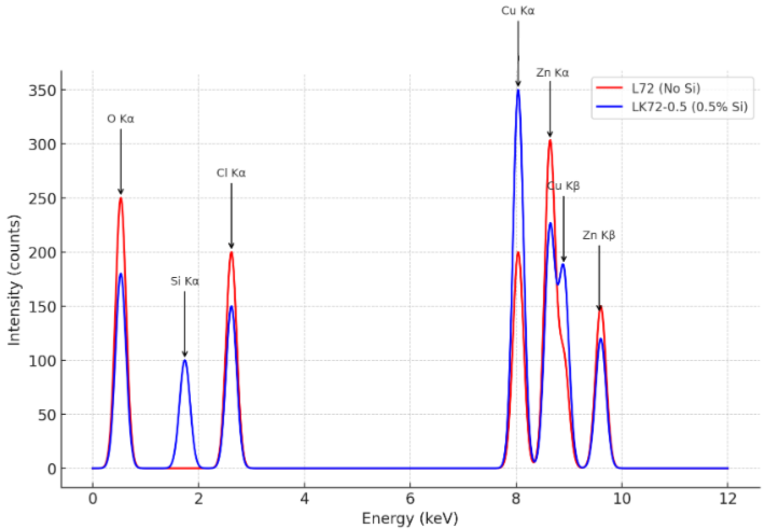

Fig. 5. EDX spectra of brass samples in a 3.5% NaCl solution after 24 hours.

Tab. 3. Surface's chemical composition of brass without and containing 0.5% Si in NaCl 3.5% solution after 24 hours

Sample	**Cu**	**Zn**	**Si**	**O**	**Cl**
L72	24.56	48.89	-	20.21	6.34
LK72-0.5	46.28	30.16	0.16	14.15	9.25

The presence of oxygen and chlorine elements indicates that the corrosion products may be a mixture of oxides, and the odors are mainly copper and zinc. Zn selectively dissolves into a metal surface from two brass samples, L72 and LK72-0.5, enriching the area where Zn dissolves with Cu. The corrosion process produces its products at the sites where Zn dissolves. The products of the corrosion process include the various salts of Cu and Zn, which some research works have calculated to include Cu_2O, $CuCl$, $CuCl_2$, $Cu_2(OH)_3Cl$, $Zn_5(OH)_8Cl_2Zn(OH)_2$,… [6], [8]. There are two types of structures that contain Zn in each sample because of the corrosion products that form a layer over the Cu in the alloy. The corrosion process can be summarized as follows:

$$CuZn \rightarrow Cu^* + Zn^{2+} + 2e \tag{2}$$

$$2Cu + H_2O \rightarrow Cu_2O + 2H^+ + 2e \tag{3}$$

$$Cu_2O + H_2O \rightarrow 2CuO + 2H^+ + 2e \tag{4}$$

$$Cu + 2Cl^- \rightarrow CuCl_2^- + e \tag{5}$$

$$Cu^+ + 2Cl^- \rightarrow CuCl_2^- \tag{6}$$

$$Zn^{2+} + 2Cl^- \rightarrow ZnCl_2 \tag{7}$$

Tab. 4. Dezincification coefficient z of brass without and containing 0.5% Si in NaCl 3.5% solution

Time **Sample**		**1 day**	**2 days**	**5 days**	**10 days**
L72	Cu	1.303	1.841	2.008	3.471
	Zn	1.252	2.602	4.073	7.367
	z	2.491	3.664	5.258	5.703
LK72-0.5	Cu	0.213	0.337	0.381	0.481
	Zn	0.101	0.484	0.914	1.226
	z	1.219	1.697	2.691	2.940

The dezincification coefficient (z) shows how much zinc is lost compared to copper and indicates selective corrosion in brass alloys when they are in chloride environments. According to the data, the brass L72 shows a significantly higher z value across all immersion times in a 3.5% NaCl solution, increasing from 2.491 on day 1 to 5.703 after 10 days. This pattern shows that zinc is being lost quickly and increasingly from the alloy, which weakens its strength and ability to resist corrosion. In contrast, the LK72-0.5 exhibits much lower z values, ranging from 1.219 to 2.940 over the same period, indicating a slower and less intense dezincification process. The consistently lower z values for LK72-0.5 indicate that adding 0.5% Si greatly reduces the loss of zinc by improving the protective layer and stabilizing the alloy's structure. Importantly, even though both samples show rising z values over time, the Si-containing brass has a slower increase, showing that silicon is important for reducing the loss of zinc. These results match earlier studies on electrical and surface properties, supporting the idea that adding silicon is a good way to enhance the long-term resistance to corrosion and strength of brass alloys in environments with a lot of chloride.

4. Conclusion

This study demonstrates that adding 0.5% Si to L72 brass significantly improves its resistance to dezincification in saline environments. The positive effects include: (i) enhanced electrochemical stability, with a shift in corrosion potential to more noble values and a marked reduction in corrosion current density and rate; (ii) formation of a smoother, less corroded surface with fewer chloride products and a more stable copper-rich layer; and (iii) a consistently lower dezincification coefficient (z), indicating better retention of structural integrity.

On the other hand, the addition of Si also introduces negative effects. Microstructural changes, such as uneven grain boundaries and microcracks, may act as preferential sites for localized corrosion initiation.

Overall, Si plays a dual role: while it markedly enhances general corrosion resistance and stability against dezincification, it also requires careful alloy design and processing to minimize the risk of localized damage. These insights are valuable for developing durable, eco-friendly, lead-free brass alloys for service in aggressive environments.

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ẢNH HƯỞNG CỦA BỔ SUNG SILIC ĐẾN QUÁ TRÌNH KHỬ KẼM HỢP KIM ĐỒNG THAU L72 TRONG MÔI TRƯỜNG CLORUA

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Tóm tắt: Hợp kim đồng thau (Cu-Zn) dễ bị khử kẽm, đặc biệt trong môi trường giàu ion clorua. Nghiên cứu này đánh giá ảnh hưởng của việc bổ sung 0,5% silic vào đồng thau L72 đến hành vi ăn mòn và khả năng chịu ăn mòn cục bộ do vi nứt gây ra. Các mẫu có và không chứa silic ngâm trong dung dịch NaCl 3,5% được phân tích bằng các phương pháp đo điện hóa, OM và EDX và xác định hệ số khử kẽm (z). Kết quả cho thấy việc bổ sung silic giúp tăng khả năng chống ăn mòn thông qua việc nâng cao thế ăn mòn, giảm mật độ dòng điện và giảm tổn thất kẽm. Phân tích OM và EDX xác nhận bề mặt hợp kim chứa silic mịn hơn và hàm lượng đồng cao hơn. Tuy nhiên, quan sát vi cấu trúc cho thấy silic có thể làm thay đổi biên hạt và thúc đẩy hình thành vi nứt, từ đó có khả năng khởi phát ăn mòn cục bộ. Những phát hiện này cho thấy mặc dù silic cải thiện khả năng chống khử kẽm tổng thể, nó cũng có thể tạo ra các điểm yếu về cấu trúc, nhấn mạnh việc tối ưu hóa thành phần hợp kim la tông chứa silic nhằm nâng cao khả năng chống ăn mòn, đồng thời hạn chế sự suy giảm do vi nứt trong môi trường khắc nghiệt.

Từ khóa: *Hợp kim la tông; la tông silic; khử kẽm; chống ăn mòn.*

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