

Reviewer A:

Introduction:

While the text mentions that the addition of Zn in CuZn alloys with compositions of 6, 9, and 12 wt.% has not been comprehensively investigated recently, it would be useful to further elaborate on the specific gap in knowledge or application. For instance, it could mention whether there is a lack of research on the combined effects of Zn content on corrosion resistance, antibacterial properties, and mechanical properties, especially in the context of medical transportation (Research Gap Clarification).

The introduction references several studies related to the properties of CuZn alloys (e.g., corrosion resistance, antibacterial properties), but the connection to the specific research focus (medical transportation) could be more explicit. For example, it would be useful to explain how the research will extend current knowledge on CuZn alloys to address issues specific to medical transportation or medical equipment.

The introduction references several studies related to the properties of CuZn alloys (e.g., corrosion resistance, antibacterial properties), but the connection to the specific research focus (medical transportation) could be more explicit. For example, it would be useful to explain how the research will extend current knowledge on CuZn alloys to address issues specific to medical transportation or medical equipment. Suggestion: After mentioning existing research, transition smoothly into explaining why the application of CuZn alloys in medical transportation specifically needs further exploration, and how this research will address that need.

The introduction ends with a mention of the research objectives but does not provide a concluding summary or a clear statement of how the research contributes to advancing the field. A strong concluding sentence would help tie together the information and set the stage for the following sections of the paper.

## Literature review:

While various studies are mentioned, the review does not explicitly highlight a research gap that this study aims to address. For example, it could mention that although there is ample research on the mechanical properties and antibacterial properties of CuZn alloys, there is a lack of comprehensive studies on their combined performance (e.g., how both properties interact or affect each other in specific applications such as medical transportation). Explicitly state the gap in current knowledge or research, especially in the context of this study's aim to investigate CuZn alloys for medical transportation, and how previous research has not sufficiently addressed this aspect.

The literature review mentions many studies but does not clearly tie them to the specific focus of the current research—CuZn alloys for medical transportation. While the properties of CuZn alloys (hardness, antibacterial, corrosion resistance) are discussed, the introduction of medical application as a central theme could be more strongly integrated throughout the review. Tie the studies more directly to the context of medical transportation, by explaining how CuZn alloys with varying Zn content may be suited for specific requirements in medical equipment (e.g., antimicrobial surfaces, durability in a medical environment, etc.).

The review could be more explicit in discussing the novelty of the current research, especially in light of recent studies. For instance, while the studies cited discuss various Zn content ranges and their effects, few studies appear to focus on the specific Zn compositions (6, 9, and 12 wt.%) or applications for medical transportation, which could be a key contribution of this research. Highlight the novelty of the study more clearly. For example, indicate that research on CuZn alloys with Zn compositions in the range of 6–12 wt.% for medical transportation purposes has not been thoroughly investigated, making the current study unique.

The review provides citations from a wide range of studies, but there is no mention of particularly recent or high-impact studies (e.g., from the last 3-5 years) that could demonstrate ongoing trends or breakthroughs in CuZn alloy research. Providing a more up-to-date review could enhance the relevance of the review and the study. Incorporate more recent studies to provide a more current perspective on CuZn alloys, especially research conducted in the last 3-5 years.

The review lacks a conclusive paragraph that ties together the findings from the various studies. A summary of key findings, as well as an explicit statement on how the research builds upon or addresses the gaps in existing knowledge, would help set up the research objectives more clearly. Conclude the literature review with a summary that synthesizes the key findings from the studies and explicitly connects them to the goals of the present research.

#### Research Methods:

While the alloy composition (Cu, Cu6Zn, Cu9Zn, and Cu12Zn) is provided, the exact weight of Cu and Zn used in each alloy should be clarified, especially since Cu is in ingot form and Zn is in powder form. Additionally, it would be useful to mention any prior treatments or cleaning steps for the materials before casting. Specify the exact quantities (in grams or other units) of Cu ingot and Zn powder used for each composition. Also, provide any details on material preparation steps (e.g., cleaning, drying) before casting.

The method of gravity casting is mentioned, but more details on the casting setup could be helpful. For instance, were the Cu and Zn components melted separately before mixing, or were they directly combined in the crucible? What type of crucible was used (material, size, etc.), and what was the cooling rate of the cast samples? Include more detailed information on the casting procedure, including the preparation of the materials, mixing process, crucible specifications, and cooling rate, if available.

XRD Measurement: Provide additional information on the data analysis process using Highscore software, such as peak identification and phase determination. Mention if any reference materials were used for comparison.

Add details on sample cleaning or any surface treatments that were done prior to polishing. Clarify whether any surface defects were removed to ensure the accuracy of the hardness measurements.

#### Result and discussion:

Interpretation of XRD Data: While the XRD results and crystal structure are discussed well, it would be useful to elaborate on the significance of the observed phases in terms of material properties (e.g., mechanical, corrosion resistance). The statement that "all samples had a cubic crystal structure" should be linked to how this structure relates to the properties of the CuZn alloy, especially in terms of the alloy's performance in real-world applications (e.g., corrosion, strength). Suggestion: Add a brief explanation of why the cubic crystal structure is significant for the alloy's properties, and how it may affect the material's performance in specific applications such as corrosion resistance or mechanical strength.

Provide a clearer explanation of the relationship between crystallite size, microstrain, and material properties like hardness and corrosion resistance. Discuss whether the trends observed are desirable for the specific application of CuZn alloys.

Relate the hardness results to the potential applications of the CuZn alloy, such as its suitability for medical implants, corrosion-resistant parts, or mechanical components. Discuss whether the decrease in hardness with Zn addition is an advantage or limitation based on the alloy's intended use.

Analyze the data from Figure 2 in more detail, including commenting on any trends, anomalies, or unexpected results. Provide a more in-depth interpretation of the hardness values, especially in relation to Zn content.

References to Other Studies: The comparison with previous studies (e.g., Nikhil et al., Ezequiel et al., Nnakwo et al.) is appropriate, but it could be expanded. For instance, is the observed trend in your study consistent with the findings of other researchers, or are there notable differences? Also, what could explain the discrepancies, if any, in the results? Suggestion: Expand the comparison to other studies by addressing whether your findings align or diverge from theirs, and discuss possible reasons for any differences.

The discussion of antibacterial activity is good, but it would benefit from a deeper analysis of the results. For example, the statement about Staphylococcus aureus showing significant reduction after 7 hours and Escherichia coli after 3 hours is helpful, but there could be more explanation as to why this difference exists, beyond the basic membrane structure explanation. Suggestion: Provide a more detailed analysis of why Staphylococcus aureus and Escherichia coli show different inhibition times, discussing the different types of antibacterial activity (e.g., ion release, surface interaction) and how the alloy's properties may influence these activities.

Linking Results to Research Gap: The discussion could benefit from stronger linkage to the research gap outlined in the introduction and literature review. For example, the studies on Zn content in CuZn alloys are mentioned, but there could be more emphasis on how your study fills the gap or provides new insights. Suggestion: Tie the results back to the research gap and highlight how the findings contribute to advancing knowledge in the field, particularly regarding the antibacterial properties of CuZn alloys in medical or industrial applications. Implications of Findings: While the results are discussed in relation to previous studies, the broader implications of these findings for real-world applications are not fully explored. For instance, how do the hardness and antibacterial properties affect the potential use of CuZn alloys in medical devices, valves, or other applications? Suggestion: Discuss the broader implications of your findings in terms of real-world applications, such as medical implants, industrial machinery, or other sectors where CuZn alloys are used.

Recommendation: Revisions Required

#### Reviewer B:

#### Abstract:

Please rewrite your abstract. Currently, it reads too narratively and focuses more on the procedural aspects of the research rather than providing a concise summary of the entire study. The abstract should include the following key points:

- 1. Purpose of the study: Briefly state the aim of your research and explain why the study was conducted.
- 2. Methodology: Outline the methods used to address the research problem.
- Results: Summarize the key findings of your study.
- 4. Implications: Discuss the practical and theoretical implications of your results. How can the findings be applied in both theory and practice?
- 5. Contribution: State the value of your study. What is the unique contribution of your research to the field of study?

## Introduction:

The introduction section needs to be expanded to provide more context and clarity. Please ensure that it includes the following:

- 1. Background: Clearly describe the context of the study and provide an overview of the current state-of-the-art in the field. This section should offer readers an understanding of the existing knowledge and research gaps.
- 2. Problem Statement: Identify the problem your study addresses. Explain why the problem is significant and why readers should be interested in the research.
- 3. Proposed Solution: Once the problem is identified, clearly outline the contribution of your work. Highlight the novel aspects of your research and provide a framework that connects it with the existing literature. You should cite at least 8 recent journal articles to support this section.
- 4. Literature Gaps: Summarize the existing gaps in the literature that your research intends to fill, and present the specific aim of your study.
- 5. Use of International Sources: Please ensure that you use international journal references, not local sources, to strengthen the relevance of your work.

#### Literature Review:

While the literature review covers relevant works, it lacks a critical analysis and synthesis of the literature. It currently reads as a list of studies, but a deeper, more critical evaluation is necessary. Here's how to improve it:

- 1. Critical Analysis: Provide a more comprehensive review of the literature, offering critical commentary and drawing connections between studies to highlight trends, contradictions, or gaps in knowledge. This should not just be a factual listing but a thoughtful analysis of the research.
- 2. Recent References: Include more recent journal articles (published from 2019-2024) to ensure the literature review reflects the current state of the field. This will also help support the argument for the novelty and relevance of your research.
- Theoretical Framework: The review should present the theoretical framework underpinning your study. Explain how the selected literature connects with your research questions and objectives.
- 4. Selection of Literature: Describe how the literature was selected for inclusion. Explain the criteria used to ensure a comprehensive and representative review of the state of the art.

# Methodology:

Please clarify the methodology section by addressing the following:

- 1. Justification of Methods: Avoid conceptual descriptions and instead explain why each method was chosen for your study. Discuss the rationale behind selecting specific approaches, tools, and techniques.
- 2. Participants and Characteristics: Provide detailed information about the participants (if applicable) and their characteristics. Who were the subjects, and why were they chosen for this research?
- 3. Data Collection and Instruments: Describe the data to be collected and the instruments used to gather that data. Include an explanation of how the instruments were applied to ensure reliability and validity in your results.
- 4. Explanation of Images: If any images are included in the methodology, add an explanation of what information can be gleaned from them.

#### Results and Discussion:

- 1. Discussion Structure: The results and discussion sections should be presented in continuous prose, without numbering or bullet points. Ensure that there is a smooth flow between sections, with connecting sentences to guide the reader.
- 2. Comparison with Previous Studies: Your discussion should compare your findings with previous research. Please include at least 15 relevant studies from international journals. This will help contextualize your results and demonstrate how your research fits within the broader body of knowledge.
- 3. Generalizations: Conclude the discussion with a broader perspective, offering generalizations or conclusions based on your findings.

## Conclusion:

The conclusion should summarize the main outcomes of the study without using numbered points. It should reflect on the implications of your findings, both theoretical and practical. Highlight the significance of your study and why it contributes to the existing literature.

# References:

- 1. Minimum Number of References: Include at least 40 references, with a minimum of 80% sourced from SCOPUS, Web of Science (WoS), or other reputable international journals.
- Citations within the Text: Ensure that all references are properly cited within the text. Avoid listing references without citation. Use a reference management tool like Mendeley to ensure proper formatting and consistency.
- 3. Recent Sources: Prioritize recent journal articles (preferably from 2019 onward) to maintain the relevance and currency of your work.

Recommendation: Revisions Required

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# EFFECT OF ZINC ADDITION IN COPPER TO STRUCTURE, HARDNESS, CORROSION, AND ANTIBACTERIAL ACTIVITY

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#### **ABSTRACT**

Brass (CuZn) is widely used today due to better mechanical, thermal, physical, and chemical properties. The present research casts CuZn alloy by adding various Zn (6, 9, and 12 wt.%) to the Cu using gravity casting. Casts of CuZn alloy by adding various Zn to the Cu to investigate optimum composition resulted in the highest inhibition of bacterial activity. In addition, structure, hardness, and corrosion were also investigated. Structure, hardness, corrosion, and antibacterial activity behavior were investigated using XRD, potentiostat, Vickers hardness equipment, and a digital camera. XRD confirmed that CuZn alloy has an alpha phase, and a face-centered cubic (FCC) crystal structure. The rise of the Zn content led to an increase in crystallite size, a decrease in the hardness, and a shift to a more negative OCP potential at 1200 s measurement. The increase of the Zn content to 9 wt.% decreased the corrosion rate. Antibacterial activity observation found that all samples had no diffusion. Moreover, 24-hour post-contact observation found that the sample places removed remained clear of bacteria. The fluid contact test shows that the Cu6Zn sample has better antibacterial performance than others. The Cu6Zn sample successfully inhibited the growth of Escherichia coli in the 3rd hour, while Staphylococcus aureus was 100 % reduced in the 7th hour. The Cu6Zn sample could be used as an alternative material for medical equipment in ambulances.

Keywords: XRD, Vickers, Electrochemical measurement, Staphylococcus aureus, Escherichia coli

#### 1. Introduction

Brass (CuZn) is an alloy widely used in the national defense, oil and gas industries, and health because it has better mechanical properties, thermal conductivity, and corrosion resistance (Bhavsar & Bali, 2023; Wang et al., 2023; Widyastuti et al., 2023). CuZn alloys for medical equipment in transportation such as ambulances need to consider two parameters: corrosion resistance and antibacterial characteristics. Commonly, NaCl media was used to investigate corrosion for ambulance equipment due exposure from medical patients eccrine sweat and saline-infused for humans (Baker & Wolfe, 2020; Tayyab et al., 2021). Furthermore, some studies found that after cleaning the ambulance, 35.37 % of bacterial contaminants were still seen (Syamsuir et al., 2023). Viegas et al. found *Staphylococcus aureus* was detectable on firefighter's ambulance equipment (Viegas et al., 2021). *Staphylococcus aureus* is a type of bacteria that could cause skin disease and is hard to treat with traditional antibiotics. *Staphylococcus aureus* bacteria tend to have methicillin resistance. According to Tajik et al., 38.4 % of *Staphylococcus aureus* methicillin resistant was found in Tehran community (Tajik et al., 2020). Moreover, this bacteria also could contaminate orthopedic implants and cause serious infections (Pietrocola et al., 2022).

Several researchers were interested in investigating the corrosion behavior of CuZn alloy in NaCl medium. Abed and Dawood investigated the corrosion behavior of Cu40Zn alloy in 3.5% NaCl and found a corrosion rate of around 0.037 mmpy (Abed & Dawood, 2022). Yin et al. investigated the corrosion behavior of Cu alloy in NaCl medium were immersed in different times. More time is immersed, resulting in more corrosion resistance of Cu (Yin et al., 2021). Chen et al. investigated Cu alloy in NaCl medium and found Cu potential around -0.305 V vs SCE and Zn potential around -1.165 V vs SCE (Chen et al., 2024). Gao et al. found that a reduction in thickness (50 to 60 %) of CuZn using cold rolling resulted in a significant decrease in corrosion current from 4.824 to 1.804 μA/cm² (investigating in 3.5 % NaCl) (Gao et al., 2021). Moreover, aluminum (Al) alloy, such as ambulances, is widely used in the transportation sector (Blanco et al., 2022; Vandersluis et al., 2020). Liu et al. have found Al alloy corrosion current between 4.868-5.251 A/cm² in a 3.5% NaCl medium (Liu et al., 2020). Comparing the studies of Liu et al. and Gao et al., Al alloy has a higher corrosion current than CuZn (Gao et al., 2021; Liu et al., 2020). Corrosion current significantly influences the corrosion rate, and a rise in the corrosion current would enhance the corrosion rate.

Recently, researchers have been interested in investigating CuZn alloy for medical applications. Azizian et al. investigated CuZn alloys for cardiovascular applications (Azizian et al., 2024). Riaz et al. investigated the structural and biological properties of CuZn alloy for orthopedic applications (Riaz et al., 2024). Moreover, Sabbouh et al. did the sonification of CuZn in an alkali solution to enhance the antibacterial inhibition zone (Sabbouh et al., 2023). Moreover, Syamsuir et al. have investigated the antibacterial activity of *Staphylococcus aureus* by presenting a Cu layer for ambulance equipment (Syamsuir et al., 2023).

The killing mechanism of bacterial activity inseparable from the ions released by the alloy (Qu et al., 2020). Cu<sup>2+</sup> ions could be adsorbed on the cytoplasmic membrane surfaces, then penetrate the bacteria, react with sulfhydryl groups, and cause the cell to die (Zeng et al., 2022). The released Zn<sup>2+</sup> ions could penetrate the cell membrane and cause cell death (Du et al., 2021). Zhang et al. have stated that Cu<sup>2+</sup> and Zn<sup>2+</sup> ions could act as antibacterial agents and inhibit *Staphylococcus aureus* growth (E. Zhang et al., 2021).

According to the literature review, research on CuZn alloys with Zn compositions in the range of 6–12 wt.% for medical transportation purposes has not been thoroughly investigated. CuZn can transform into Cu and Zn ions. As mentioned above, the killing mechanism of bacterial activity depend on Cu and Zn ions. Therefore, the present research casts CuZn alloy by adding various Zn to the Cu to investigate optimum composition, resulting in a higher killing mechanism of bacterial activity. Moreover, different alloy compositions would result in different corrosion behavior and mechanical properties. The present study investigated structure, hardness, corrosion, and antibacterial activity using XRD, Vickers hardness equipment, potentiostat, and digital camera.

#### 2. Literature Review

Many techniques are used to make CuZn alloys, including gravity and investment casting (Hendrawan et al., 2021; Ziat et al., 2020). Gravity casting is simple, inexpensive, and can rapidly fill complex geometry (Huang et al., 2024; Nuryadi et al., 2020). Moreover, in the fabrication of CuZn alloys, one thing needs to be considered to produce specific properties, namely alloy composition. Researchers focused on adding various Zn compositions onto Cu for different purposes. Strzępek et al. investigated the mechanical properties of Cu and alpha brass (Cu2.5Zn and Cu6.5Zn) wire (∅ 3.8 mm). Increased Zn content causes increases in ultimate tensile strength, yield strength and hardness (Strzępek et al., 2019). Situmorang et al. fabricated Cu with various Zn additions (10, 20, 38, and 45 wt.%) and found that the higher the Zn composition, the higher the antibacterial properties (Situmorang et al., 2019). Daniyan et al. found that hardness and tensile strength would increase with increasing Zn content (5, 10, 15, 20, and 30 %) in the alloy (Afolabi Daniyan et al., 2021). Iqbal et al. melted Cu28.7Zn using a furnace and cast to investigate hardness and morphology (Iqbal et al., 2021). Shahriyari et al. added Zn (5, 15, 20, and 30 wt.%), increasing the hardness due to the alloy's rise in the Zn content (Shahriyari et al., 2022). Akhyar et al. melted Cu28.7Zn using a gas furnace and cast to investigate the tensile strength (Akhyar et

al., 2023). Morath et al. have created Zn0.8Cu and Zn1.5Cu using casting methods to investigate the biological aspect of arterial implants (Meng et al., 2024). Azizian et al. have added Cu with compositions 1, 2, and 5 wt.% to CuZn alloy by melting in the induction furnace to investigate microstructure, mechanical properties, and cytotoxicity for cardiovascular application (Azizian et al., 2024). Generally, a CuZn alloy with a Zn composition of less than 37 wt.% would produce a single alpha phase with an FCC crystal structure (Clement & Auger, 2023; Mousavi et al., 2020).

#### 3. Research Methods

#### 3.1 Material Preparation

Sample preparation used the gravity casting method. Cu ingot (98.798 %) and Zn powder (99 %) were cast with the alloy composition CuxZn (x=0, 6, 9, and 12 wt.%, namely as Cu, Cu6Zn, Cu9Zn, and Cu12Zn, respectively). Before melting was conducted, the ingot and the apparatus, such as the crucible, were cleaned using water to avoid impurities and then dried. The Cu ingot was first filled into a silicon carbide crucible (3kg) and then inserted into a muffle furnace. Melting was carried out in a crucible at 1100 °C under atmospheric pressure. After the Cu has melted, remove it from the muffle furnace, mix it with Zn powder, stir it manually, and pour it into a permanent mold. For comparison, Cu ingot was melted and poured into a permanent mold without Zn addition. The as-cast ingots, such as XRD, hardness, electrochemical measurement, and antibacterial activity observation, were cut for further characterization.

#### 3.2 XRD Measurement

XRD was measured using the PANalytical (Cu  $K_{\alpha}1$   $\lambda$ =1.5405980) apparatus. XRD was scanned from 20 to 100°, using step size 0.0217°. The Highscore software was used to refine and collect peak, phase and crystallographic parameters of as-cast samples. By using that software, full width at half maximum (FWHM) are also found. FWHM data is used to calculate crystallite size.

## 3.3 Hardness Measurement

Before testing, samples with  $20\times20\times6$  mm dimensions were polished using silicon carbide up to #3000 grit. Afterward, the polished sample was cleaned using water, followed by alcohol, and then dried using drier equipment. The hardness test was conducted using the Vickers method. An FV-300e hardness test was performed on top of various samples using 1 kg of load. Ten repeatable measurements were conducted.

#### 3.4. Electrochemical Measurement

Two electrochemical measurements were conducted in the present research, such as OCP and LSV, using Digi-Ivy (DY2311) potentiostat in 0.9 % NaCl at room temperature. OCP was scanned until 1200 s using a sampling scan rate of 0.02 s, while LSV was conducted using a scan rate of 1 mV/s. Cu/CuZn samples are used as the working electrode, platinum wire as the counter electrode, and Ag/AgCl as the reference electrode. LSV data was examined using the Tafel extrapolation method to see corrosion potential (Ecorr) and current density (icorr). The corrosion rate could be found by inserting icorr in the following equation (Ahmad, 2006).

Corrosion rate 
$$(mmpy) = K \frac{M \times icorr}{o \times n}$$
 (1)

Where K is corrosion constant (3.27 mmpy), M is atomic weight (g/mol),  $\rho$  is material density (g/cm<sup>3</sup>), and n is the number of electrons involved.

# 3.5 Antibacterial Activity Observation

The sample dimension used for antibacterial activity is  $20\times20\times6$  mm. Before testing, samples with  $20\times20\times6$  mm dimensions were polished using silicon carbide up to #3000 grit. The experimental procedure for antibacterial activity is similar to that of the previous report (Syamsuir et al., 2023). Moreover, the recent study uses *Staphylococcus aureus* ATCC 25923 for Direct contact kill assay (24 hours) and Fluid contact assay (8 hours). In comparison, *Escherichia coli* 25922 was also used in the present study. Afterward, documentation was captured using a digital

camera. In addition, 24-hour post-contact observation and Fluid contact test were also captured using a digital camera.

#### 4. Results and Discussions

#### **4.1 XRD**

The diffraction pattern of Cu/CuZn with (111), (200), (311), and (222) planes can be seen in Figure 1. The four diffraction patterns match the alpha phase, which is shown to align with another study (Heidarzadeh et al., 2022). The acquired XRD data was then analyzed using Highscore software, and the parameters are listed in Table 1. All samples had a face-centered cubic (FCC) crystal structure, indicating that the Cu and Zn atoms dissolve into one another. According to Jinlong et al., the FCC quantity of surface energy is (111)>(001) >(110), and the FCC sample with the (111) plane has the lowest corrosion rate (Jinlong et al., 2016). Moreover, the FCC sample with the preferred orientation of the (111) plane has the highest surface atomic density (Soegijono et al., 2020).

Presenting Zn in the alloy reduces crystallite size from 227.20 to 109.95 nm and increases Zn content from 6 to 12 wt.%, leading to an increase in crystallite size from 109.95 to 316.26 nm. This behavior is similar to Özdemir and Karahan's study that showed Zn in the alloy leads to decreased crystallite size, and an increase in Zn content in the alloy leads to increased crystallite size (Özdemir & Karahan, 2014). Moreover, the microstrains of the as-cast sample are independent of Zn content, which perfectly agrees with Karahan and Özdemir's study ((Karahan & Özdemir, 2014). The smallest microstrain is seen in the Cu9Zn sample.

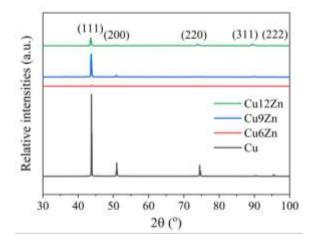


Fig. 1. XRD of various casting samples.

Table 1 - Crystallographic parameters of various casting samples.

Tuble 1 Crystanographic parameters of various custing samples.					
Domormoton	Sample				
Parameter	Cu	Cu6Zn	Cu9Zn	Cu12Zn	
Crystal structure			FCC		
Lattice constant $a = b = c (Å)$	2.964	8.929	2.981	2.964	
Cell volume (Å <sup>3</sup> )	26.03	90	26.49	26.05	
d-spacing (Å)	1.69	1.18	1.69	2.07	
Crystallite Size (nm)	227.20	109.95	204.95	316.26	
Micro strain	0.37	0.53	0.16	0.32	

#### 4.2 Hardness

Figure 2 shows the average hardness of various casting samples. Nikhil et al. have found that pure Cu has a hardness of 140 HV when treated at 400 and 600 °C and then held for two hours, followed by quenching in tap water, resulting in a hardness of 100 and 60 HV (Nikhil et al., 2021). The higher heat treatment temperatures led to a decrease in the hardness of the pure Cu. Therefore, the hardness of Cu may vary depending on heat treatment. In the present study, Cu re-casting has a hardness of 74.54 HV.

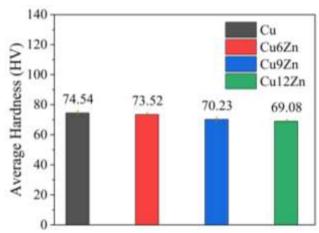


Fig. 2. Average hardness of various casting samples.

According to Figure 2, it can be seen that an increase in Zn content led to a decrease in the hardness. Another research study found that Cu (as-cast) has a hardness of around 100 HV, while Cu15 Zn (as-cast) has a hardness of 75 HV (Ezequiel et al., 2024). Nnakwo et al. also found that increased Zn content in the Cu alloy leads to decreased hardness due to increased in grain size and solid solution region (Nnakwo et al., 2021). According to a study by Qu et al. and García-Mintegui et al., pure Zn hardness is between 41-42 HV (García-Mintegui et al., 2021; Qu et al., 2020). Therefore, it could be concluded that Zn in the alloy leads to decreased hardness due to Zn hardness less than Cu.

Several researchers correlated measured hardness to crystallite size (Augustin et al., 2016; Syamsuir et al., 2023). Augustin et al. have found an increase in Cu's crystallite size, promoting a decrease in scratch and micro-hardness (Augustin et al., 2016). Syamsuir et al. found a decrease in Cu's crystallite size, leading to an increase in hardness (Syamsuir et al., 2023). Comparing Table 1 with Figure 2, it can be seen that an increase in the Zn content led to an increase in crystallite size and a decrease in the hardness. On the contrary, while as-cast samples do not form an alloy (Cu), the resulting hardness is not aligned with the crystallite size. It seems that it cannot compare the crystallite size were found with measured hardness between alloy (CuZn) and unalloy (Cu) material.

Several transportation sectors, such as ambulance equipment, are made from Al alloy (Blanco et al., 2022; Vandersluis et al., 2020). According to Hajizadeh et al., Al alloy hardness is between 32-52 HV (Hajizadeh et al., 2017). Therefore, all specimens have hardness still higher than Al alloy.

#### **4.3 OCP**

Figure 3 shows the OCP measurement result of various casting samples in 0.9 % NaCl at room temperature. Generally, increased Zn content in the alloy promoted more negative potential, which perfectly agrees with the Cocco et al. study (Cocco et al., 2016). Dridi et al. have found that  $E_{\text{OCP}}$  CuZn30 and CuZn39 are -0.578 and -0.604 V/MSE at 3 % NaCl, which means an increase in the Zn promoted to more negative potential (Dridi et al., 2020). Cu, Cu6Zn, Cu9Zn, and Cu12Zn samples have  $E_{\text{OCP}}$  potential at 1200 s measurement -0.014, -0.023, -0.027, and -0.032 V vs Ag/AgCl, respectively.

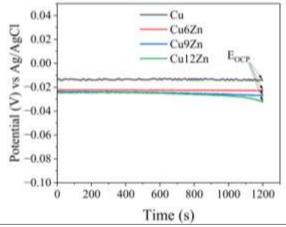


Fig. 3. OCP measurement of various casting samples.

According to Figure 3, Cu, Cu6Zn, and Cu9Zn samples are steady at initial measurements until 1200 s, indicating that the protective layer formed has good protection. In contrast, the Cu12Zn sample is steady at initial measurements until 600 s, then moves in a more negative direction. These phenomena indicated that the formed protective layer had initially dissolved at 600 s; therefore, the measurement continuously moved forward in a negative direction until the measurement reached 1200 s.

#### **4.4 LSV**

Eccrine sweat and saline-infused for humans are nearly 0.9 % of NaCl (Bond & Lieu, 2014; Tayyab et al., 2021). Therefore, LSV measurement was conducted in 0.9 % NaCl at room temperature. Luo et al. found Cu<sub>2</sub>O crystalline growth on the Cu surface when exposed to 0.9 % NaCl, while when exposed to pure water, Cu<sub>2</sub>O crystalline was not seen (Luo et al., 2020). Commonly, Cu<sub>2</sub>O crystallines are formed, which is preceded by the formation of CuCl when the specimen is tested in a chloride solution. Moreover, Zhang et al., in their study, found ZnO and Zn<sub>5</sub>Cl<sub>2</sub>(OH)<sub>8</sub>·H<sub>2</sub>O as corrosion products on top of Cu40Zn surfaces in a chloride environment (X. Zhang et al., 2016). The reaction of Cu in chloride solution is as follows (Milošev et al., 2024).

The reaction of Zn in chloride solution is as follows (Milošev et al., 2024).

$$Zn \to Zn^{2+} + 2e^{-}$$
 (8)  
 $Zn^{2+} + 2OH^{-} \to Zn(OH)_{2} \to ZnO + H_{2}O$  (9)  
 $5Zn^{2+} + 8OH^{-} + 2Cl^{-} + H_{2}O \to Zn_{5}(OH)_{8}Cl_{2} \cdot H_{2}O$  (10)

LSV measurement results in 0.9 % NaCl can be seen in Figure 4.

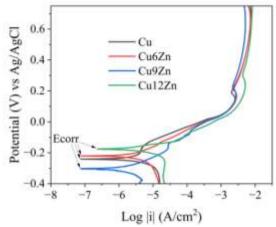


Fig. 4. LSV scans result of various casting samples.

According to Figure 4, corrosion Ecorr and icorr can be found using the Tafel extrapolation method. Moreover, the corrosion rate could be calculated by inserting icorr into expression (1). Table 2 presented Ecorr, icorr, and the corrosion rate of various casting samples. It appears Ecorr is independent of Zn addition; however, Zn is dependent on icorr and corrosion rate (except for the Cu12Zn sample). This behavior is probably due to a protective layer that was formed. Compared to the OCP result, it can be seen that the Cu12Zn sample continuously moves forward in a negative direction from 600 until 1200 s of measurement. Therefore, that sample has a higher icorr and corrosion rate.

Table 2 - Corrosion parameters of various casting samples.

Sample name	Ecorr (V) vs Ag/AgCl	Icorr (A/cm²)	Corrosion rate (mmpy)
Cu	-0.241	$4.42 \times 10^{-6}$	$5.13 \times 10^{-2}$
Cu6Zn	-0.220	$3.33 \times 10^{-6}$	$3.86 \times 10^{-2}$
Cu9Zn	-0.304	$2.15 \times 10^{-6}$	$2.49 \times 10^{-2}$
Cu12Zn	-0.175	$6.19 \times 10^{-6}$	$7.18 \times 10^{-2}$

Qu et al. have found that an increase in Zn content led to an increase in corrosion resistance, which perfectly agrees with the present study (except for Cu12Zn) (Qu et al., 2020). Milošev et al. investigated Cu, Cu10Zn, Cu40Zn, and Zn in 3 % NaCl and found icorr after stabilized at 1 hour around 1.573, 1.456, and 2.114, and 5.21  $\mu$ A/cm² respectively (Milošev et al., 2024). According to equation (1), icorr strongly influences the corrosion rate. The more icorr, the higher the corrosion rate. Moreover, a limitation in Zn content in the Cu alloy could influence the corrosion resistance. Presenting the Zn content  $\leq$  11 wt.% in the alloy could enhance the corrosion resistance; however, Zn of more than 10 wt.% could decrease corrosion resistance (Milošev et al., 2024).

According to Table 1, the Cu9Zn sample has the lowest microstrain than others. The measured microstrain could be associated with the sample's crystal defect (Soegijono et al., 2020). Based on Table 2, the Cu9Zn sample has the lowest microstrain, which confirms that the sample has the lowest corrosion rate. Moreover, the FCC sample with the preferred orientation of the (111) plane could offer a lower corrosion rate due to the highest surface atomic density (Jinlong et al., 2016; Soegijono et al., 2020). Compared to other samples, the Cu9Zn sample has the higher preferred orientation of the (111) plane. Even though the (111) plane of the Cu sample is the highest. Unfortunately, the (220) and (200) planes are still present and relatively high.

# 4.5 Antibacterial Activity

Figure 5 shows the direct contact kill of *Staphylococcus aureus* and *Escherichia coli* after 24 hours of incubation. The present study focused on *Staphylococcus aureus*, but *Escherichia coli* was also used for comparison. There is no diffusion in the sample; therefore, the inhibition zone could not be seen.

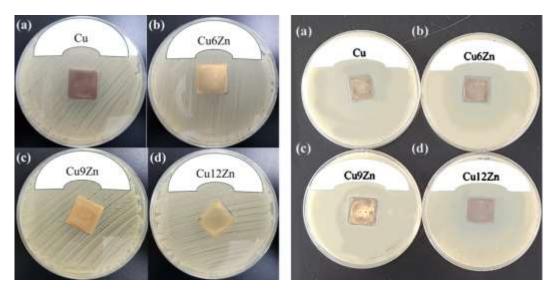


Fig. 5. Antibacterial activity test using *Staphylococcus aureus* (left) and *Escherichia coli* (right) (a) Cu, (b) Cu6Zn, (c) Cu9Zn, and (d) Cu12Zn.

Antibacterial activity after 24 hours of post-contact with various casting samples using *Staphylococcus aureus* and *Escherichia coli* can be seen in Figure 6. The removed sample places remain clear (with no regrowth) from bacterial activity. The antibacterial behavior was significantly influenced by Cu or Zn ions (Qu et al., 2020). Villapún et al. found that releasing Cu ions leads to the highest killing activity of *Staphylococcus aureus* (Villapún et al., 2016). Excess in the Cu ion could be bacteriostatic (Sabbouh et al., 2023). Moreover, Cu ions could be adsorbed on the cytoplasmic membrane surfaces, then penetrate the bacteria, react with sulfhydryl groups and cause the cell to die (Zeng et al., 2022). Furthermore, Cu ions could form hydroxyl groups in the presence of oxygen in nature, which could destroy cell membranes (Dou et al., 2022). Hutchings et al. stated that Zn<sup>2+</sup> successfully inhibits the growth of *S. epidermidis* (Hutchings et al., 2021). This behavior is associated with the generation of reactive oxygen or the formation of (E. Zhang et al., 2021).

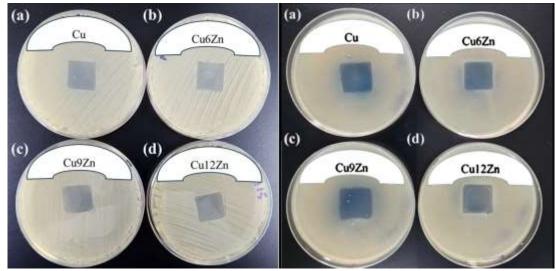


Fig. 6. Antibacterial activity after 24 hours of post-contact (regrowth assessment) towards *Staphylococcus aureus* (left) and *Escherichia coli* (right) (a) Cu, (b) Cu6Zn, (c) Cu9Zn, and (d) Cu12Zn

Figure 7 shows the fluid contact test of *Staphylococcus aureus* and *Escherichia coli*. The orientation of the test materials is mapped within the yellow box. For the Cu6Zn sample, *Escherichia coli* was killed on the 3rd hour. However, there is no significant reduction within the

fluid because there are no diffusible materials. Also, there is no visible growth after 3<sup>rd</sup> hour for the fluid in contact with the metal.

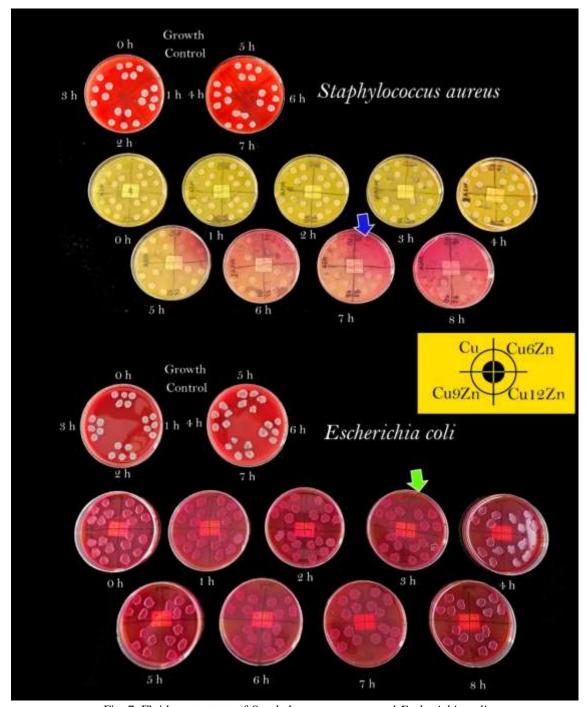


Fig. 7. Fluid contact test of Staphylococcus aureus and Escherichia coli

Moreover, it should be noted that the reduction of the colony is significant in the 7<sup>th</sup> hour for *Staphylococcus aureus* (blue arrow), but for *Escherichia coli*, the inhibition of *Escherichia coli* growth is shown within the 3<sup>rd</sup> hour of contact (green arrow), for Cu6Zn sample. The reduction of colonies is significant on the surface of the metal. While on the remaining fluid, the reduction is insignificant until 8 hours. This behavior is because *Staphylococcus aureus* and *Escherichia coli* have different membrane structures and thick cell walls, therefore could inhibit ion exchange and restrain the antibacterial effect of Cu and or Zn ions (Di et al., 2022). Cu killing is more effective in Gram-negative bacteria (e.g., *Escherichia coli*) because peptidoglycan affects the cell's susceptibility. The thicker the peptidoglycan layer, the harder it became for the Cu ions to

reach the membrane (Xhafa et al., 2023). Therefore, *Escherichia coli* was killed in the 3<sup>rd</sup> hour. Another reason Cu6Zn has better antibacterial performance than others is probably due to its smaller crystallite size. Researchers found that smaller crystallite sizes promote the enhancement of antibacterial effects (Syamsuir et al., 2023). This behavior is attributed to an increase in surface area due to the crystallite size (Sangeetha et al., 2015). The Cu6Zn sample could be used as an alternative material for medical equipment in ambulances.

#### 5. Conclusion

CuZn has been successfully fabricated. XRD confirmed that CuZn alloy has a single alpha phase with an FCC crystal structure. The rise of the Zn content in the alloy led to a decrease in the hardness due to an increase in crystallite size and led to a shift to more negative OCP potential at 1200 s measurement. Moreover, the rise of the Zn content to 9 wt.% decreased the corrosion rate. It appears there is a limitation in Zn content in the copper alloy that influences the corrosion rate, as shown when Zn content around 12 wt. % is promoted to increase the corrosion rate. Antibacterial activity observation found that all samples had no diffusion. Moreover, 24-hour post-contact observation found that sample places removed from the sample remained clear of bacteria. The Cu6Zn has better antibacterial performance than others due to the smallest crystallite size. According to the fluid contact test, the reduction of the colony of *Staphylococcus aureus* is significant in the 7<sup>th</sup> hour. The inhibition of *Escherichia coli* growth is also shown within the 3<sup>rd</sup> hour of contact. This behavior is because *Staphylococcus aureus* and *Escherichia coli* have different membrane structures and thick cell walls, therefore, could inhibit ion exchange and restrain the antibacterial effect of Cu and or Zn ions.

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# [JAETS] Editor Decision

2024-11-19 10:50 AM

Lisa Samura, Mustamina Maulani, Cahaya Rosyidan, Kartika Fajarwati Hartono, Suryo Prakoso, Evi Ulina Margareta Situmorang, Daniel Edbert, Bambang Soegijono, Muhammad Yunan Hasbi, Ferry Budhi Susetyo:

We have reached a decision regarding your submission to Journal of Applied Engineering and Technological Science (JAETS), "EFFECT OF ZINC ADDITION IN COPPER TO STRUCTURE, HARDNESS, CORROSION, AND ANTIBACTERIAL ACTIVITY".

Our decision is to: Accept Submission

<u>Journal of Applied Engineering and Technological Science (JAETS)</u>