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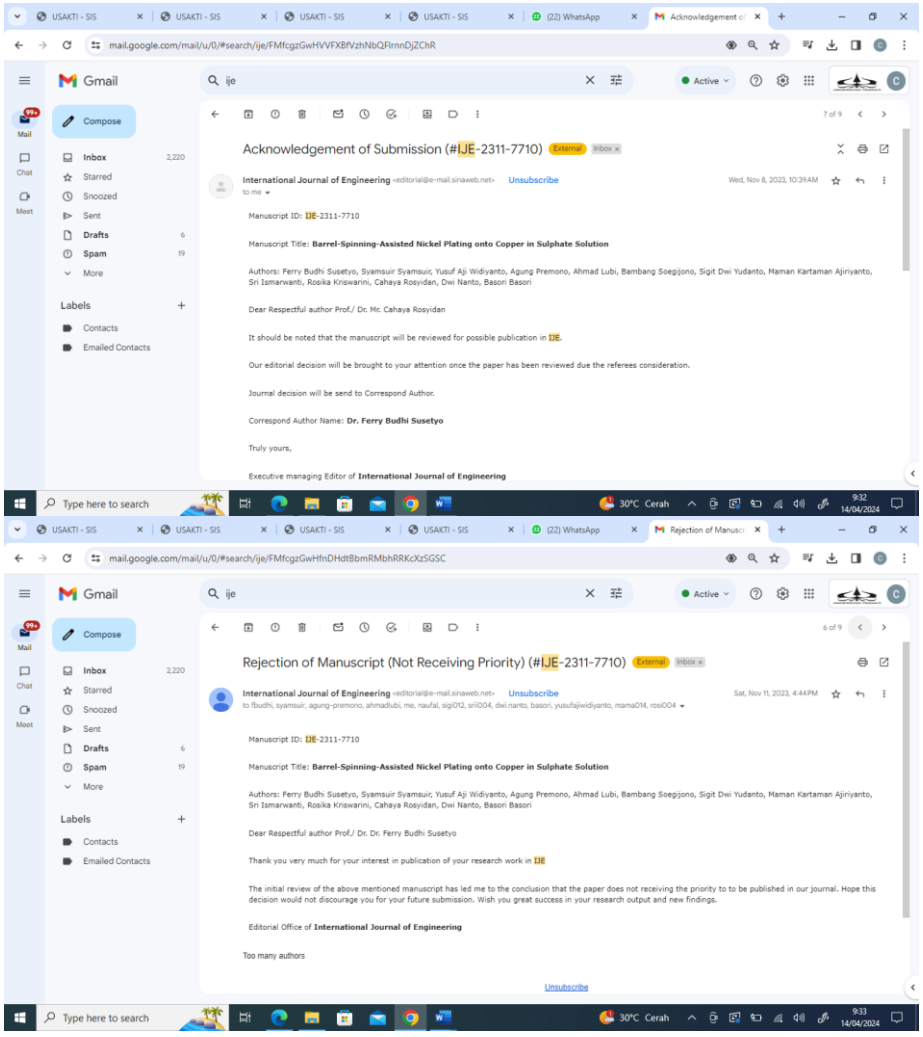

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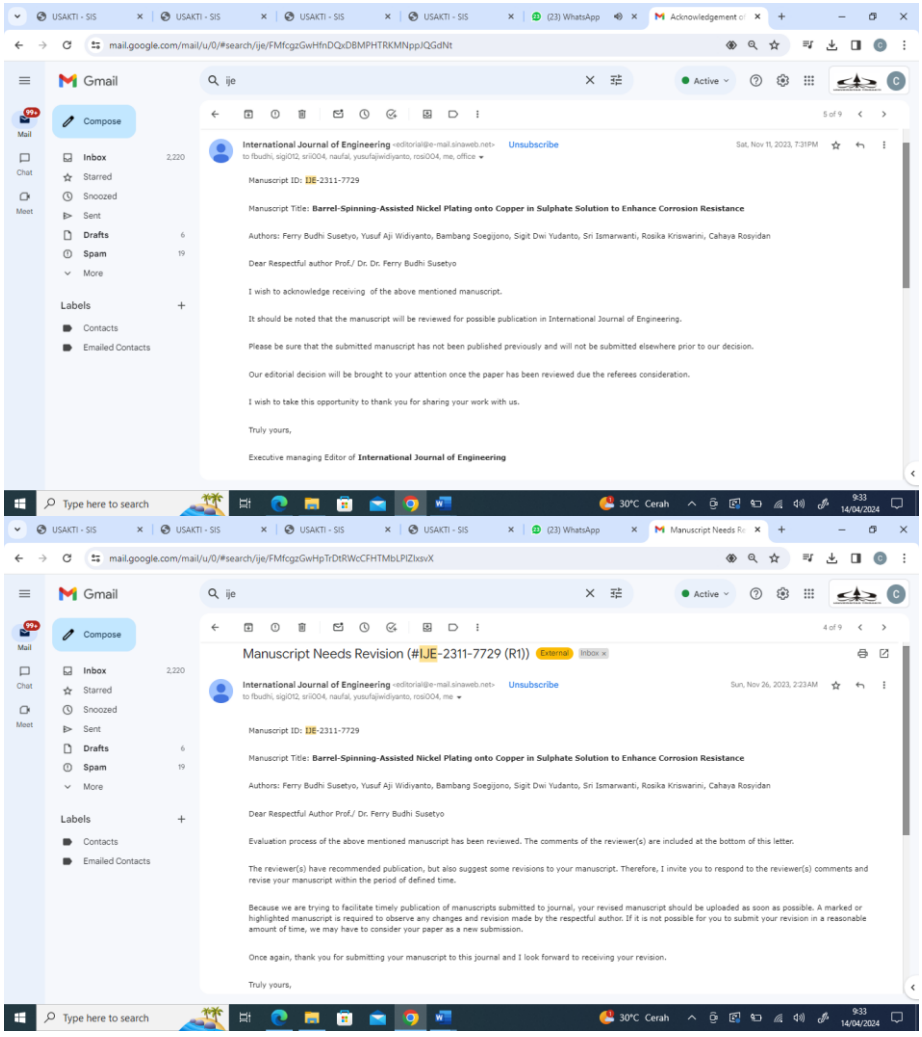

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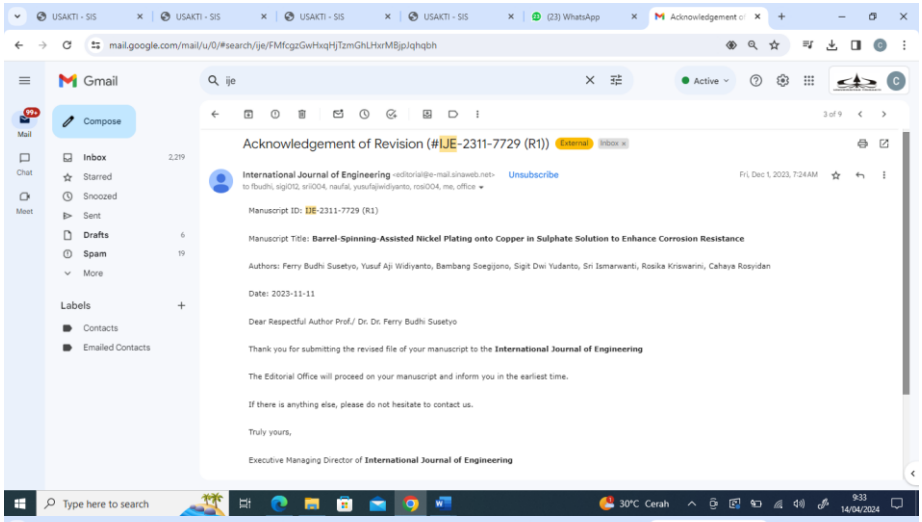
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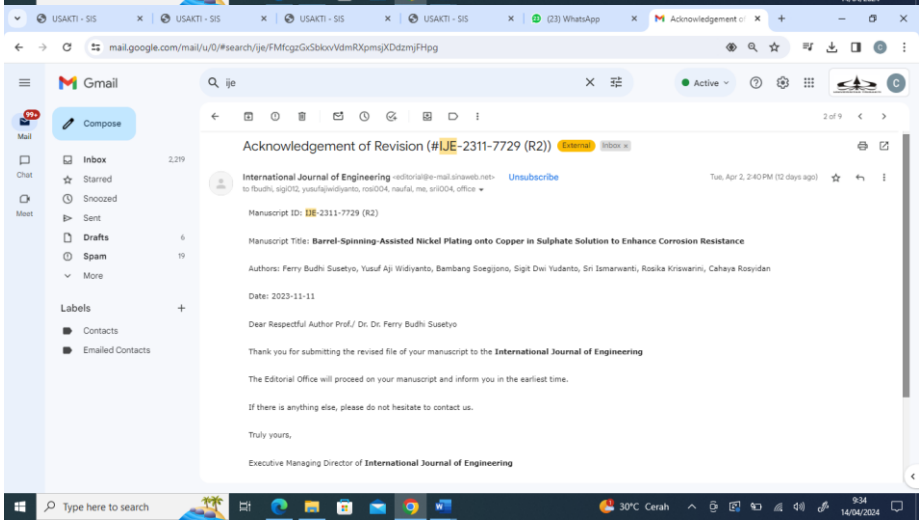
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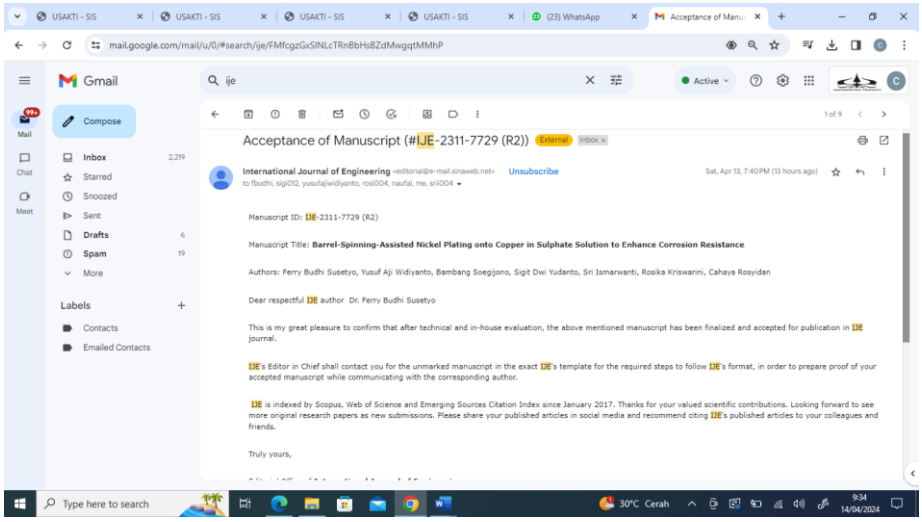
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We look forward to the outcome of your assessment.

We hope the revised version is suitable for publication and look forward to hearing from you in due course.

Yours Sincerely,

Cahaya Rosyidan

Department of Physics

Universitas Indonesia

Indonesia

E-mail: budhy.kurniawan@sci.ui.ac.id

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Effect of Current Density on Magnetic and Hardness Properties of Ni-Cu Alloy Coated on Al via Electrodeposition

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ABSTRACT

Nickel (Ni)-rich single-phase nickel-copper (Ni-Cu) alloy coatings were produced on aluminum (Al) substrates by electrodeposition in stabilized citrate baths. Electrodeposition experiments were performed at four different current densities. Increasing the current density resulted in the metal deposition rate increasing faster than the hydrogen evolution rate; thus, the cathodic current efficiency increased. The crystal systems of the Ni-Cu alloys were face center cubic (fcc), with the (111) plane as the preferred crystal plane. Scanning electron microscopy with energy dispersive X-ray spectroscopy (SEM-EDS) measurements showed that the Ni content in the coating increased with increasing current density. The Ni-Cu 40 sample had the most Ni content and showed a homogeneous and compact morphology. It was found that the higher the concentration of Ni in the solution, the smaller the grain size. Measurements recorded with a vibrating sample magnetometer (VSM) showed that the Ni-Cu 40 sample provided magnetic saturation, with the highest value being 0.108 emu/g. The microhardness method produced 404 HV on the Ni-Cu 40 sample. In conclusion, higher current densities were associated with a higher Ni composition and increased thickness, which were responsible for the increases in the magnetic properties and hardness.
doi: 10.5829/ije.2023.36.01a.00

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NOMENCLATURE			
Ce	Cathodic current efficiency	Wi	Initial weight of the substrate
σ	Lattice strain	Wf	Weight
Ws	Final weight of the substrate	I	Total current
Wm	Ratio of the final weight of the substrate	t	Deposition time
μ	Texture coefficient of the unique plane	F	Faraday's constant
I(hkl)	Measured intensity	fni	Nickel deposit weight ratio
mcu	Copper's atomic weight	mni	Nickel's atomic weight

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1. INTRODUCTION1

Researchers widely study nickel (Ni) and copper (Cu) alloys as engineering materials due to their unique mechanical, magnetic, and anti-corrosion properties [1, 2]. Ni-Cu alloys are known as monel in the industry and are typically comprised of 70 wt% Ni and 30 wt% Cu [3]. These alloys have outstanding capabilities in acidic and alkaline environments [4]. Ni-Cu alloys are single-phase alloys throughout their composition on the phase diagram, and these alloys formed because Ni and Cu are fully soluble in their solid and liquid states [5]. Ni and Cu both have a face center cubic (fcc) crystal structure, and they have almost similar electronegativity and atomic radii [6, 7].

Given that conventional casting as a manufacturing method for monel results in substantial production costs and that Ni-Cu alloy coated on aluminum (Al) has potential as a replacement for monel as a bulk material [8], Al-based metals have received considerable attention [9, 10]. They are lightweight and demonstrate high resistance to wear and corrosion and a high strength–stiffness combination [11].

Several techniques have been proposed to successfully modify the surface morphology and chemical composition, including sol-gel, chemical etching, chemical vapor deposition, thermal embossing, and electrodeposition [12, 13]. The electrodeposition technique is a cost-effective, scalable, and easy-to-control process for coating Ni-Cu alloy [14]. Specific methods have also been developed to determine the structure, morphology, and phase composition of the coated Ni-Cu alloys [15, 16]. Goranova et al. investigated how changing the concentration of Ni ions and the current density affected the structure and composition of Ni-Cu alloys formed by electrodeposition in alkaline citrate baths [17]. Higher concentrations of Ni ions in the bath led to notably smoother deposits and enhanced current efficiency. However, producing a uniform Ni-Cu coating can be challenging due to the difference in reduction potential between Ni and Cu. The reduction potential of Ni atoms is -0.25 V vs. SHE, and that of Cu atoms is +0.34 V vs. SHE [18]. As a result, controlling the concentrations of Ni and Cu is vital. Complexing agents must be added to narrow the potential difference between Ni and Cu. The most frequently used complexing agent is citrate due to its low toxicity, cost-effectiveness, and buffering characteristics [19].

The electrodeposition process affects the physical properties of the resultant Ni-Cu alloy, as does the current density. A high current density causes the crystal plane to be oriented in the (111) plane, the lattice size to be smaller, and the atomic distance to be less [20]. The grain size becomes smaller when the current density is high, and the morphological shape resembles that of a cauliflower [21]. As a result of a high current density, the coating will be thicker, and the composition of the Ni weight fraction will also be higher. The amount of Ni deposited on the substrate and the thickness of the coating both have an impact on the product's magnetic properties [22]. In addition, a smaller grain size results in an increase in hardness [23]. Kanukaran et al. reported a hardness of 153 HV when the current density was 40 mA/cm2 [24], and Karunakaran and Pugazh Vadivu reported a magnetic saturation value of 0.0004 emu/g at 40 mA/cm2 [25]. Nevertheless, the researchers did not examine the impact of the coating electrodeposition factors, structure, and morphology on the magnetic and hardness properties.

The aims of this research were 1) to produce a Ni-rich Ni-Cu alloy coating on Al via electrodeposition and 2) to investigate the link between magnetic and hardness properties and the coating's microstructure and surface morphology. We varied the current density, and the process was conducted at room temperature. We examined the influence of various process variables on the cathodic current efficiency, structure, morphology, composition, grain size, and thickness of the produced coatings. Finally, the magnetic properties and hardness of the coatings were investigated.

2. MATERIAL AND METHODS

2. 1. Material and Electrodeposition Process The chemical composition of the Al substrate (cathode) used was Fe = 1.63 wt%, Mg = 1.49 wt%, and Al = 96.88 wt%. The chemical composition of the Ni (anode) used was Al = 0.02 wt%, Ca = 0.04 wt%, Fe = 0.23 wt%, Y = 1.61 wt%, Zr = 0.04 wt%, Nb = 0.05 wt%, and Ni = 98.01 wt%. The chemical composition of the Cu (anode) used was P = 0.22 wt%, Cd = 0.684 wt%, Si = 0.137 wt%, and Cu = 98.959 wt%. The Al was cleaned from the oxide coating with sandpaper before deposition using DELTA D68H for 5 min. Ni-Cu electrodeposition was carried out using a SANFIX 305 E DC power supply. The samples produced using

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a current density of 15 mA/cm2, 20 mA/cm2, 30 mA/cm2, and 40 mA/cm2 were designated as Ni-Cu 15, Ni-Cu 20, Ni-Cu 30, and Ni-Cu 40, respectively. Table 1 shows the bath composition and deposition parameters.

TABLE 1. Bath composition and deposition parameters

Bath composition and condition	Quantity
NiSO4.6H2O (Merck)	0.5 M
CuSO4.5H2O (Merck)	0.04 M
Na3C6H5O7 (Merck)	0.2 M
pH	4.2
Temperature	25 °C
Deposition time	1 h

2. 2. Characterization The deposition rate was calculated using the previously reported method [26]. The following formula, Equation 1, was used to calculate the efficiency of the cathodic current [27]:

$$C_e = {W_m}/{W_f} \quad (1)$$

Wm and Wf were calculated using Faraday's law, as shown in Equations (2) and (3).

$$W_m = W_s - W_i, \quad (2)$$

$$W_f = \{(m_{ni}/2) * f_{ni} + (m_{cu}/2) * f_{cu}\} * I.\frac{t}{F}. \quad (3)$$

The crystal structure of the Ni-Cu coating was determined using X-ray diffraction (XRD-PANanalytical Aeris Instrument Suit) (Cu-Kα radiation, λ = 0.15418 nm). XRD data were collected from 20° to 80° with a step size of 0.020°. The Materials Analysis Using Diffraction (MAUD) program was used to determine the crystal parameters of the sample after Rietveld refinement.
The preferential crystallite orientation was determined from the texture coefficient μ, as shown in Equation 4 [28]:

$$\mu = \frac{I(hkl)/I_0(hkl)}{(\frac{1}{N})\sum[I(hkl)/I_0(hkl)]}, \quad (4)$$

Based on the XRD results, the lattice strain σ was calculated using Equation (5) [29]:

$$\sigma = \left(\frac{\beta}{4 \times \tan\theta}\right) \quad (5)$$

SEM-EDS (Thermofisher Quanta 650 EDAX EDS Analyzer) with 1000× magnification was used to analyze the surface morphology of the Ni-Cu coatings. EDS was used to determine the chemical composition of the coatings, and the statistical distribution of grain sizes was calculated using ImageJ software. The cross sections of the coated samples were also examined to assess how the current density and CCE affected the coating thickness. Measurement of magnetic properties was conducted using a vibrating sample magnetometer (VSM, Oxford 1.2H). The hardness of the Ni-Cu coatings was measured using a MicroMct® 5100 Series Microindentation Hardness Tester. The ATM E384 standard was used for the tests, which were performed with a load of 100 g for 10 s at five places.

3. RESULTS AND DISCUSSION

3. 1. Cathodic Current Efficiency and Deposition rate

Figure 1 depicts the relationship between the average CCE and deposition rate in the citrate electrolyte bath. The CCE was found to be high, with a value of 82–89%. The basic concept of current efficiency can be understood as the fraction of total current used for metal plating [30]. Apart from metal deposition, hydrogen evolution is the only other necessary process that must occur on the substrate surface. Ni and Cu precipitation are both antagonistic to the hydrogen evolution reaction. In this study, the metal deposition rate increased faster than the hydrogen evolution rate when the current density increased from 15 to 40 mA/cm2; hence, the CCE increased. The highest CCE (89.96%) was associated with the Ni-Cu 40 sample, and the lowest CCE (82.55%) was associated with the Ni-Cu 15 sample. Basori et. al [26] and Syamsuir et. al [31] found that the deposition rate and current efficiency are considered linear.

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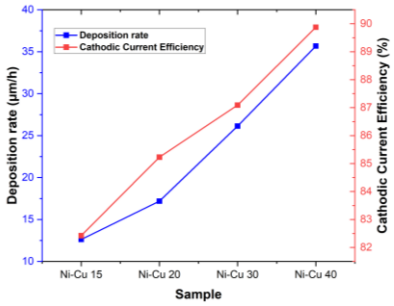
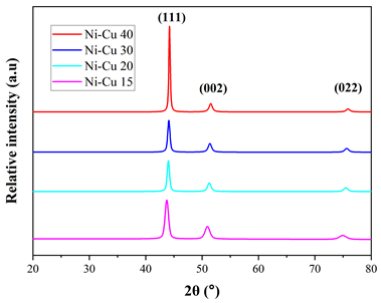


Figure 1. The cathodic current efficiency and deposition rate at a current density of 15, 20, 30, and 40 mA/cm2

3. 2. Structural Properties Figure 2(a) depicts the XRD spectra of the Ni-Cu alloy samples produced at various current densities. According to the XRD analysis results, each Ni-Cu alloy sample consisted of a single phase with an fcc structure. The evolution of the lattice parameters of the Ni-rich (111) phase over the range of current densities is shown in Figure 2(b). The peaks of each sample were found between the peaks of the fcc of 2θ = 43.3o for pure Cu and 2θ = 44.5 o for pure Ni [32, 33]. As the Ni content of the alloy coating increased, the diffraction angle also increased.



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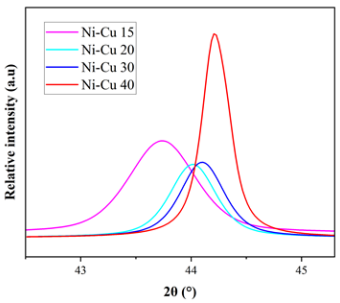


(b)

Figure 2. (a) X-ray diffraction spectra of Ni-Cu coatings electrodeposited at various current densities and (b) the extended view of the Ni-Cu (111) plane, showing peak shifts

Crystal size calculation using MAUD resolved refinement was used to determine the size of the crystallites in the Ni-Cu alloys, and the results (Table 2) show that the crystallite size of the Ni-Cu coating ranged from approximately 24 to 50 nm. The crystallite size of a pure Ni layer is 60 nm, meaning that the Ni-Cu alloys had smaller crystallite sizes than a pure Ni layer. This result is also similar to that obtained by Li et al. [34]. In contrast to the typical watt-Ni coating, we found that the presence of a sodium citrate complexing agent resulted in a finer crystallite size. This is consistent with the findings of Sarac and Baykul [35], who observed that Cu atoms affect grain refinement in Ni-Cu alloys. Cu atoms can restrain the surface diffusion of Ni atoms during the deposition process and inhibit the growth of crystallites.

The evolution of the crystallographic orientation of the Ni-Cu coatings produced with varying current densities is shown in detail in Figure 3. It can be observed that a strong (111) fiber texture appeared in all the samples, while the (002) texture gradually decreased as the current density increased.

The μ values of different crystal planes are also used to evaluate the degree of crystallographic orientation [34]. Moreover, the Ni-Cu coating electrodeposited at the current density of 40 mA/cm2 was found to have a strong (111) texture.

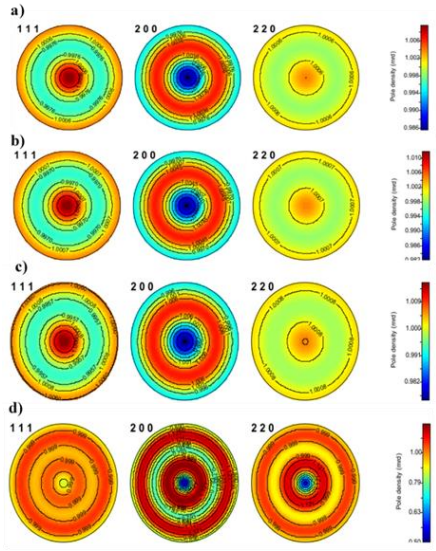


Figure 3. The simulated 2D pole figures for the (a) Ni-Cu 15, (b) Ni-Cu 20, (c) Ni-Cu 30, and (d) Ni-Cu 40 samples

TABLE 2. Parameters of the Ni-Cu alloys after Rietveld refinement using MAUD

Sample				
Parameter	Ni-Cu 15	Ni-Cu 20	Ni-Cu 30	Ni-Cu 40
Crystal structure	Cubic fcc			
Space group	Fm-3m			
Lattice constant (Å) a = b = c	3.582	3.560	3.554	3.545
Volume (Å3)	45.975	45.152	44.905	44.557
d-spacing (Å)	1.791	1.780	1.695	1.691
Crystallite size (nm)	24.55	26.82	32.29	50.78
Rwp (100%)	3.640	4.484	5.394	5.139
GOF	1.94	1.72	2.06	2.03
Lattice strain	0.663	0.453	0.431	0.276

The texture coefficient for every preference was calculated using Equation 4 to ascertain the preferred crystal orientation direction of each Ni-Cu alloy obtained at the various current densities, and the results are shown in Table 3 [32].
It seems that the texture coefficient was also dependent on the peak current, and the preferred orientation was the (111) plane. Li et al. [34] found that the higher the current density, the more dominant the (111) plane. The findings suggest that the (111) crystallographic orientation was preferable for all the Ni-Cu coatings electrodeposited at the tested current densities.

TABLE 3. Texture coefficient analysis of Ni-Cu deposits

Sample	μ (hkl)		
	[111]	[002]	[022]
Ni-Cu 15	1.12	0.86	0.77
Ni-Cu 20	1.17	0.74	0.81
Ni-Cu 30	1.18	0.74	0.78
Ni-Cu 40	1.48	0.34	0.32

The lattice strain of the prepared coatings was determined using Equation 5, and Figure 4 illustrates the changes in the crystal size and lattice strain of the Ni-Cu coatings based on the current density applied in the plating bath. The crystal size increased and the lattice strain decreased as the current density increased [36]. This result aligns with that obtained by Devi et al. [33], who showed that the higher the current density, the more the crystal size increased. A possible

reason for this is that the composition of Ni increases as the current density increases.

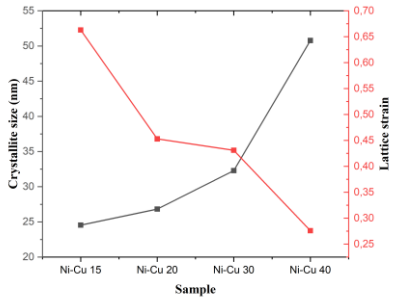


Figure 4. The lattice strain and crystallize size of the Ni-Cu alloys coated on Al at various current densities

3. 3. Surface Morphology EDS was used to determine the elemental composition of the Ni-Cu coatings, and the results are shown in Figure 5 and Table 4. Cu and Ni were the only elements present in the deposits. The alloy composition was influenced by the current density: as the current density increased, the Cu content decreased. This phenomenon can be caused by the [Ni2+]/[Cu2+] ratio in the bath, which changes the composition of Ni and Cu. Goranova et al. discovered that as the Cu content of deposits decreased, so did the CCE [37]. This phenomenon occurs because of the orderly deposition of Ni and Cu [38]. In regular deposition, increasing the current density leads to an increased proportion of less noble metals in the deposited material [39]. In our scenario, Ni is the less noble metal. As a result, when the current density was higher, the deposits were richer in Ni.

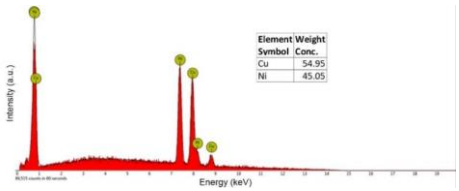


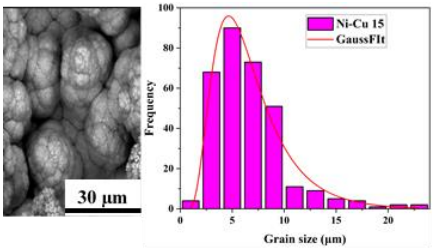
Figure 5. Eds graph of the Ni-Cu 15

Another notable feature was that as the current density increased, the peak shifted to the right (i.e., to higher 2θ values). The change in the alloy composition may have also caused this peak shift. The Ni concentration increased as the current density increased (see Table 4). Because Ni and Cu combine to produce a single-phase alloy, the diffraction peak shifted toward that of pure Ni as the Ni percentage increased. This finding is similar to the observations of Goranova et al. [17], who found that the fcc reflection for Ni-rich Ni-Cu alloy deposits shifted to the right as the Ni concentration increased. Indeed, it is logical to expect the Ni-Cu alloy peak to shift as the Ni content increase.

TABLE 4. The chemical composition of Ni-Cu coatings on the Al substrate prepared at different current densities

Sample	Cu, wt%	Ni, wt%
Ni-Cu 15	54.95	45.05
Ni-Cu 20	39.83	60.17
Ni-Cu 30	29.80	70.20
Ni-Cu 40	19.64	80.36

The surface morphological structure and the cross section of the coated samples were observed using SEM. Figure 6 shows SEM micrographs of the four samples' surface morphological structures. The deposits developed a fine-grained and compact spherical shape when lower deposition current densities were applied (Figure 5(a)). Deo et al. [27] and Goranova et al. [37] also observed this morphology at low current densities. The shape changed to a coarser cauliflower form when higher current densities were applied (Figure 6(d)). A diffusion-limited deposition mechanism in which a multigeneration spherical diffusion layer creates a cauliflower shape is likely to produce this type of morphology [40]. As the current density increased, the cauliflower-like protrusions became more spaced and separated, creating gaps. The Ni-Cu 40 sample, produced with the highest current density, was found to have the largest gaps between the cauliflower-like bulges.



(a)

Commented [WU7]: Why is there a significant difference in the results for Ni-Cu 40 sample?

Commented [H8R7]: According to Figure 2(b), the FWHM for peak (111) decrease with increasing current density. This result is consistent with earlier studies in which increasing current density reduces FWHM. DOI: 10.24203/ajas.v8i4.6287

Commented [WU9]: Is eds accurate enough to determine chemical composition?
The change in composition is very impressive.

Commented [H10R9]: Yes accurate. The chemical composition of small objects is determined using the EDS method, which involves placing the material in a vacuum chamber and lighting it from above with a focused electron beam. Ionised atoms emit discrete energy X-ray quanta characteristic of the chemical contents of the material under analysis.

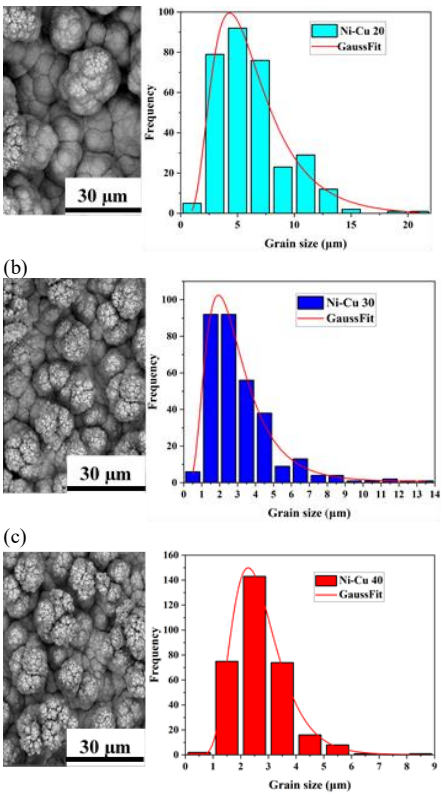


Figure 6. Surface SEM images of the deposited Ni-Cu alloy coatings and plots showing the statistical distribution of the grain size

The increased nucleation rate can explain the observed decrease in grain size with increasing current density [41][42]. Ni-Cu ion flow to the cathode is faster at higher current densities. Further investigation is needed to determine the exact relationship between the current density and the grain size of the coating. In the Ni-Cu 15, Ni-Cu 20, Ni-Cu 30, and Ni-Cu 40 samples, the peak that corresponded to the (111) plane shifted toward the right (see Figure 2b). The reduction in d-spacing is ascribed to residual stress induced at a higher deposition rate [43]. The statistical results of the grain size distribution presented in Figure 6 indicate that the grain size ranged from 4.63 to 1.94 µm. The decrease in grain size with the increase in current density is evident in the data shown in Table 5.

TABLE 5. Average grain size found in each sample

Sample	Average grain size (µm)
Ni-Cu 15	4.63 ± 0.269
Ni-Cu 20	4.38 ± 0.365
Ni-Cu 30	2.28 ± 0.068
Ni-Cu 40	1.94 ± 0.032

Figure 7 depicts the relationship between the deposited alloy composition and the applied current density. EDS was used to determine the composition.

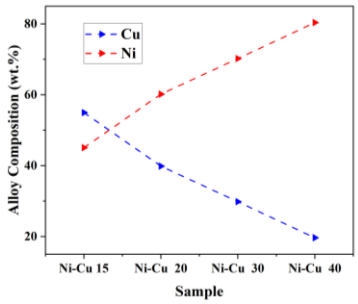


Figure 7. The dependence of the deposited alloy's composition (shown as wt% of the single electrolytes) on current density

Figure 8 (a–d) displays SEM cross-section images of the produced Ni-Cu coatings. The absence of cracks between the substrate and coating demonstrates that appropriate adhesion occurred between the two entities [44]. The thickness of the electrodeposited Ni-Cu coating was also measured for each sample [27], and the following results were recorded: Ni-Cu 15 = 32 µm, Ni-Cu 20 = 42 µm, Ni-Cu 30 = 49 µm, and Ni-Cu 40 = 50 µm. The effect of the current density on the thickness of the Ni-Cu coating is depicted in Figure 8; the thickness increased as the current density increased [19]. Hence, a higher current density results in more mass and a thicker coating. The findings presented in Figure 1 show that as the current density increased, so too did the CCE.

According to Faraday's law, when the deposition time remains constant for all samples, coatings formed at lower current densities will be thinner than those produced at higher current densities. A thinner covering may lead to severe interference from the Al substrate. The cathodic current density also affects the coating, as Deo et al. [27] discovered that increasing the current density improves the thickness of the film due to an increase in the CCE.

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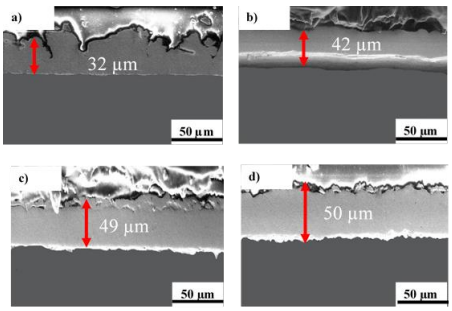
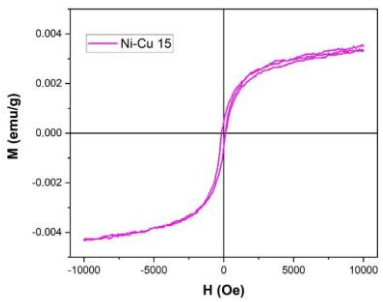
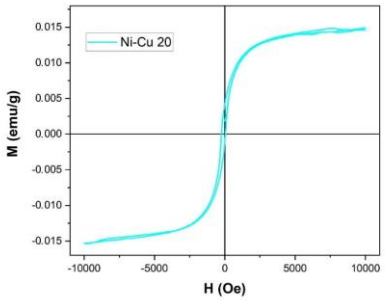


Figure 8. SEM cross-section images of the Ni-Cu coating on Al in the (a) Ni-Cu 15, (b) Ni-Cu 20, (c) Ni-Cu 30, and (d) Ni-Cu 40 samples

3. 4. Magnetic Properties Figure 9 depicts the magnetic characteristics and fluctuations in the current density magnetization measured with a VSM at room temperature [45]. The results of the VSM analysis demonstrate that the coatings in the Ni-Cu 15, Ni-Cu 20, Ni-Cu 30, and Ni-Cu 40 samples displayed ferromagnetic activity. The low ferromagnetic activity of the Cu54.95Ni45.05 alloy film of Ni-Cu 15 could be attributed to Ni diffusion in the Cu matrix, as Cu is a diamagnetic metal and Ni is a ferromagnetic metal [46]. The ferromagnetic properties of the Ni-Cu films of Ni-Cu 20, Ni-Cu 30, and Ni-Cu 40 increased with the Ni content of the alloy coatings.



(a)



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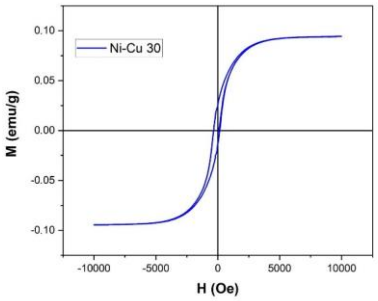

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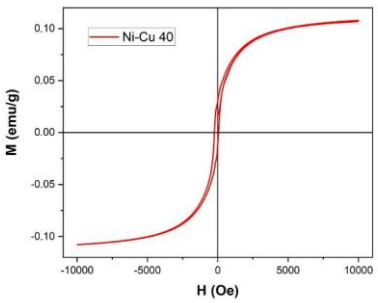

(d)

Figure 9. Hysteresis loops of multilayers generated at different current densities

As the Ni content of the Ni-Cu alloy coatings increased, so did the saturation magnetization (see Table 6). Wang et al. reported that saturation magnetization depends on the Ni content of Ni-Cu

alloy coatings [43]. In addition, Awasthi reported that magnetization is enhanced by increasing the coating thickness [22]. A possible reason for this enhanced magnetization is the magnetic disorder caused by the coating. It has been shown that the trend in saturation magnetization enhancement is associated with the coating level [47]. Demidenko et al. [48] found that monel has paramagnetic properties at room temperature, while the Ni-Cu alloys in this study had ferromagnetic properties. Here, we have improved upon the results of previous studies in which phosphorus (P) and tungsten (W) were added [25]. In this study, the Ni-Cu 40 sample exhibited the strongest magnetic properties.

TABLE 6. The results of the magnetic analysis of the Ni-Cu/Al samples

Sample	Hc (Oe)	Mr (emu/g)	Ms (emu/g)
Ni-Cu 15	143.829	0.00047	0.003
Ni-Cu 20	140.081	0.004	0.015
Ni-Cu 30	256.215	0.025	0.094
Ni-Cu 40	144.023	0.032	0.108

3. 5. Hardness Figure 10 depicts the dependence of the microhardness of the Ni-Cu coatings on the current density in the plating bath. From the data presented in Figure 10, it is clear that the coating of the Ni-Cu 40 sample had the highest microhardness value (404 HV). In general, the microhardness increased with the current density and was attributed to the grain size and thickness of the coatings [42]. Pingale et al. found that hardness increased with the thickness of the coating [19]. The coating thickness could influence the hardness of Ni-Cu films [26].

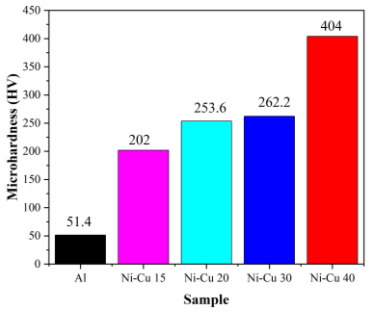

Figure 10. The microhardness of the electrodeposited Ni-Cu coatings of the samples produced with different current densities

The results indicate that alloys with a greater Ni content are mechanically harder. The overall dependence of hardness and microhardness on the percentage of Ni is shown in Figure 11, and the data indicate that microhardness increases as the percentage of Ni increase. This result is similar to Marenych's finding that the hardness value is highest with the highest Ni composition [49].

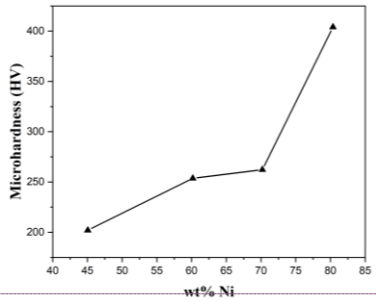

Figure 11. The relationship found between wt% Ni and microhardness, based on the data obtained from the four experimental samples

Moreover, the hardness reported in previous studies that resulted from electrodeposition of Ni-Cu on Al in the presence of P was lower than that recorded in the present study [24]. This is due to the smaller grain size that resulted from applying a different current density. In addition, the increase in microhardness reported here is related to the role that Ni atoms play in grain refinement [31]. Ramkumar et al. [50] reported a monel hardness value of 165 HV, which is lower than the peak hardness value recorded in the current study.

4. CONCLUSION

In this study, Ni-Cu alloys were electrodeposited onto Al substrates using citrate baths. The effects of adding a citrate solution on the properties of the deposited coatings were studied, and the results illustrate that single-phase Ni-Cu alloy layers were produced on the Al surface at all current densities. The CCE increased as the current density increased. The coatings formed at lower current densities showed a more compact and spherical morphology, while those formed at higher current densities showed a less uniform structure with a cauliflower-like morphology. Both the surface morphology and composition of the coating showed a strong dependence on the current density. The Ni-Cu alloy coating deposited at a low current density had a layer thickness of 32 μm, while the coating deposited

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at a high current density had a thickness of 50 μm. The saturation magnetization of the coating increased with the Ni content in the Ni-Cu alloy and with the coating thickness. The hardness increased with the coating thickness, grain size, and Ni composition in the solution. The hardness of the produced Ni-Cu alloy coatings was found to be greater than that of monel.

5. ACKNOWLEDGMENTS

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Effect of Current Density on Magnetic and Hardness Properties of Ni-Cu Alloy Coated on Al via Electrodeposition

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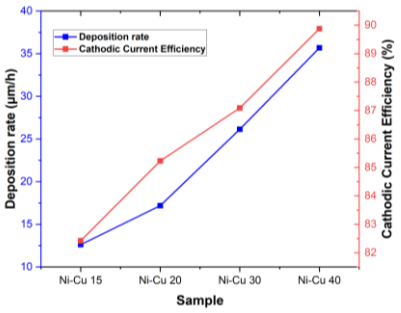
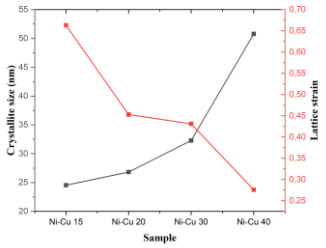
Please cite IJE's recent published articles related to your work. DOI for all refernces required.

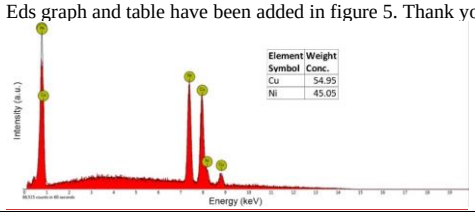
Response:

Seven IJE-published papers were related, and we have cited them with pleasure (ref 3, 9, 10, 11, 12, 13, 31, 36, 42, and 44). We would recommend our colleagues for IJE's recently published paper for future publication in any journal. We hope IJE's ranking always increases shortly. DOI, all listed references have been added. Thank you.

#Reviewer 1

<table>
<tr><th>No.</th><th>Comment from Reviewer</th><th>Revision</th></tr>
<tr><td>1</td><td>Add Ni-Cu coating</td><td>Ni-Cu coating was added. Thank you</td></tr>
<tr><td>2</td><td>Why are these two materials mentioned?</td><td>I apologize there is a little mistake in the writing, it should be Ni and Cu alloy</td></tr>
<tr><td>3</td><td>Why is not crystallite size measured by Rietveld, which has higher accuracy?</td><td>Thanks for your input, the crystal size has been recalculated with the Rietveld refinement method. In Rietveld refinement, we can model the entire crystal structure and parameters in one analysis. This analysis includes parameters such as crystal structure, lattice parameter, gof, rwp, atomic position, diffraction pattern, including intensity and position of diffraction peaks, which enables a more comprehensive analysis. So that it can provide a complete picture of the influence of inter-parameters. Therefore, it can improve the accuracy of crystallite size measurement.
<table>
<tr><th rowspan="2">Parameter</th><th colspan="4">Sample</th></tr>
<tr><th>Ni-Cu 15</th><th>Ni-Cu 20</th><th>Ni-Cu 30</th><th>Ni-Cu 40</th></tr>
<tr><td>Crystal structure</td><td colspan="4">Cubic fcc</td></tr>
<tr><td>Space group</td><td colspan="4">Fm-3m</td></tr>
<tr><td>Lattice constant (Å) a = b = c</td><td>3.582</td><td>3.560</td><td>3.554</td><td>3.545</td></tr>
<tr><td>Volume (Å^3)</td><td>45.975</td><td>45.152</td><td>44.905</td><td>44.557</td></tr>
<tr><td>d-spacing (Å)</td><td>1.791</td><td>1.780</td><td>1.695</td><td>1.691</td></tr>
</table></td></tr>
</table>

		Crystallite size (nm)	24.55	26.82	32.29	50.78
		Rwp (100%)	3.640	4.484	5.394	5.139
		GOF	1.94	1.72	2.06	2.03
		Lattice strain	0.663	0.453	0.431	0.276
4	is this consistent with similar research?	Yes, this is consistent with other studies, we add the deposition rate graph and the same study. 				
5	Why is there a significant difference in the results for Ni-Cu 40 sample?	According to Figure 2(b), the FWHM for peak (111) decrease with increasing current density. This result is consistent with earlier studies in which increasing current density reduces FWHM. DOI: 10.24203/ajas.v8i4.6287 				
6	Is eds accurate enough to determine chemical composition? The change in composition is very impressive	Yes accurate. The chemical composition of small objects is determined using the EDS method, which involves placing the material in a vacuum chamber and lighting it from above with a focused electron beam. Ionised atoms emit discrete energy X-ray quanta characteristic of the chemical contents of the material under analysis. Besides using EDS, there are also using XRF and AAS. The advantages of the XRF method are that the samples analyzed do not need to be tampered with, have high accuracy, can determine the elements in the material in the absence of standards, and can determine the mineral content in biological materials and the body directly. The disadvantage of the XRF method is that it cannot analyze elements below atomic number 10. Samples are solutions, and mostly water. Thus those of interest to study such as soils, animal tissues, plants, and minerals require complex preparation to form test-ready analytes. The decomposition of these materials usually requires harsh treatment at high temperatures, resulting in the potential loss of analytes by evaporation or as particulates in the smoke. Can only measure metals in total, cannot distinguish between species such as oxidation state, free metal or bound to other molecules such as metalloproteins, organometallics, etc.				

7	EdS graph and table is added for at least one sample	Eds graph and table have been added in figure 5. Thank you 
8	Hc of Ni-Cu 3, why is it high?	The largest Hc means that this sample is difficult to demagnetized. It is due to magnetic anisotropy. It is predicted, since Ni-Cu 30 has the largest Hc, it revealed that the magnetic anisotropy in the Ni-Cu 30 is also highest.

#Reviewer 2

No.	Comment from Reviewer	Revision
1	Please update references and try to use some references from the following journals: International journal of engineering Advanced ceramic progress or ACERP published by MERC	Yes,A reference journal has been added. Thank you. We have cited them with pleasure (ref 3, 9, 10, 11, 12, 13, 31, 36, 42, and 44)