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Effect of Current Density on Magnetic and Hardness Properties of Ni-Cu Alloy Over Al Substrate by Electrodeposition Method

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ABSTRACT

Ni-rich single-phase Ni-Cu alloy coatings were obtained on Al substrates by electrodeposition from stabilized citrate baths. Electrodeposition experiments were carried out at four different current densities. Increasing the current density can increase the cathodic current efficiency, so the change in metal deposition rate has increased faster than the rate of hydrogen evolution, thus increasing the cathodic current efficiency. In this study, all the resulting crystal systems of Ni-Cu alloys are fcc, according to x-ray diffraction (XRD) data, 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 obtained the most Ni content and showed a homogeneous and compact morphology. This study found that the higher the concentration of Ni in solution, the smaller the grain size. The measurements of the vibrating sample magnetometer (VSM) found that the Ni-Cu 40 sample provided magnetic saturation with the highest value at 0.108 emu/g. The microhardness method produces 404 HV on Ni-Cu 40. In conclusion, higher current densities result in higher Ni composition and increased thickness, which are responsible for the higher magnetic properties and hardness.

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NOMENCLATURE			
k	Constant of 0.9	W_i	Initial weight of the substrate
β	Full width at half maximum (FWHM)	W_f	Weight
λ	X-ray wavelength (Cu Kα is 0.154 nm)	I	Total current
θ	Bragg's angle	t	Deposition time
μ	Texture coefficient of the unique plane	F	Faraday's constant
$I(hkl)$	Measured intensity	f_{ni}	Nickel deposit weight ratio
C_e	Cathodic current efficiency	m_{ni}	Nickel's atomic weight
W_m	Ratio of the final weight of the substrate	m_{cu}	Copper's atomic weight
W_s	Final weight of the substrate	σ	Lattice strain

1. INTRODUCTION

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, with a composition of 70 wt% Ni and 30 wt%Cu. This alloy has outstanding capabilities in acidic and alkaline

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environments [3]. Ni-Cu alloys are single-phase alloys throughout their composition on the phase diagram. This alloy is formed because Ni and Cu are fully soluble in solid and liquid states [4]. Ni-Cu alloys have an face center cubic (fcc) crystal structure and have almost similar electronegativity and atomic radii that are not much different [5, 6]. Using conventional casting as a manufacturing method for monel leads to substantial production costs. Ni-Cu alloys coating on aluminum (Al) is known to have many potentials to replace monel as a bulk material [7]. Since then, Al-based metals have received considerable attention due to their lightweight, high corrosion resistance, high strength-stiffness combination, and wear resistance [8].

Several techniques have been proposed to successfully modify surface morphology and chemical composition, including sol-gel, chemical etching, chemical vapor deposition, thermal embossing, and electrodeposition [9, 10]. The electrodeposition technique is a cost-effective, scalable, and easy-to-control process for coating Ni-Cu alloy [11]. Researchers can obtain the Ni-Cu alloy's structure, morphology, and phase composition by employing specific methods [12, 13]. Goranova et al. investigated how changing the concentration of Ni ions and current density affected the structure and composition of Ni-Cu alloys formed by electrodeposition in alkaline citrate baths [14]. Higher concentrations of Ni ions in the bath led to notably smoother deposits and enhanced current efficiency. However, making the Ni-Cu coating together takes work due to the difference in reduction potential between Ni atoms and Cu atoms. As known, Ni atoms has a reduction potential (-0.25 V vs. SHE) and Cu atoms (+0.34 V vs. SHE) [15]. As a result, controlling the concentration of Ni and Cu is very important. Complexing agents need to 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 [16].

The electrodeposition process with variations in current density, will affect to the crystal plane that is oriented in the (111) plane; besides that, it will also affect the smaller lattice size and the closer atomic distance [17]. The grain size becomes smaller due to the influence of the current density, and the morphological shape resembles to be cauliflower [18]. As a result of the influence of the current density, the thickness of 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 magnetic properties [19]. Hardness properties will be higher due to the smaller grain size [20]. M. Kanukaran found a hardness of 153 HV at a current density of 40 mA/cm^2 [21]. It is also reported a magnetic saturation value of 0.0004 emu/g at 40 mA/cm^2 [22]. Nevertheless, the researchers did not examine the impact of coating electrodeposition factors, structure, and morphology on the magnetic and hardness properties.

This research aims to obtain electrodeposition of a Ni-rich Ni-Cu alloy coating on Al and investigate the link between magnetic and hardness properties and the coating's microstructure and surface morphology. This study used some variations in current density and was conducted at room temperature. Coatings were studied, and the influence of various process variables on cathodic current efficiency, structure, morphology, composition, grain size, and thickness was investigated. Finally, the behavior of 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 (Fe = 1.63 wt.%; Mg = 1.49 wt.%; Al = 96.88 wt.%). The chemical composition of pure Ni (Al = 0.02 wt.%; Ca = 0.04 wt.%; Fe = 0.23 wt.%; Y = 1.61 wt.%; Zr = 0.04 wt.%; Nb = 0.05 wt.%; Ni = 98.01 wt.%). The chemical composition of pure Cu (P = 0.22 wt.%; Cd = 0.684 wt.%; Si = 0.137 wt.%; Cu = 98.959 wt.%). Al was cleaned of the oxide coating with sandpaper before deposition using DELTA D68H for 5 minutes. Ni-Cu electrodeposition was carried out using SANFIX 305 E DC power supply. The sample was designated as Ni-Cu 15, Ni-Cu 20, Ni-Cu 30, and Ni-Cu 40 for current density 15 mA/cm^2, 20 mA/cm^2, 30 mA/cm^2, and 40 mA/cm^2, respectively. Table 1 depicts the bath composition and parameters.

TABLE 1. Bath composition and deposition parameters.

Bath composition and condition	Quantity
$NiSO_4.6H_2O$ (Merck)	0.5 M
$CuSO_4.5H_2O$ (Merck)	0.04 M
$Na_3C_6H_5O_7$ (Merck)	0.2 M
pH	4.2
Temperature	25 °C
Time deposition	1 h

2. 2. Characterization The cathodic current efficiency (CCE) was calculated similarly by the methode as was previously reported [23]**.** The following formula is used to calculate the efficiency of the cathodic current. Equation 1 [23]:

$$C_e = {W_m}/{W_f} \tag{1}$$

W_m and W_f is calculated by Faraday’s law, as shown in Equations (2) and (3).

$$W_m = W_s - W_i, \tag{2}$$

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

The crystal structure of the Ni-Cu coating was determined by using XRD (Panalytical Aeris) (Cu-Kα radiation, λ = 0.15418 nm). XRD data were collected from 20° to 80° with a step size of 0.020°. MAUD (Materials Analysis Using Diffraction) was used to observe the crystal parameters of the sample after Rietveld refinement. The calculation of crystal size was measured using the Debye-Scherrer formula, Equation 4 [24]:

$$D = \frac{0.9\lambda}{\beta \cos\theta} \tag{4}$$

The preferential crystallite orientation determined from the texture coefficient as follows in Equation 5 [25]:

$$\mu = \frac{I(hkl)/I_0(hkl)}{(\frac{1}{N})\sum[I(hkl)/I_0(hkl)]}, \tag{5}$$

Where µ is the texture coefficient of the specific (*hkl*). I(hkl) and *I₀(hkl)* are the (*hkl*) peak intensities of the coatings and standard Ni-Cu powders, respectively.

The lattice strain is calculated using Equation (6) based on XRD results [26]:

$$\sigma = \left(\frac{\beta}{4 \times \tan\theta}\right) \tag{6}$$

Where β is the full width at half maximum (FWHM) and θ the Bragg's angle corresponding to fcc (111) peak.

SEM-EDS (Thermofisher Quanta 650 EDAX EDS Analyzer) with 1000 magnification was used to analyze the surface morphology of a Ni-Cu coating experiment on Al. EDS was used to determine the chemical composition of the coatings and the statistical distribution of grain sizes was calculated using ImageJ software. The coated samples' cross-sections were also examined to see how current density and cathodic current efficiency affect coating thickness. Measurement of magnetic properties was conducted using a vibrating sample magnetometer (VSM Oxford 1.2H). The hardness of the Ni-Cu coating 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 seconds at five places.

3. RESULT AND DISCUSSION

3. 1. Cathodic Current Efficiency Figure 1 depicts the relationship between average cathodic current efficiency and current density in a citrate electrolyte bath. A high cathodic current efficiency of 82–89% has been found. The basic concept of current efficiency can be understood as the fraction of total current used for current-efficient metal plating [27]. Aside from metal deposition, hydrogen evolution is the only necessary process on the substrate surface. Ni and Cu precipitation are both antagonistic to the hydrogen evolution reaction. The metal deposition rate has increased faster than the rate of hydrogen evolution from 15 to 40 mA/cm^2, cathodic current efficiency increases. The 40 mA/cm^2 sample had the maximum current efficiency, around 89%, while the 15 mA/cm^2 sample had the lowest, around 82%.

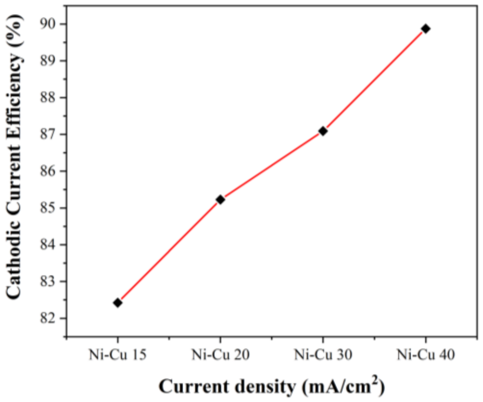
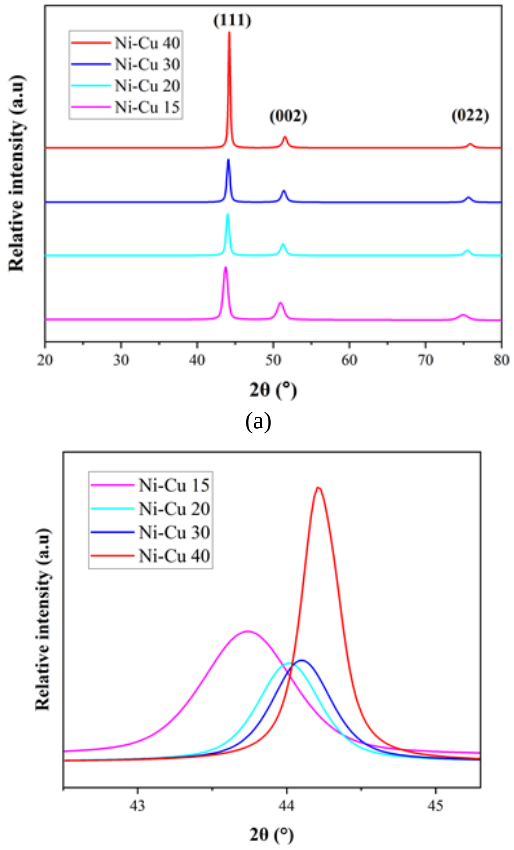


Figure 1. Effect of current efficiency at current density 15, 20, 30 and 40 mA/cm^2

3. 2. Structural properties Figure 2(a) depicts a Ni-Cu alloy's XRD at various current densities. According to XRD analysis, the Ni-Cu alloy was a single phase with an fcc structure. The evolution of the lattice parameters of the Ni-rich (111) phase with current density is shown in Figure 2(b). Each alloy's peaks were found between the peaks of the fcc of 2θ = 43.3 for pure Cu and 2θ = 44.5 for pure Ni [28, 29]. As the Ni atoms content of the alloy coating increased, the diffraction angle shifted to a higher angle.



(a)

(b)

Figure 2. (a) XRD spectrum of Ni-Cu coatings electrodeposited at various current densities and (b) extended view of Ni-Cu (111) plane showing shifting.

Based on Scherrer Equation 4, the size of crystallites in the Ni-Cu alloy was determined and reported in Table 2. It shows that the crystallite size of the Ni-Cu coating increases to 10-20 nm. Meanwhile, the pure Ni layer has a crystallite size of 60 nm, meaning that the Ni-Cu alloy has a smaller crystallite size than the pure Ni layer. This result is also similar to that obtained by Li et al. [30]. In contrast to the typical watt-Ni coating, the presence of a sodium citrate complexing agent results in a finer crystallite size. This is consistent with Sarac and Baykul [31], who observed Cu atoms affect to 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 detailed crystallographic orientation evolution of the Ni-Cu coating with varying current density is shown in Figure 3. It can be observed that a strong (111) fiber texture appears in all samples, while the (002) texture gradually decreases with increasing current density.

The μ (the relative texture coefficient) values of different crystal planes are also used to evaluate the degree of crystallographic orientation [30]. Moreover, strong (111) texture is present on Ni-Cu coating electrodeposited at the current density of 40 mA/cm^2.

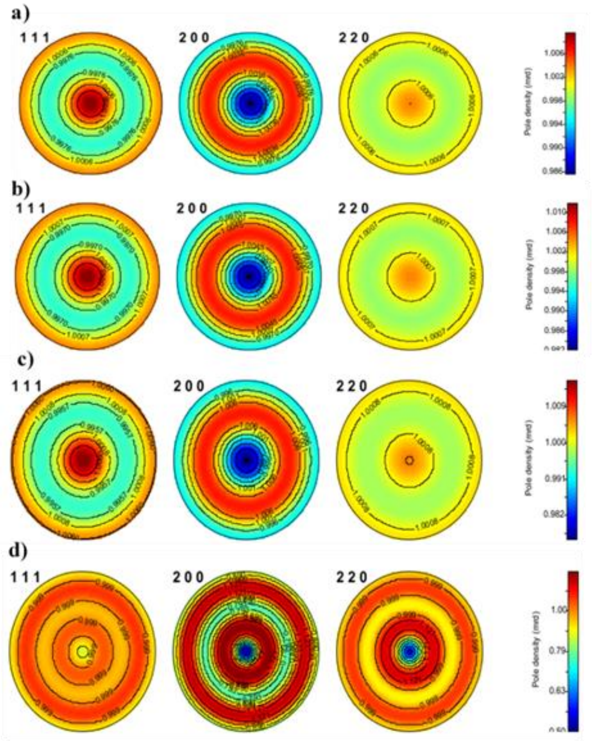


Figure 3. The simulated 2D pole figure (a) Ni-Cu 15, (b) Ni-Cu 20, (c) Ni-Cu 30, and (d) Ni-Cu 40

TABLE 2. Parameters of Ni-Cu after rietveld refinement using MAUD

	Sample			
Parameters	**Ni-Cu 15**	**Ni-Cu 20**	**Ni-Cu 30**	**Ni-Cu 40**
Crystal structure	Cubic FCC			
Space Group	Fm-3m			
Laticce constan (Å) a = b = c	3.582	3.560	3.554	3.545
Volume (Å^3)	45.975	45.152	44.905	44.557
d-spacing (Å)	1.791	1.780	1.695	1.691
Crystallite Size (nm)	10.74	15.91	16.47	20.98
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 (5) to ascertain the preferred direction of the crystal orientation of Ni-Cu deposits obtained at various current densities [28]. The texture coefficient values are shown in Table 3.

It can be seen from Figure 2(a) that the most intensive peak is the (111) plane. This indicates the (111) plane is the preferred orientation. It seems that the texture coefficient also depends on the peak current. As shown by µ values calculated in Table 3, the preferred orientation is the (111) plane. Li et al. [30] found that the higher the current density, the (111) plane is dominated. The findings suggest that the (111) crystallographic orientation is favoured in all Ni-Cu coatings electrodeposited with different current density variations in the citrate bath.

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 is determined using Equation 6, and Figure 4 illustrates the changes in crystal size and lattice strain of the Ni-Cu coating based on the current density applied in the plating bath. It is seen that the crystal size increases and the lattice strain decreases with the increasing function of the variation of current density in the plating bath. This result is the same as that obtained by Devi et al. [29], the higher the current density, the more the crystal size increases. The possible reason is that the composition of Ni increases as the current density increases.

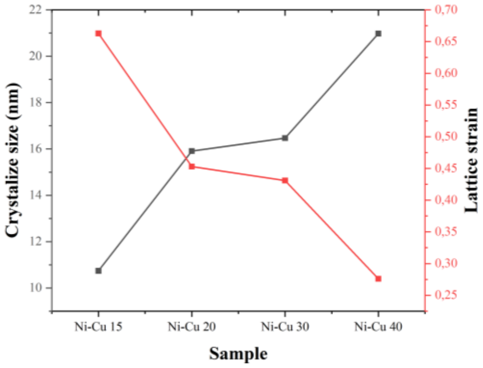


Figure 4. Ni-Cu lattice strain and size on Al at various current densities in the plating bath.

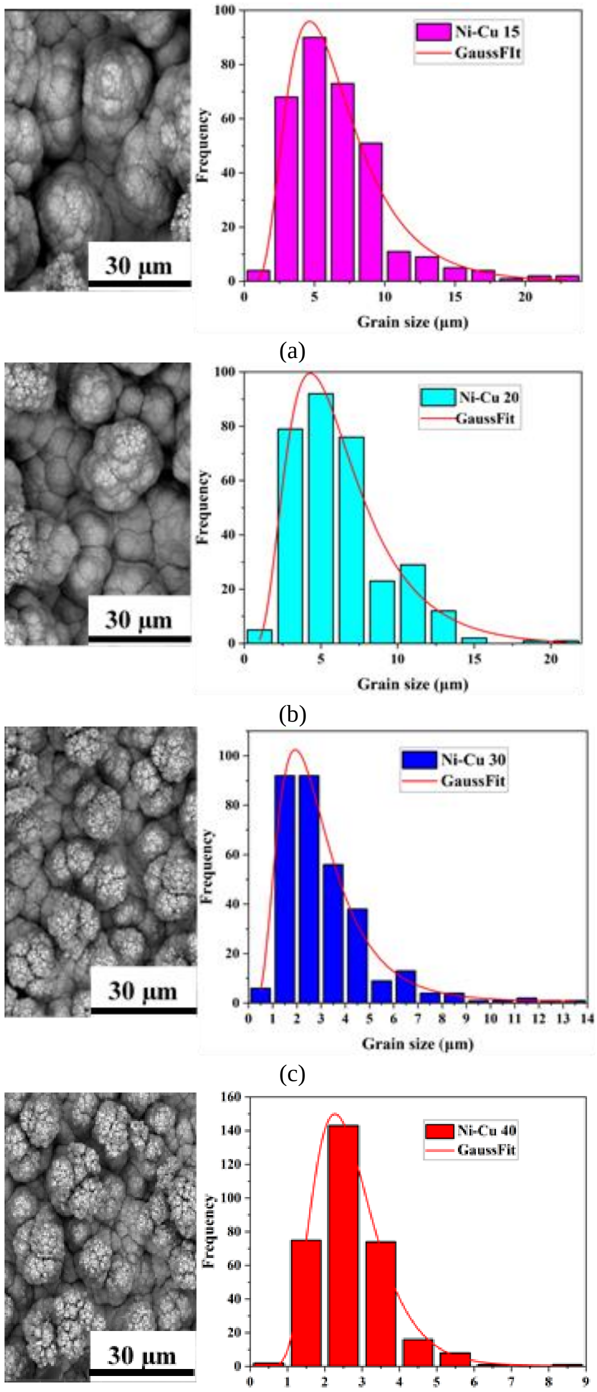
3. 3. Surface Morphological EDS was used to determine the elemental composition of the coating; Cu and Ni were the only elements present in the deposit; Table 4 reveals the Ni-Cu composition. The alloy composition is influenced by variations in the current density. As the current density increases, the deposit's Cu content decreases. This phenomenon can be caused by the bath's $[Ni^{2+}]/[Cu^{2+}]$ ratio, which changes the composition of Ni and Cu. Goranova et al. discovered that as the Cu content of the deposit fell, so did the cathodic current efficiency [32]. This phenomenon occurs because of the orderly deposition process of Ni and Cu [33]. In regular deposition, increasing current density leads to an increased proportion of less noble metals in the deposited material [34]. Ni is the less noble metal in this scenario. As a result, when the current density increases, the deposit gets richer in Ni.

Another essential feature is that as the current density increases, the peak shifts to the right, i.e. to higher 2θ values. The change in alloy composition could also cause the peak shift. The Ni concentration increases as the current density increases (see Table 4). Because Ni and Cu constitute to a single-phase alloy, the diffraction peak will shift toward pure Ni as the Ni percentage increases. This is similar to the observations of Goranova et al. [14], who found that the fcc reflection for Ni-rich Ni-Cu alloy deposits shifts to the right as the Ni concentration increases. Indeed, it is logical to expect the Ni-Cu alloy peak to change.

TABLE 4. The Chemical composition of Ni-Cu on Al coating prepared at the different current density

Samples	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, as well as the cross-section of the coated samples, were observed using SEM. Figure 5 shows SEM micrographs of the four samples' surface morphological structures. The deposits develop a fine-grained and compact spherical shape at lower deposition current densities (Figure 5(a)). Deo et al. [23] and Goranova et al. [32] also observed this morphology at low current densities. The shape shifts to a coarser cauliflower form at greater current densities (Figure 5(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 [35]. As the current density increases, the cauliflower-like protrusions become more spaced and separated, creating gaps. The Ni-Cu 40 sample with the highest current density had the largest gap between the cauliflower bulges.



(d)

Figure 5. Surface SEM image of the deposited pure Ni-Cu alloy coating and statistical distribution of grain size.

The increased nucleation rate can explain the decrease in grain size with increasing current density [36]. Ni-Cu ion flow to the cathode is faster at higher current densities. Further investigation is needed to determine the exact relationship between the coating's current density and the grain size. At a coating current density of Ni-Cu 15, Ni-Cu 20, Ni-Cu 30 and Ni-Cu 40, the peak corresponding to the (111) plane has shifted towards the right (see Figure 2b). The reduction in d-spacing is ascribed to residual stress induced at a higher deposition rate [37]. The statistical results of the grain size distribution in Figure 5 indicate that the grain size is in the range of 4.63 to 1.94 nm. The decrease in grain size with the increase in current density is evident in Table 5.

TABLE 5. The result of calculating the average grain size

Sample	Grain size average (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 6 depicts the relationship between the deposited alloy composition and the applied current density. EDS was used to determine the composition.

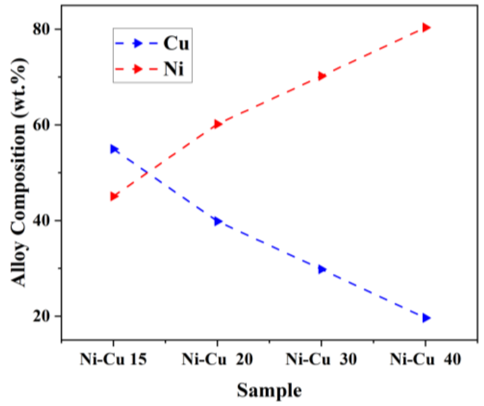


Figure 6. The dependence of the alloy composition deposited from the single electrolyte on current density

Figure 7 (a-d) displays the SEM cross-section of the Ni-Cu coating, which demonstrates appropriate adhesion between the substrate and the coating due to the absence of cracks between them. At different current densities, the thickness of the Ni-Cu coating electrodeposited was measured [23]. The thickness of Ni-Cu 15 32 µm, Ni-Cu 20 42 µm, Ni-Cu 30 49 µm, and Ni-Cu 40 50 µm. The effect of various current densities on the thickness of the Ni-Cu coating is depicted in Figure 7; the thickness increases as the current density increases [16]. As a result, a higher current density means more mass and a higher coating. Compared to Figure 1, it can be seen a shift to the higher current density on electrodeposited Ni-Cu coatings, delivering a higher cathodic current efficiency.

According to Faraday's law, when the deposition time remains constant for all samples, coatings formed at lower current densities will have a lesser thickness than those produced at higher current densities. A thinner covering may cause severe interference from the Al substrate. The cathodic current density also affects the coating, as Deo et al. [23] discovered that increasing the current density improves the thickness of the film due to an increase in cathodic current efficiency.

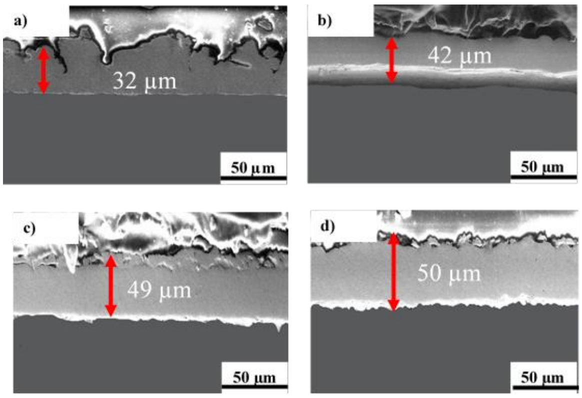
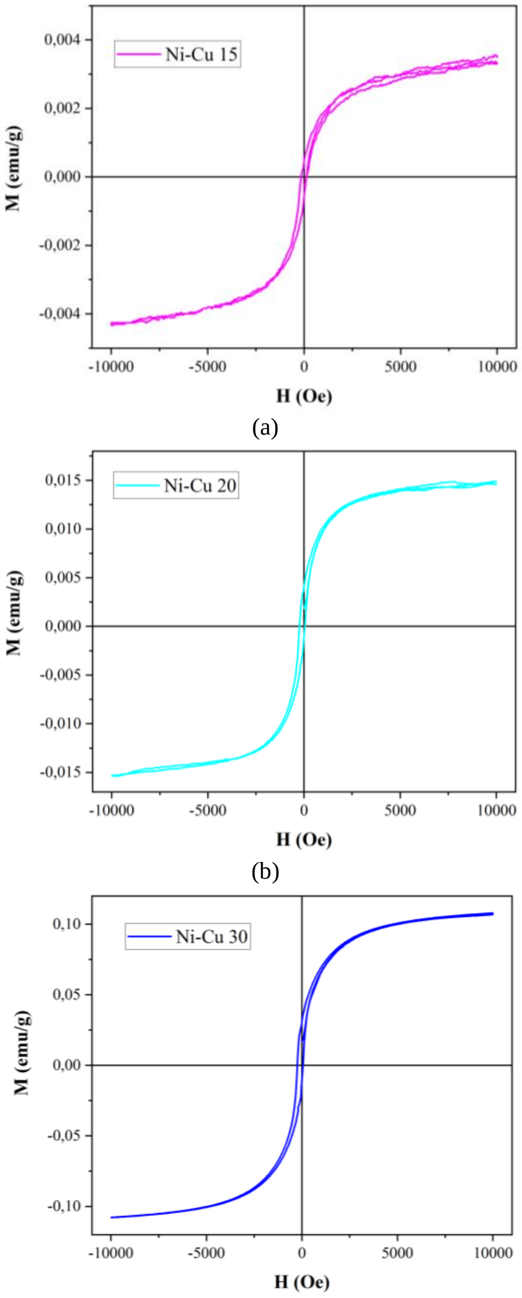


Figure 7. SEM cross sectional of the Ni-Cu over Al at (a) Ni-Cu 15, (b) Ni-Cu 20, (c) Ni-Cu 30, and (d) Ni-Cu 40

3. 4. Magnetic Properties

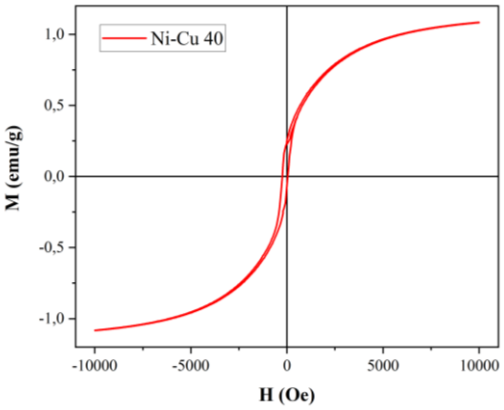
Figure 8 depicts the magnetic characteristics and fluctuations in current density magnetization measured with a VSM at room temperature [38]. The VSM analysis demonstrated that the coatings created using Ni-Cu 15, Ni-Cu 20, Ni-Cu 30, and Ni-Cu 40 displayed ferromagnetic activity among the four types of Ni-Cu alloy films studied. The low ferromagnetic activity of the $Cu_{54.95}Ni_{45.05}$ alloy film at Ni-Cu 15 could be attributed to Ni diffusion in the Cu matrix, as Cu is diamagnetic and Ni is a ferromagnetic metal [39]. The ferromagnetic properties of the Ni-Cu films at Ni-Cu 20, Ni-Cu 30, and Ni-Cu 40 were discovered, and the ferromagnetic properties increased with the increasing in Ni content of the alloy coatings.



(a)

(b)

(c)



(d)

Figure 8. Hysteresis loops of multilayers grown at different various current density

As the Ni content of Ni-Cu alloy coatings increases, so does their saturation magnetization (see Table 6). Wang et al. reported on the dependence of saturation magnetization on the Ni content of Ni-Cu alloy coatings [37]. Awasthi reported that magnetization is enhanced by increasing the coating thickness [19]. The possible reason for this magnetization-enhanced phenomenon might be the magnetic disorder caused by the coating. The trend in the saturation magnetization enhancement is associated with the coating level [40]. Demidenko et al. [41] found that monel has paramagnetic properties at room temperature, while the Ni-Cu alloy in this study has ferromagnetic properties. The current study was able to improve on the results of previous studies that added phosphorus (P) and tungsten (W) [22]. Therefore the Ni-Cu 40 sample produces the highest magnetic properties.

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

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 9 depicts the dependence of the microhardness of Ni-Cu coatings on the variation of current density in the plating bath. From Figure 9, it is clearly visible that the microhardness of Ni-Cu 40 coatings is higher than else. In general, it increases with the variation in current density in the plating bath. Ni-Cu 40 coating shows a maximum microhardness value of 404 HV. The increase in microhardness coatings is attributed to the grain size refinement and thickness. Pingale et al. found that hardness increases with the thickness of the coating [16]. The thickness could influence the hardness of Ni-Cu films [42].

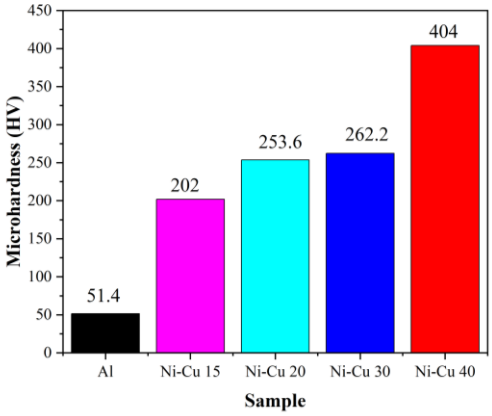


Figure 9. Variation in microhardness electrodeposited Ni-Cu coatings at various current densities

A larger percentage of Ni indicates 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 10, indicating that microhardness increases. This result is similar to what Marenych found: the hardness value is highest with the highest Ni composition [43].

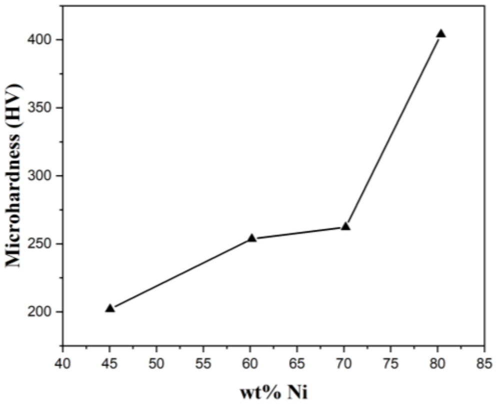


Figure 10. Relation wt% Ni vs microhardness

Moreover, compared to previous studies, the electrodeposition of Ni-Cu over Al by adding phosphorus (P) resulted in lower hardness than the present study [21]. This is due to the smaller grain size resulting from the change in current density. In addition, the increase in microhardness is related to the role of Ni atoms in grain refinement [44]. Ramkumar et al. [45] reported a monel hardness value of 165 HV. This result is lower than the hardness value in the current study.

4. CONCLUSION

Ni-Cu atoms were electrodeposited onto Al substrates using citrate baths in the present study. The effects of adding citrate solutions on the properties of the deposited layers were studied, and the following results are reported:

1. Electrodeposition was used to create single-phase Ni-Cu alloy coatings on Al surfaces at all current densities.
2. The cathodic current efficiency is affected by hydrogen evolution. The cathodic current efficiency increases if the metal deposition rate exceeds the hydrogen evolution rate.
3. Coatings formed at lower current densities exhibit a more compact and globular morphology, whereas those formed at higher current densities display a less uniform structure with a cauliflower-like morphology.
4. Both the surface morphology and composition of the coating showed a strong dependence on the current density.
5. Ni-Cu alloy samples deposited at low current densities have a coating thickness of 32 μm, while samples deposited at high current densities have a coating thickness of 50 μm.
6. Saturation magnetization increases with the increase in Ni content in the Ni-Cu alloy and also with the thickness of the coating.
7. Hardness increases as the current density increases. Hardness increases as the composition of Ni in the solution increases. In addition, hardness is also affected by grain size and coating thickness. The hardness of the Ni-Cu alloy is harder than that of monel.

5. ACKNOWLEDGMENTS

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Effect of Current Density on Magnetic and Hardness Properties of Ni-Cu Alloy Over Al Substrate by Electrodeposition Method

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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.

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NOMENCLATURE

C_e~~k~~	Cathodic current efficiency~~Constant of 0.9~~	W_i	Initial weight of the substrate
σ~~β~~	Lattice strain~~Full width at half maximum (FWHM)~~	W_f	Weight
W_s~~λ~~	Final weight of the substrate~~X-ray wavelength (Cu Kα = 0.154 nm)~~	I	Total current
W_m~~θ~~	Ratio of the final weight of the substrate~~Bragg's angle~~	t	Deposition time
μ	Texture coefficient of the unique plane	F	Faraday's constant
$I(hkl)$	Measured intensity	f_{ni}	Nickel deposit weight ratio
m_{cu}~~C_e~~	Copper's atomic weight~~Cathodic current efficiency~~	m_{ni}	Nickel's atomic weight

1. INTRODUCTION

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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/cm^2 [24], and Karunakaran and Pugazh Vadivu reported a magnetic saturation value of 0.0004 emu/g at 40 mA/cm^2 [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 ~~pure~~ 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 ~~pure~~ 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 a current density of 15 mA/cm^2, 20 mA/cm^2, 30 mA/cm^2, and 40 mA/cm^2 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
$NiSO_4.6H_2O$ (Merck)	0.5 M
$CuSO_4.5H_2O$ (Merck)	0.04 M
$Na_3C_6H_5O_7$ (Merck)	0.2 M
pH	4.2
Temperature	25 °C
Deposition time	1 h

2. 2. Characterization

The ~~cathodic current efficiency (CCE)~~deposition rate was calculated using the previously reported method [26] ~~[24]~~. The following

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formula, Equation 1, was used to calculate the efficiency of the cathodic current [27]:

$$C_e = {W_m}/{W_f} \tag{1}$$

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

$$W_m = W_s - W_i, \tag{2}$$

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

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 crystallite size was calculated using the Debye-Scherrer formula, as shown in Equation 4 [24]:~~

~~$D = \frac{0.9\lambda}{\beta cos\theta}$ (4)~~

The preferential crystallite orientation was determined from the texture coefficient μ, as shown in Equation ~~5~~ 4 [28]:

$$\mu = \frac{I(hkl)/I_0(hkl)}{(\frac{1}{N})\sum[I(hkl)/I_0(hkl)]}, \qquad (~~5~~\ 4)$$

Based on the XRD results, the lattice strain σ was calculated using Equation (~~6~~ 5) [29]:

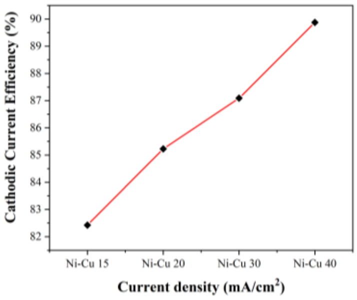
$$\sigma = \left(\frac{\beta}{4 \times \tan\theta}\right) \qquad (~~6~~\ 5)$$

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 ~~and current density~~ 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/cm^2; 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.



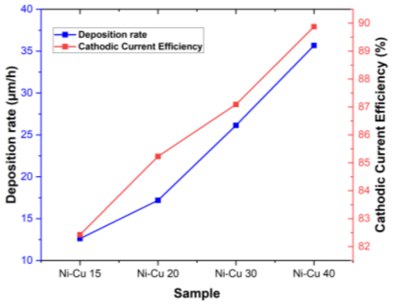


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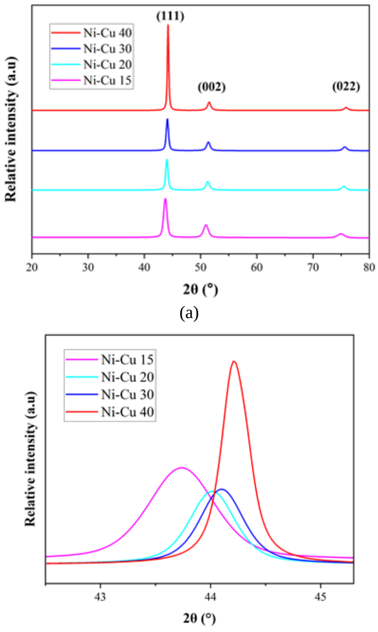
Figure 1. The cathodic current efficiency and deposition rate at a current density of 15, 20, 30, and 40 mA/cm^2

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 = 43.3° for pure Cu and 2θ = 44.5° for pure Ni [32, 33]. As the Ni content of the alloy coating increased, the diffraction angle also increased.



(a)

(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 ~~The Debye-Scherrer formula (Equation 4)~~ 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 ~~10~~ 24 to ~~20~~ 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/cm^2 was found to have a strong (111) texture.

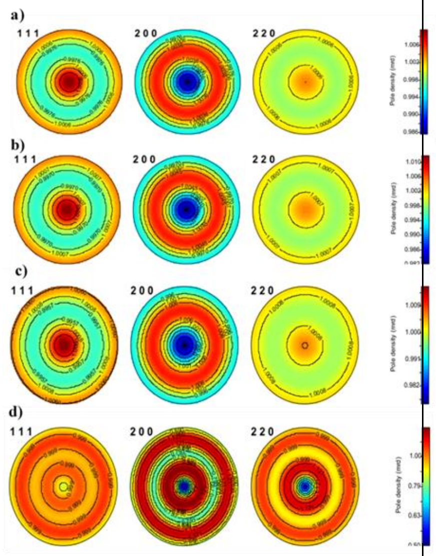


Figure 3. The simulated 2D pole figures for the (a) Ni-Cu 15, (b) Ni-Cu 20, (c) Ni-Cu 30, and (d) Ni-Cu 40 samples

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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 (Å^3)	45.975	45.152	44.905	44.557
d-spacing (Å)	1.791	1.780	1.695	1.691
Crystallite size (nm)	~~10.74~~ 24.55	~~15.91~~ 26.82	~~16.47~~ 32.29	~~20.98~~ 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 ~~5~~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~~6, 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.

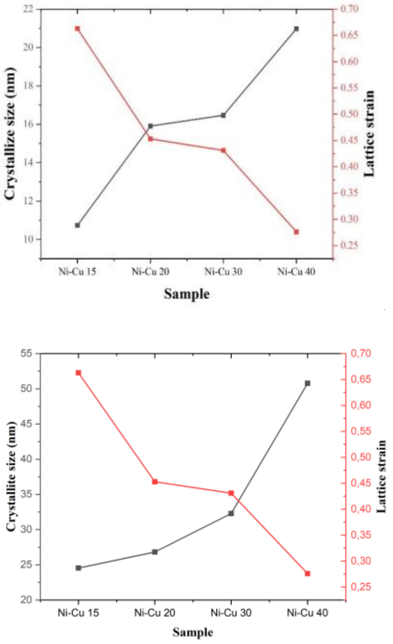

Figure 4. The lattice strain and crystallize size of the Ni-Cu alloys coated on Al at various current densities

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 [Ni^{2+}]/[Cu^{2+}] 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

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The change in composition is very impressive.

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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.

~~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.~~

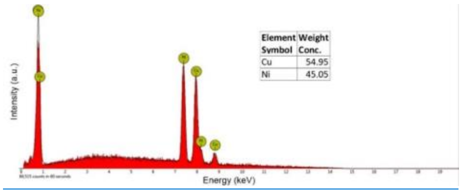


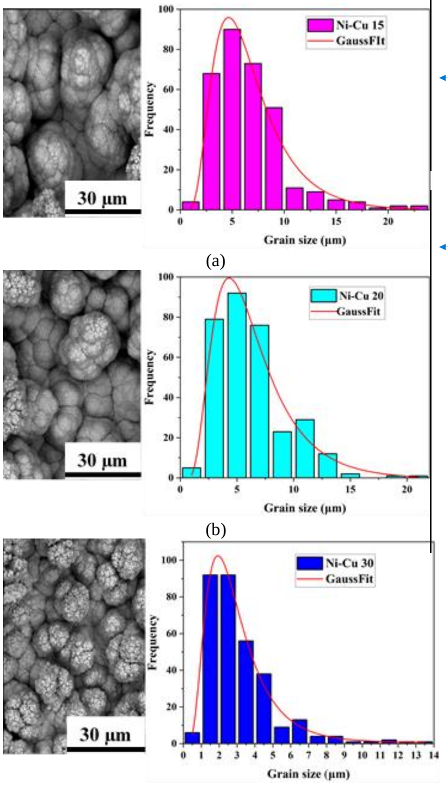
Figure 5. Eds graph of the Ni-Cu 15

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~~5~~ 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~~5~~(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.



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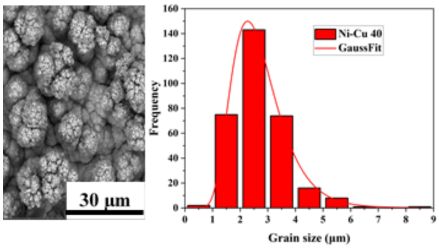

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Figure 6~~5~~. Surface SEM images of the deposited Ni-Cu alloy coatings and plots showing the statistical distribution of the grain size

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~~5~~ 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~~6~~ depicts the relationship between the deposited alloy composition and the applied current density. EDS was used to determine the composition.

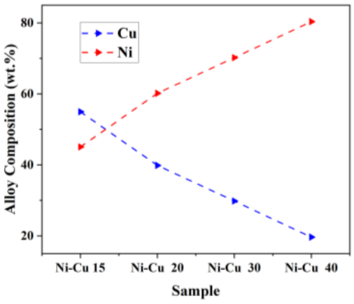

Figure 7~~6~~. The dependence of the deposited alloy's composition (shown as wt% of the single electrolytes) on current density

Figure ~~7~~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~~7~~; 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.

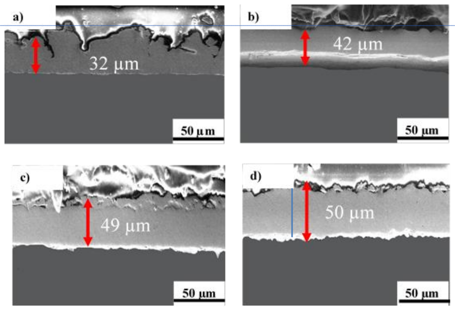

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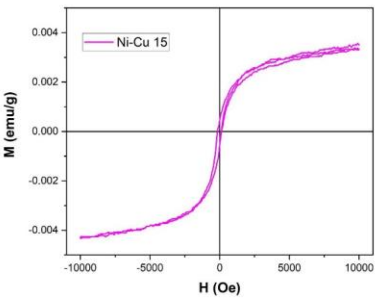
Figure ~~7~~8. SEM cross-section images of the Ni-Cu coating on Al in the (a) Ni-Cu 15, (b) Ni-Cu 20, (c) Ni-Cu 30, and (d) Ni-Cu 40 samples

3. 4. Magnetic Properties

Figure 9~~8~~ 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 $Cu_{54.95}Ni_{45.05}$ 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.



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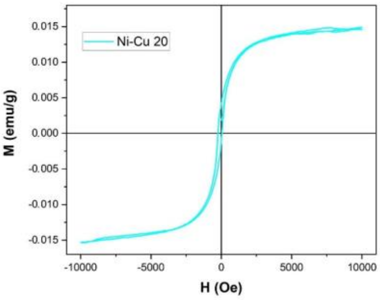

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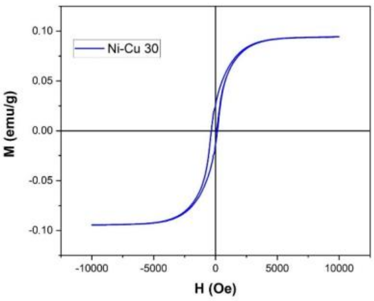

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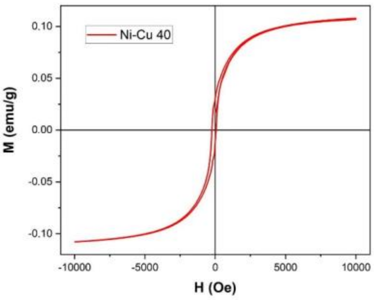

(d)

Figure 9~~8~~. 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~~9~~ 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~~9~~, 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].

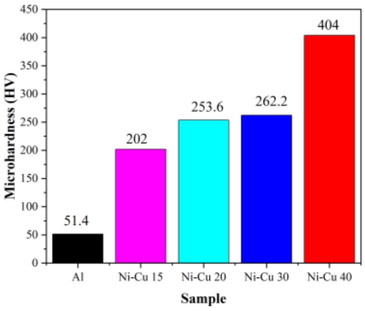

Figure ~~9~~10. The microhardness of the electrodeposited Ni-Cu coatings of the samples produced with different current densities

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~~0~~, 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].

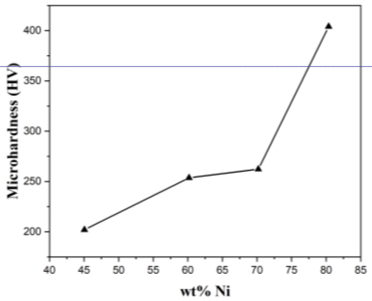

Figure 11~~0~~. The relationship found between wt% Ni and microhardness, based on the data obtained from the four experimental samples

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 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

The authors extend their gratitude to the Ministry of Research, Technology and Higher Education of the

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Republic of Indonesia for the financial support of Hibah Penelitian Disertasi Doktor No: NKB-971/UN2.RST/HKP.05.00/2022.

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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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3. Bukti menjawab reviewer
(7 September 2023)

Title : Effect of Current Density on Magnetic and Hardness Properties of Ni-Cu Alloy Coated on Al via Electrodeposition
Authors : Cahaya Rosyidan, Budhy Kurniawan, Bambang Soegijono, Valentinus Galih Vidia Putra, Dicky Rezky Munazat, Ferry Budhi Susetyo
Corresponding author : Budhy Kurniawan

#Editorial Office of International Journal of Engineering

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></th><th colspan="4">Sample</th></tr>
<tr><th>Parameter</th><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>
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<table>
<tr><td></td><td></td><td>
<table>
<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>
<tr><td>Crystallite size (nm)</td><td>24.55</td><td>26.82</td><td>32.29</td><td>50.78</td></tr>
<tr><td>Rwp (100%)</td><td>3.640</td><td>4.484</td><td>5.394</td><td>5.139</td></tr>
<tr><td>GOF</td><td>1.94</td><td>1.72</td><td>2.06</td><td>2.03</td></tr>
<tr><td>Lattice strain</td><td>0.663</td><td>0.453</td><td>0.431</td><td>0.276</td></tr>
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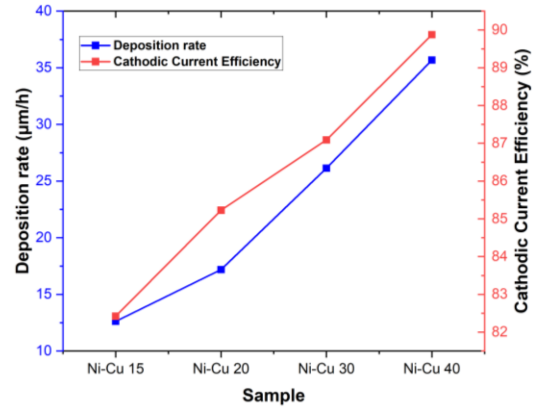
</td></tr>
<tr><td>4</td><td>is this consistent with similar research?</td><td>
Yes, this is consistent with other studies, we add the deposition rate graph and the same study.



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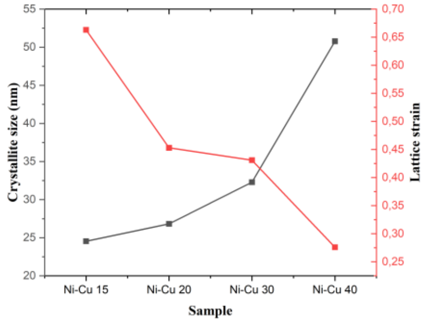
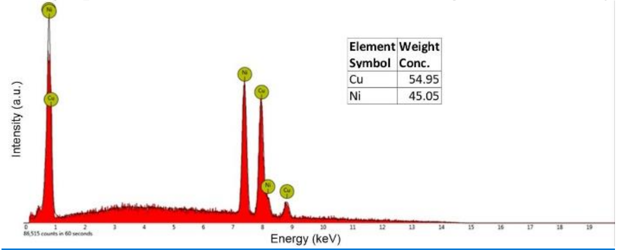
<tr><td>5</td><td>Why is there a significant difference in the results for Ni-Cu 40 sample?</td><td>
According to Figure 2(b), the FWHM for peak (111) decrease with increasing current density. This result is consistent with earlier studies in which increasing current density reduces FWHM. DOI:

10.24203/ajas.v8i4.6287



</td></tr>
<tr><td>6</td><td>Is eds accurate enough to determine chemical composition?
The change in composition is very impressive</td><td>
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
</td></tr>
</table>

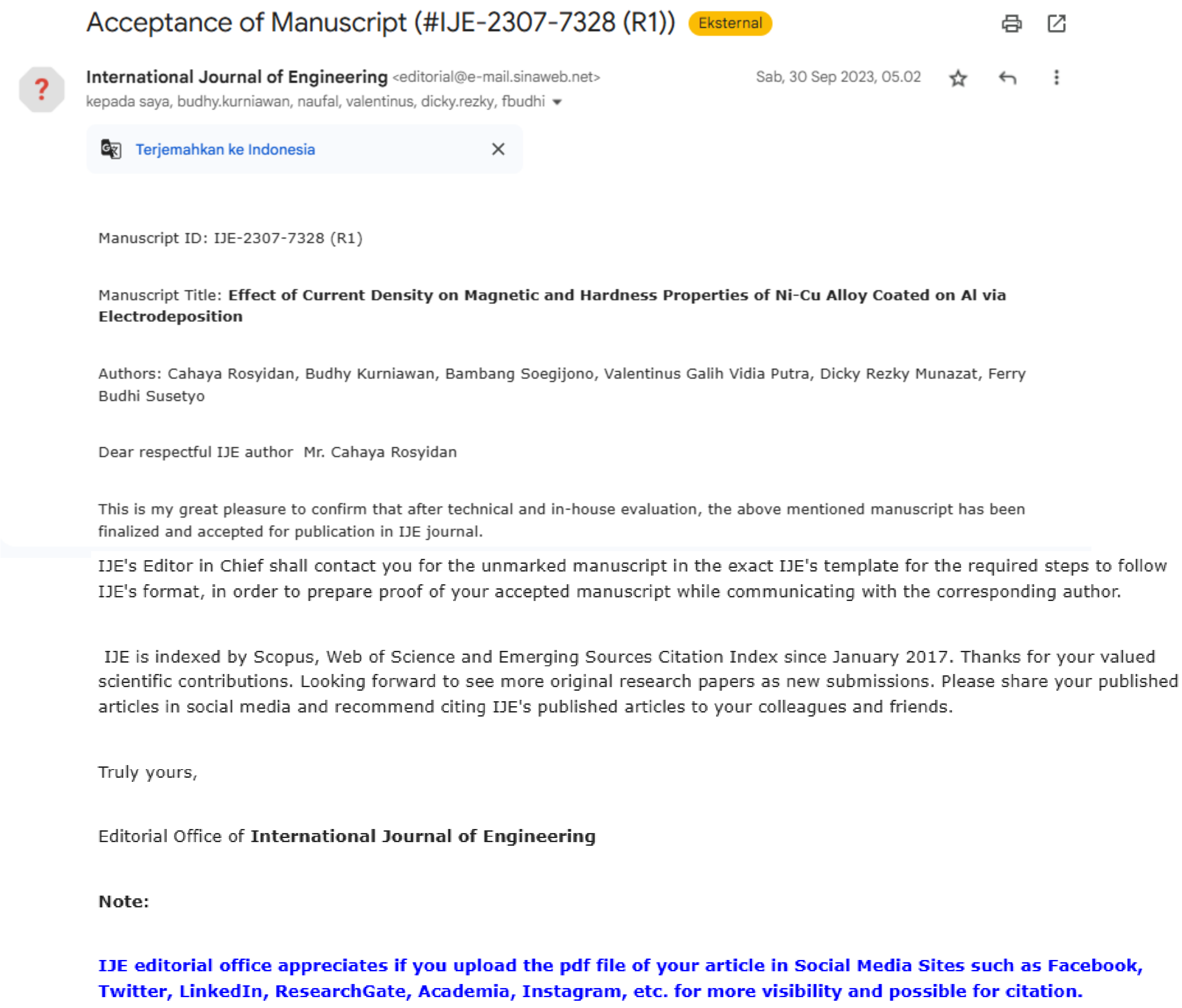
		temperatures, resulting in the potential loss of analytes by evaporation or as particulates in the smoke. Can only measure metals in total, cannot distinguish between species such as oxidation state, free metal or bound to other molecules such as metalloproteins, organometallics, etc.
7	EdS graph and 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)

4. Keputusan Editor
(30 September 2023)

Bukti dari Email



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Authors: Cahaya Rosyidan, Budhy Kurniawan, Bambang Soegijono, Valentinus Galih Vidia Putra, Dicky Rezky Munazat, Ferry Budhi Susetyo

Dear respectful IJE author Mr. Cahaya Rosyidan

This is my great pleasure to confirm that after technical and in-house evaluation, the above mentioned manuscript has been finalized and accepted for publication in IJE journal.

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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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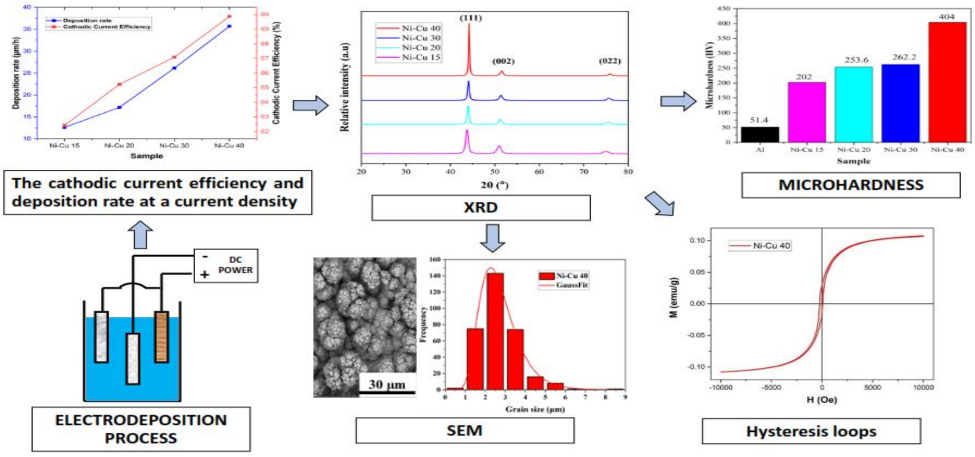
ABSTRACT

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.2024.37.02b.01*

Graphical Abstract



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NOMENCLATURE

C_e	Cathodic current efficiency	W_i	Initial weight of the substrate
σ	Lattice strain	W_f	Weight
W_s	Final weight of the substrate	I	Total current
W_m	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	f_{ni}	Nickel deposit weight ratio
m_{cu}	Copper's atomic weight	m_{ni}	Nickel's atomic weight

1. INTRODUCTION

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. (17) 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. 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). Karunakaran et al. (24) reported a hardness of 153 HV when the current density was 40 mA/cm^2, and Karunakaran and Pugazh (25) Vadivu reported a magnetic saturation value of 0.0004 emu/g at 40 mA/cm^2. 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 a current density of

15 mA/cm^2, 20 mA/cm^2, 30 mA/cm^2, and 40 mA/cm^2 were designated as Ni-Cu 15, Ni-Cu 20, Ni-Cu 30, and Ni-Cu 40, respectively. Table 1 summarized the bath composition and deposition parameters.

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):

$$C_e = {W_m}/{W_f} \tag{1}$$

W_m and W_f were calculated using Faraday's law, as shown in Equations 2 and 3.

$$W_m = W_s - W_i, \tag{2}$$

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

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):

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

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

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

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.

TABLE 1. Bath composition and deposition parameters

Bath composition and condition	Quantity
$NiSO_4.6H_2O$ (Merck)	0.5 M
$CuSO_4.5H_2O$ (Merck)	0.04 M
$Na_3C_6H_5O_7$ (Merck)	0.2 M
pH	4.2
Temperature	25 °C
Deposition time	1 h

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/cm^2; 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. and Syamsuir et al. (31) found that the deposition rate and current efficiency are considered linear.

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

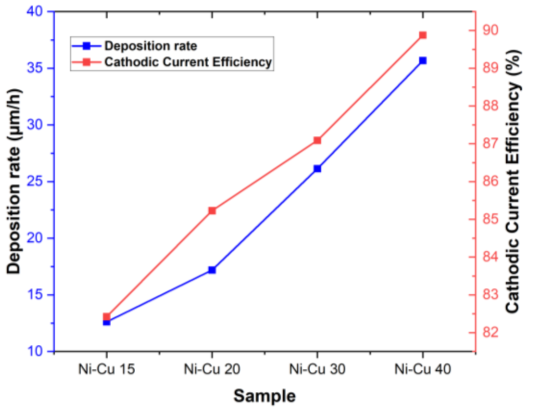
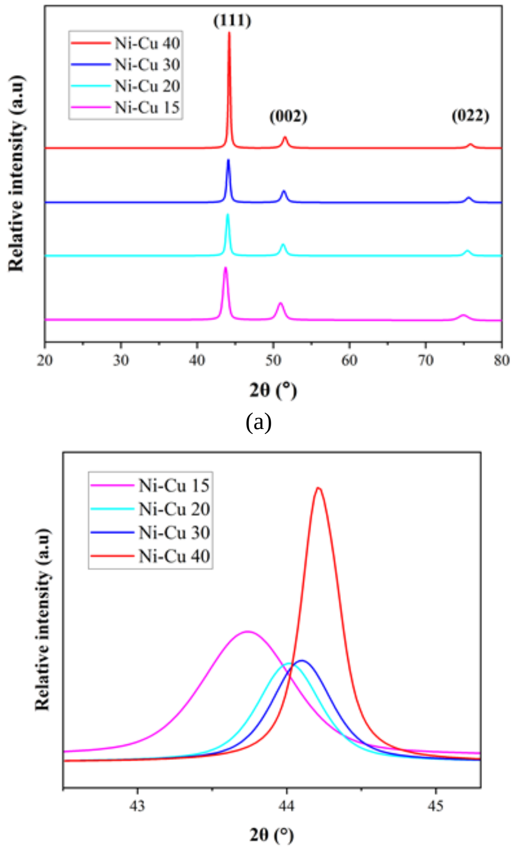


Figure 1. The cathodic current efficiency and deposition rate at a current density of 15, 20, 30, and 40 mA/cm^2



(a)

(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

peaks of each sample were found between the peaks of the fcc of 2θ = 43.3° for pure Cu and 2θ = 44.5 ° for pure Ni (32, 33). As the Ni content of the alloy coating increased, the diffraction angle also increased.

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/cm^2 was found to have a strong (111) texture.

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.

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

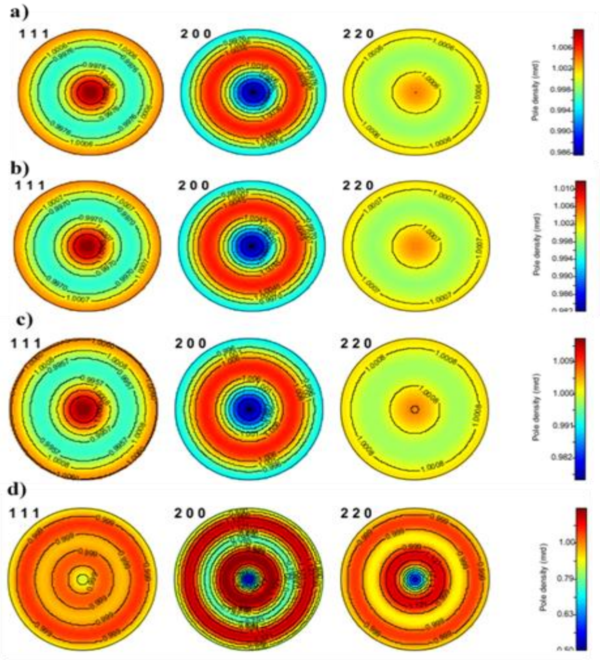


Figure 3. The simulated 2D pole figures for the (a) Ni-Cu 15, (b) Ni-Cu 20, (c) Ni-Cu 30, and (d) Ni-Cu 40 samples

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 (Å^3)	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

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

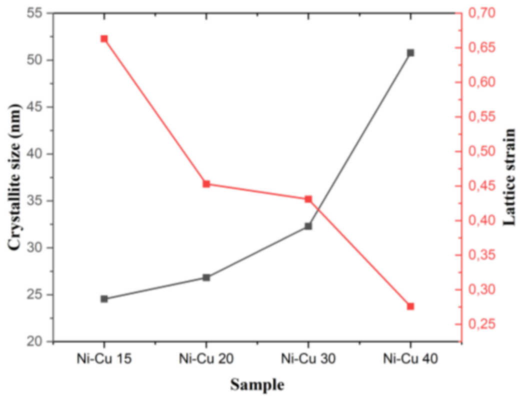


Figure 4. The lattice strain and crystallize size of the Ni-Cu alloys coated on Al at various current densities

crystal size increased. A possible reason for this is that the composition of Ni increases as the current density increases.

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 [Ni^{2+}]/[Cu^{2+}] 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.

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.

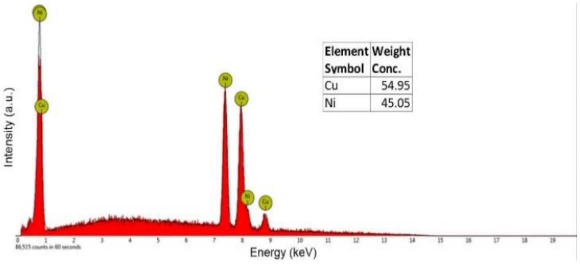


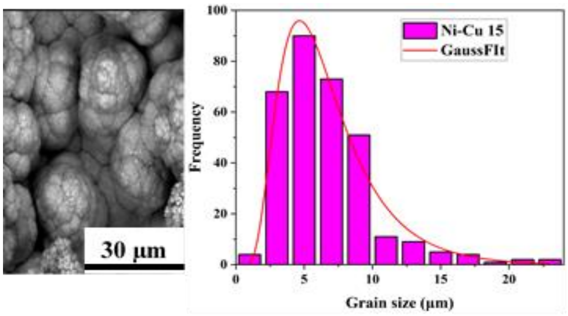
Figure 5. Eds graph of the Ni-Cu 15

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

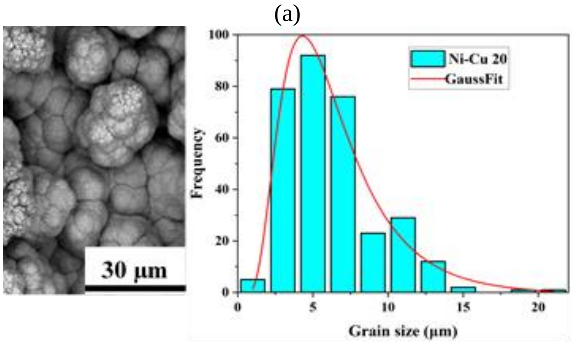
Sample	Cu, wt%	Ni, wt%
Ni-Cu 15	54.95	45.05
Ni-Cu 20	39.83	60.17
Ni-Cu 30	29.80	70.20
Ni-Cu 40	19.64	80.36

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.

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.



(a)



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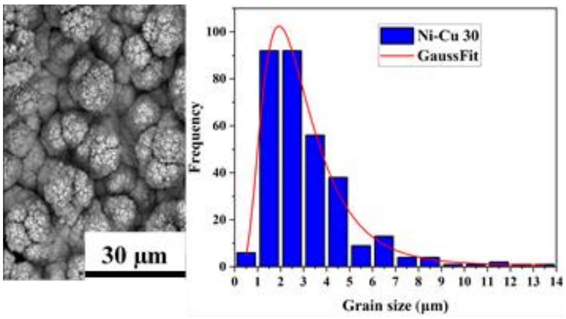

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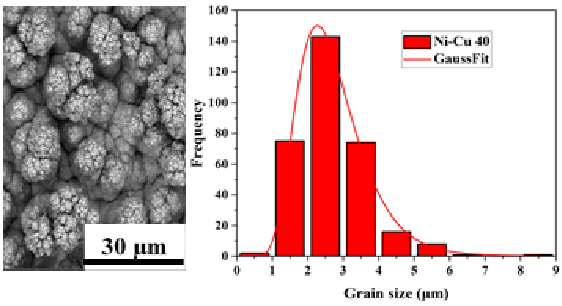


(d)

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

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

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

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

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.

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 $Cu_{54.95}Ni_{45.05}$ 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.

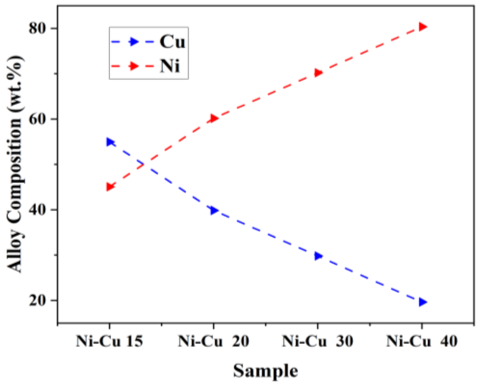


Figure 7. The dependence of the deposited alloy's composition (shown as wt% of the single electrolytes) on current density

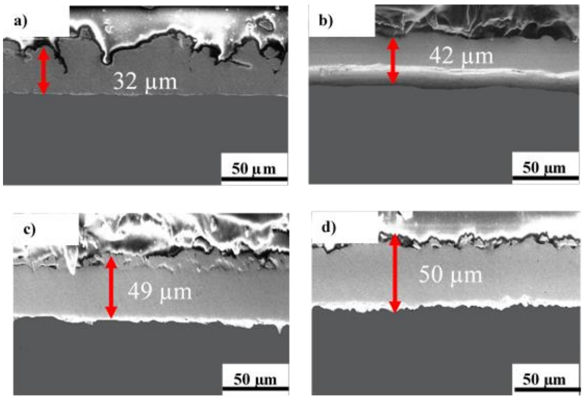
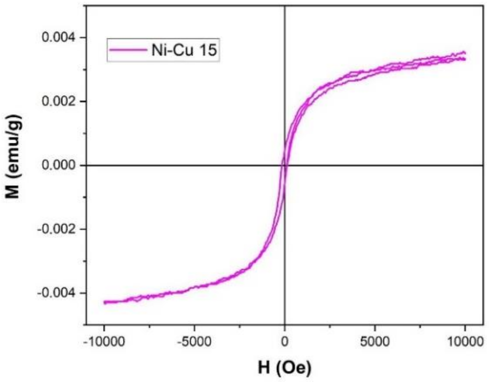
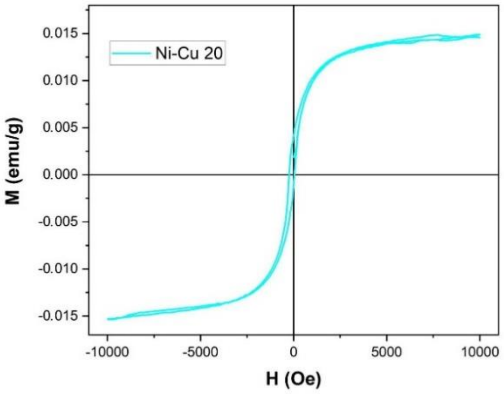


Figure 8. SEM cross-section images of the Ni-Cu coating on Al in the (a) Ni-Cu 15, (b) Ni-Cu 20, (c) Ni-Cu 30, and (d) Ni-Cu 40 samples



(a)



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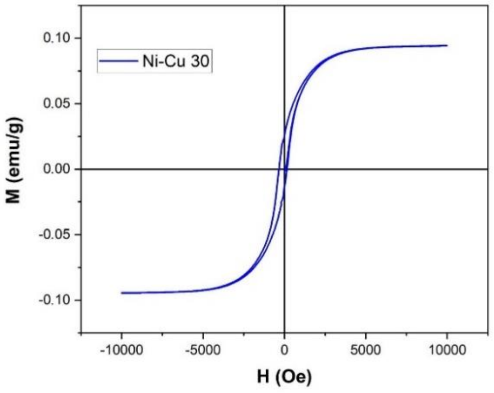

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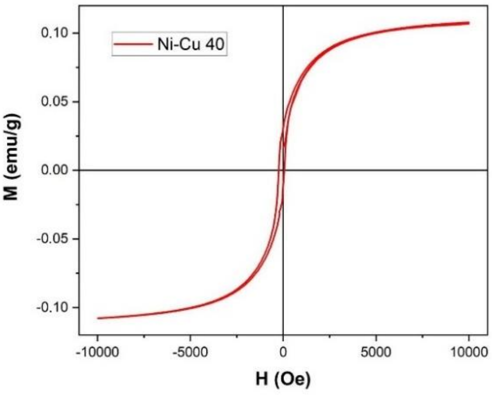

(d)

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. (43) reported that saturation magnetization depends on the Ni content of Ni-Cu alloy coatings. In addition, Awasthi (22) reported that magnetization is enhanced by increasing the coating thickness. 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.

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. (19) found that hardness increased with the thickness of the coating. The coating thickness could influence the hardness of Ni-Cu films (26).

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

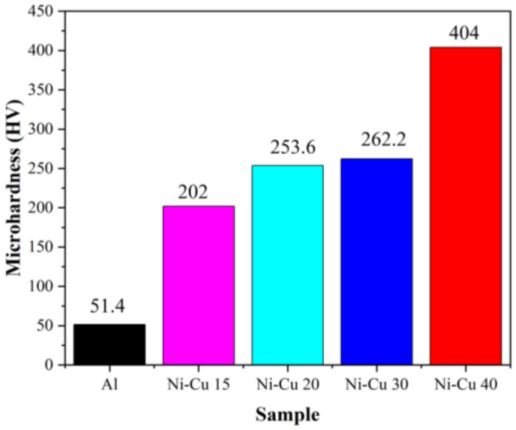

Figure 10. The microhardness of the electrodeposited Ni-Cu samples produced with different current densities

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 (49) finding that the hardness value is highest with the highest Ni composition.

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.

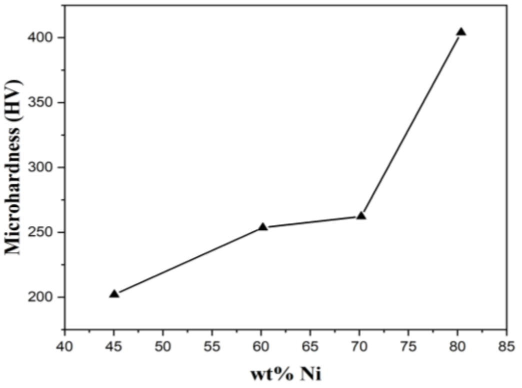

Figure 11. The relationship found between wt% Ni and microhardness, based on the data obtained from the four experimental samples

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 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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Persian Abstract

چکیده

پوشش‌های آلیاژی تک فاز نیکل مسغنی از نیکل (Ni) بر روی بسترهای آلومینیومی (Al) با رسوب الکتریکی در حمام سیترات تثبیت شده تولید شدند. آزمایش‌های رسوب الکتریکی در چهار چگالی جریان مختلف انجام شد. افزایش چگالی جریان منجر به افزایش سرعت رسوب فلز سریعتر از نرخ تکامل هیدروژن شد. بنابراین، راندمان جریان کاتدی افزایش یافت. سیستم‌های کریستالی آلیاژهای Ni-Cu مکعبی در مرکز سطح (FCC) بودند، با صفحه (۱۱۱) به عنوان صفحه کریستالی ترجیحی. میکروسکوپ الکترونی روبشی با اندازه‌گیری‌های طیف‌سنجی پرتو ایکس پراکنده انرژی (SEM-EDS) نشان داد که محتوای نیکل در پوشش با افزایش چگالی جریان افزایش می‌یابد. نمونه Ni-Cu 40 بیشترین مقدار نیکل را داشت و مورفولوژی همگن و فشرده را نشان داد. مشخص شد که هر چه غلظت نیکل در محلول بیشتر باشد، اندازه دانه کوچکتر است. اندازه‌گیری‌های ثبت‌شده با یک مغناطیس‌سنج نمونه ارتعاشی (VSM) نشان داد که نمونه Ni-Cu 40 اشباع مغناطیسی را ارائه می‌کند، با بالاترین مقدار ۰.۱۰۸ emu/g. روش میکروسختی ٤٠٤ HV بر روی نمونه Ni-Cu 40 تولید کرد. در نتیجه، چگالی جریان بالاتر با ترکیب نیکل بالاتر و افزایش ضخامت همراه بود که مسئول افزایش خواص مغناطیسی و سختی بود.