

The Effect of Electrodeposition Current and Electrolyte Composition on Electrodeposition of Fe-Ni Alloys

Ghina Imani Rofi, Maria Paristiowati*

Department of Chemistry, Faculty of Mathematics and Natural Sciences, Universitas Negeri Jakarta
Jl. Rawamangun Muka, Jakarta 13320, Indonesia

*Corresponding author: maria.paristiowati@unj.ac.id

Received

30 May 2022

Received in accepted form

23 June 2022

Accepted

23 June 2022

Published online

30 June 2022

DOI

<https://doi.org/10.56425/cma.v1i2.27>



Original content from this work may be used under the terms of the [Creative Commons Attribution 4.0 International License](https://creativecommons.org/licenses/by/4.0/).

Abstract

This research was conducted to determine the effect of solution variations, electrical current, thickness, weight, metal composition, and the characteristics of the Fe-Ni alloy synthesized using the electrodeposition method. The instruments used in this study were atomic absorption spectroscopy (AAS) to determine the metal composition in the deposit, X-ray diffraction to determine the diffraction patterns, and scanning electron microscopy (SEM) to observe morphology and energy dispersive X-ray spectroscopy for elemental analysis. The SEM micrographs obtained showed that morphology of the Fe-Ni alloy was in the form of refined grains. The percentage of Fe in the deposit decreases with the increase in the used current. The AAS data also showed that a high Ni composition in solution will affect the atomic percent of each metal in the deposit. The formation of the alloy was confirmed by diffraction peaks at 2θ of 44° , 53° , and 76° that associated with reflection planes of the face centered cubic structure of Fe-Ni with the crystallite size range from 5 nm to 35 nm.

Keywords: alloy, Fe-Ni, electrodeposition

1. Introduction

The magnetic alloy of Fe-Ni nanoparticles attracts much attention due to various applications in electronic devices [1][2][3][4]. The Fe-Ni nanoparticles can be synthesized using chemical or electrochemical methods known as electrodeposition. The electrodeposition method is preferred between the two methods because the synthesis process is more straightforward and requires fewer reagents so that relatively less waste compared to that of chemical synthesis [2].

The Fe-Ni alloy nanoparticles synthesized using the electrodeposition method were obtained in the form of a thin film with a thickness that could be adjusted through time and the applied current. The Fe-Ni alloy nanoparticle film thickness is directly proportional to the current flowing and the deposition time. The properties of an alloy particle are primarily determined by the phase, structure, and composition of each element in the resulting alloy [5]. Indirectly, the magnetic properties of the Fe-Ni alloy are also influenced by the electrodeposition parameters and the characteristics of the substrate used. Based on a previous [6], there is a causal correlation between current

and alloy composition on the crystal structure of the resulting Fe-Ni alloy. When the Fe-Ni alloy has a higher Fe composition, the crystal structure obtained tends to be body-centered cubic (BCC). On the other hand, when the Ni composition in the alloy is higher, the crystal structure tends to face center cubic (FCC) [6].

The electrodeposition method can be carried out with a constant current or constant potential during the process [7]. The electrodeposition method using a constant current during the process is preferred because it can produce a more homogeneous thin film that is firmly attached to the substrate than that prepared with a constant potential during the process [8][9]. In this paper, the Fe-Ni alloys were electrodeposited with various applied currents and deposition times from different electrolyte compositions. The effect of deposition conditions on deposit thickness, chemical composition, and deposit structures are reported.

2. Materials and Method

The synthesis of Fe-Ni alloy was carried out by electrodeposition method using a three-electrode

electrochemical cell with an external current source originating from the eDAQ modular potentiostat and an electrolyte solution in the form of a metal solution. The metal solution used in this electrodeposition process consists of two types of solutions with varying concentrations of iron and nickel. The ratio of the concentrations of iron and nickel in the first solution was 0.04 M and 0.06 M, while in the second solution, it was 0.01 M and 0.09 M. The two metal solutions used were added with buffers and additives in the form of H_3BO_3 (boric acid) 0.4 M and $C_7H_4NNaO_3S$ (sodium saccharin) 0.009 M.

The ITO (Indium Tin Oxide) substrate used for the Fe-Ni alloy electrodeposition process is rectangular with a width of 1 cm and a length of 2.5 cm. Before being used as a substrate, ITO was first rinsed using ethanol and distilled water. After rinsing, the ITO substrate was dried at room temperature. Then the ITO substrate was weighed using an analytical balance to determine the weight of the substrate. The electrodeposition process was carried out using three electrodes consisting of an Ag/AgCl reference electrode, a Pt auxiliary electrode, and a working electrode or ITO substrate. The three electrodes are mounted in a series of electrochemical cells. A thin film of Fe-Ni alloy was synthesized using the electrodeposition method with variations in current and time. The currents applied during deposition were 10 mA, 25 mA, and 50 mA. The deposition times were varied to 1, 5, and 10 minutes. Then the Fe-Ni alloy deposited on the ITO substrate was rinsed using distilled water. The obtained samples were characterized by Atomic Absorption Spectroscopy (AAS) to analyze the composition percentage of Fe-Ni, Scanning Electron Microscope (SEM) JSM-6510LA SEM JEOL, and Energy Dispersive X-Ray Spectroscopy (EDX) JEOL type JED -2300 to see the micrograph and composition of the sample, analyze the phase with X-Ray Diffractometer (XRD), and modular potentiostat eDAQ.

3. Results and Discussion

The metal alloy between iron and nickel or Fe-Ni has a characteristic grey color and has been successfully synthesized on a clear or transparent Indium Tin Oxide (ITO) substrate. Based on Figure 1, it can be seen that along with the increase in the current, there was an increase in the thickness of the Fe-Ni alloy electrodeposition on the ITO substrate for 5 minutes. In the previous research, a similar experiment with time variations of 1, 5, and 10 minutes resulted in different weights of Fe-Ni alloys. This happens because the electrodeposition time is one of the parameters that affect the properties and characteristics of the resulting Fe-Ni alloy [11].

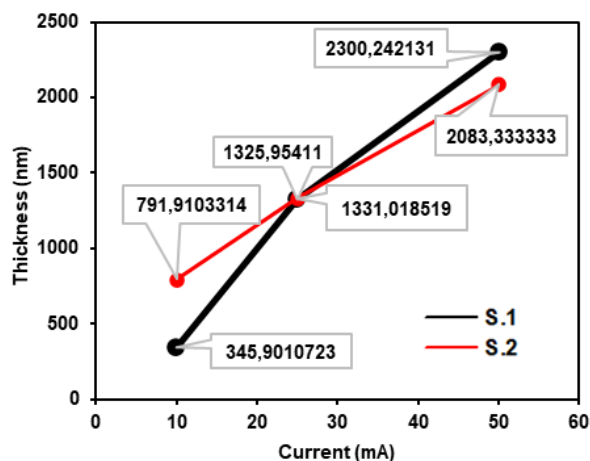


Figure 1. The curve of the relationship between the electrical current and the thickness of the Fe-Ni Alloy produced through the electrodeposition, solution 1 (black) solution 2 (red)

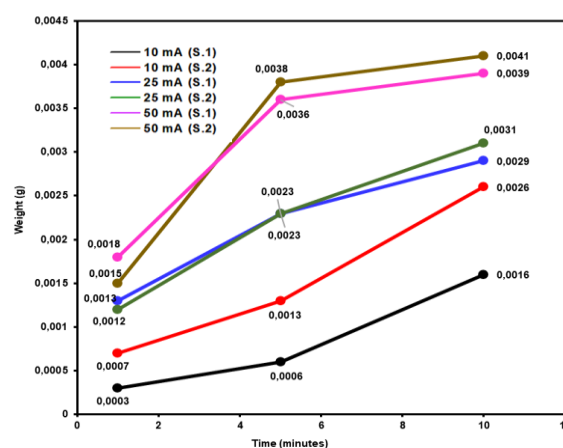


Figure 2. The curve of the relationship between time and weight of Fe-Ni alloy produced through the electrodeposition process with currents of 10 mA, 25 mA, and 50 mA for 1, 5, and 10 minutes (S: solution)

Figure 2 shows the relationship between the effect of time variations on the weight of the Fe-Ni deposits. The weight of the Fe-Ni alloy raises as the time deposition increases. The increase in weight due to the discharge time of the metal cations in the second layer becomes shorter as the deposition time increases [11].

SEM micrographs in Figure 3 show that the morphology of the Fe-Ni alloy obtained from electrodeposition using a current of 10 mA is granular. This is by the results of previous work which also produced Fe-Ni alloy nanoparticles with granular morphology [12]. It can be seen that the Fe-Ni alloy particles are evenly dispersed on the substrate. This uniform dispersion causes the Fe-Ni alloy to have a reasonably high homogeneity. The figure also shows that the resulting Fe-Ni alloy particles have a relatively uniform size range between 100nm to 200nm. The micrograph obtained by transverse observation shows two parts with different colors, namely the black and grey

sections. The black part in the image is an ITO micrograph, while the grey part is a Fe-Ni alloy particle micrograph [13].

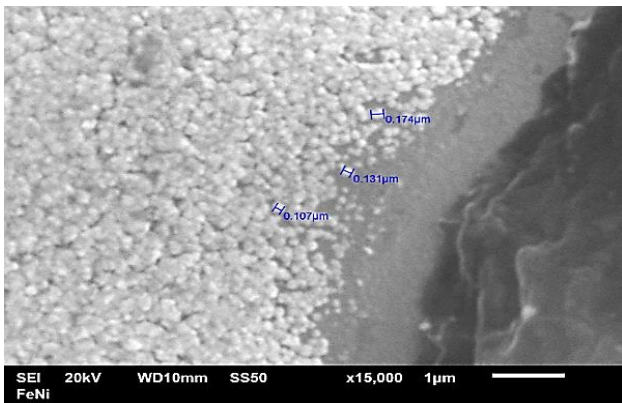


Figure 3. The morphology of the Fe-Ni alloy particles on the ITO substrate vertically

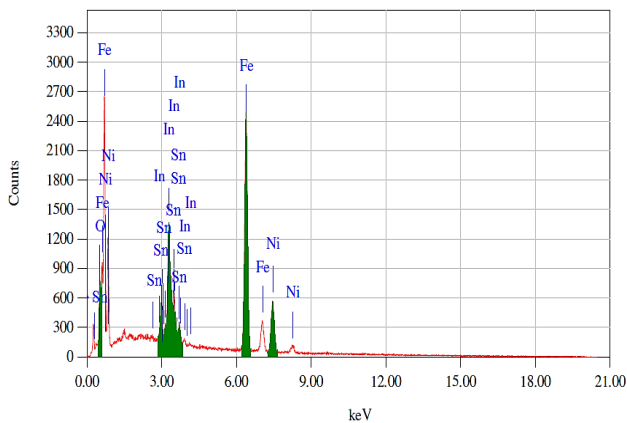


Figure 4. EDX spectrum of Fe-Ni alloy on ITO substrate

The EDX spectrum of the synthesized Fe-Ni alloy with a current of 10 mA for 1 minute is shown in Figure 4. The spectrum shows peaks of Fe and Ni that confirmed the formation of the Fe-Ni deposit on the ITO substrate with the atomic percentages are 77.63% (Fe) and 22.37% (Ni). Other peaks, such as Sn, In, and O, are obtained from the ITO substrate [14][15]. The percentage of Fe atoms is greater than the percent of Ni atoms. The atomic percent can change when the parameters of the current and the electrodeposition time used also change. This is due to the influence of chemical potential in the system on differences in the nanoparticle growth process [16].

Table 1. The concentration of Fe and Ni in solutions 1 and 2

Solution 1		Solution 2	
Fe (ppm)	Ni (ppm)	Fe (ppm)	Ni (ppm)
111.21	157.72	2780.2	23657.4

The ppm comparison between Fe and Ni in the two solutions of the AAS measurement is given in the following

Table 1. The ratio of the concentration of Fe and Ni metal ions in solution 1 is 2:3, while in solution 2, it is 1:9. The variation of the metals ion composition was carried out to observe the effect of the ratio of Fe and Ni concentrations in solution on the percentage of Fe and Ni atoms in the resulting deposit. The concentrations (ppm) of Fe and Ni metals in Fe-Ni alloys obtained by measurement using AAS were used to determine the percent of iron (Fe) and nickel (Ni) atoms in each deposit (Figure 5).

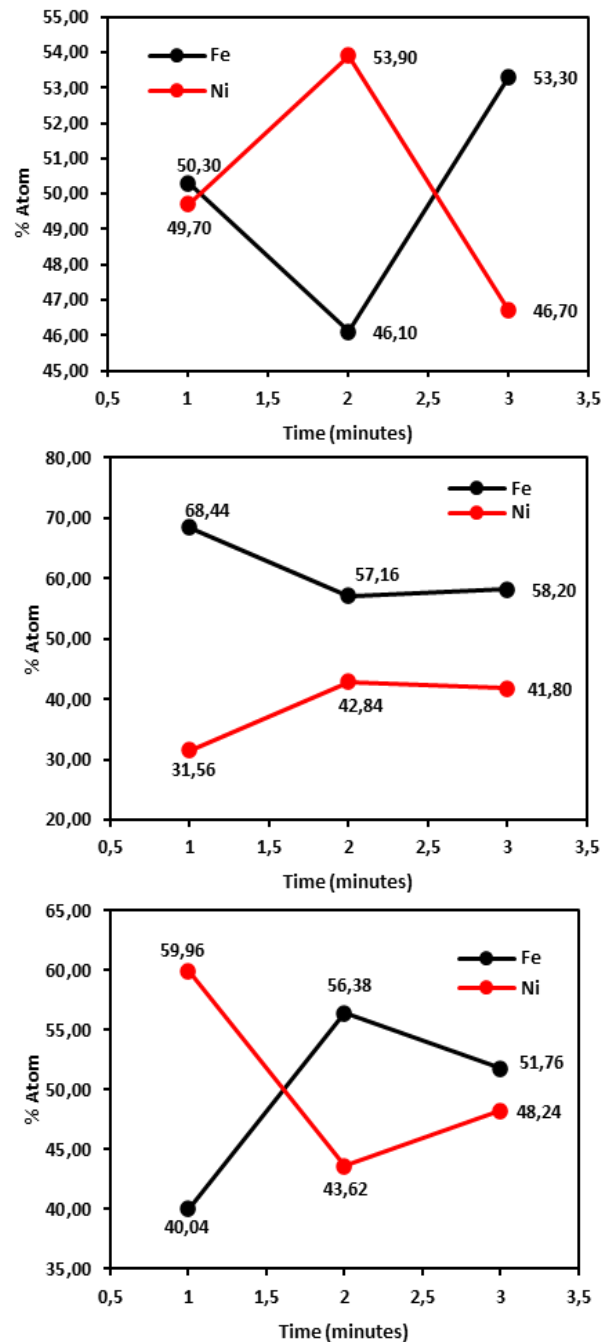


Figure 5. Comparison of Fe and Ni percentages in deposits at different applied currents (solution 1)

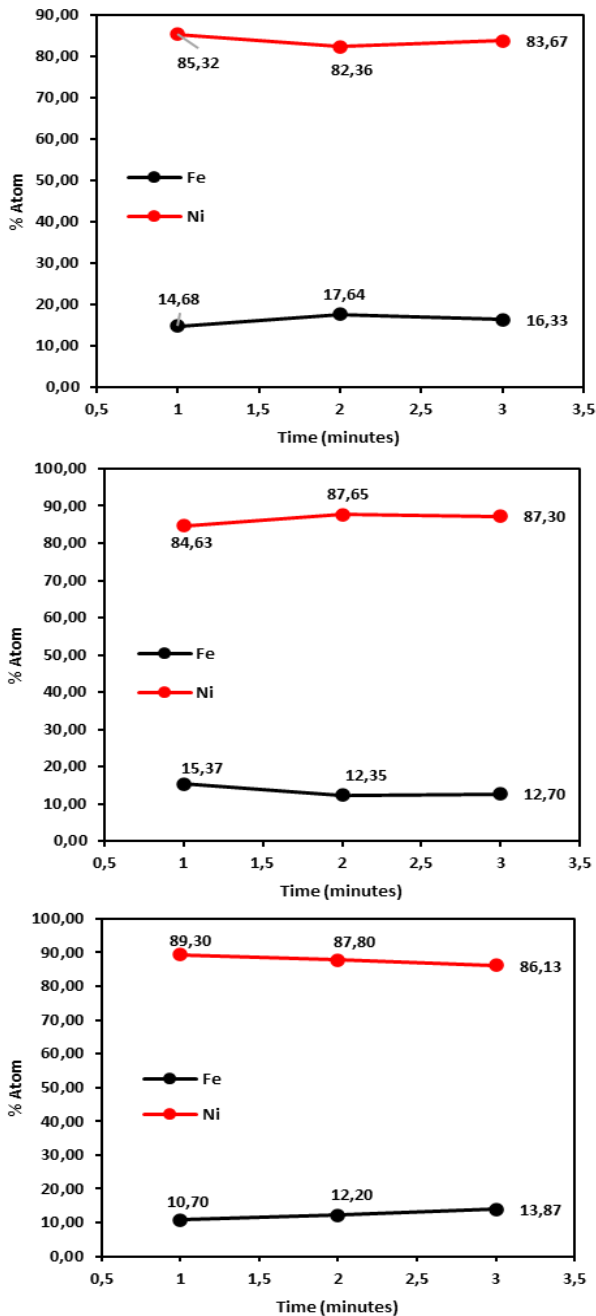


Figure 6. Comparison of Fe and Ni percentages in deposits at different applied currents (solution 2)

Electrodeposited Fe-Ni alloys using a current of 10 mA for 1 minute with solution 1 showed a higher Fe percentage than the currents of 25 mA and 50 mA. This is by the principle of electrodeposition of Fe group alloys, namely, increasing the current or temperature will increase the percent or amount of nobler metal in the deposit [12]. Another factor in Fe, which comes from the metal ion FeSO_4 , will burn when it is deposited using a high current [17]. Therefore, the highest percentage of Fe in the Fe-Ni alloy was obtained from the electrodeposition deposit with the lowest current. Meanwhile, the highest percentage of Ni in the Fe-Ni alloy was obtained from a deposit with the highest current of 50 mA.

A longer electrodeposition time causes an increase in the percentage of Fe in the deposit from solution 1. This is because the electrodeposition process in Fe-Ni alloys includes anomalous co-deposition, where nickel metal (Ni) which is nobler than iron (Fe), is deposited less in the deposit. [6]. During the electrodeposition process, iron hydroxide ions are formed on the surface of the substrate, thus inhibiting the nickel reduction process on the substrate. The reaction mechanism that occurs is as shown in Equation 1-8 [5].

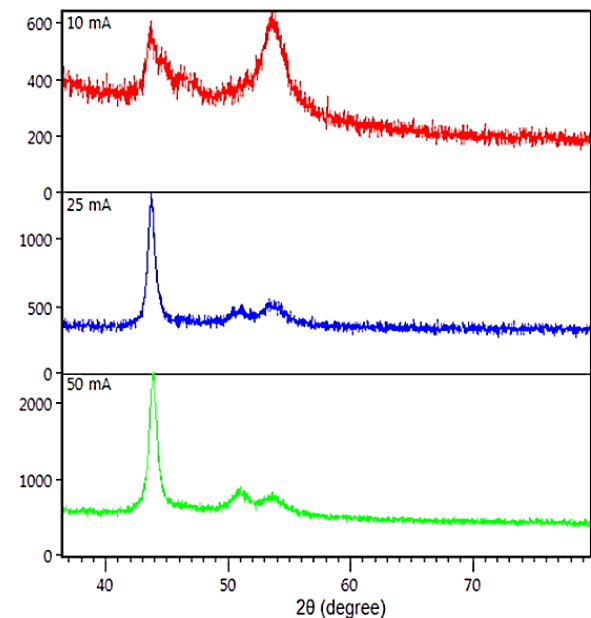
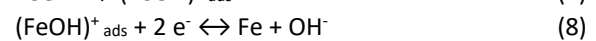
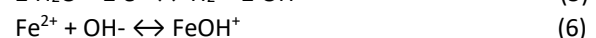
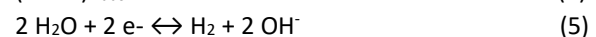
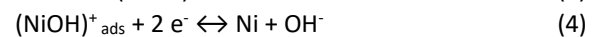
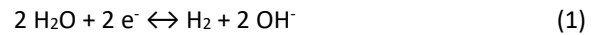


Figure 7. Diffraction Pattern of Fe-Ni alloy with current strength of 10, 25, 50 ma for 5 minutes on ITO substrate (solution 1)

The diffraction patterns of the Fe-Ni alloys electrodeposited using solution 1 with varying current show peaks at 2θ 44° and 53° at Figure 7, indicating a FCC of the structure formed over the substrate. From the XRD pattern analysis, the lattice parameter (a) of the two peaks is about 0.35 nm. The 2θ peak around 53° indicates the presence of an FCC crystal structure with a (200) plane. The highest intensity of the peak with the crystal structure of FCC (200) belongs to the Fe-Ni alloy synthesized with the applied current of 50 mA with an average particle size of 7.25 nm. This is because, at a current of 50 mA, there is a change in the resulting Fe-Ni alloy crystal structure [12].

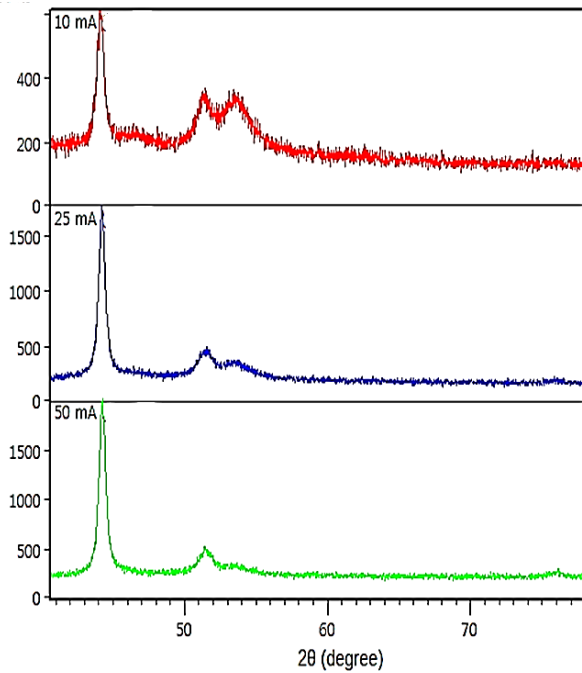


Figure 8. Diffraction pattern of Fe-Ni alloy with a current of 10, 25, and 50 mA for 5 minutes on ITO substrate (solution 2)

The diffraction patterns of the Fe-Ni alloy deposited using solution 2 with a variety of currents for 5 minutes are shown in Figure 8. The difference in the diffraction pattern between the Fe-Ni alloys from solution 1 and solution 2 is a 2θ peak in the 76° region, indicated by the deposit at a current of 50 mA. The lattice parameter (a) of the three peaks is also about 0.35 nm. This indicates that the peaks at 2θ at around 44° and 53° also indicate the crystal structure of FCC (111) and FCC (200), which are the main peaks of the crystal structure of Fe-Ni alloys with a higher percent Ni [18]. The highest intensity of the peak with the crystal structure of FCC (200) is also shown by the Fe-Ni alloy with a current of 50 mA with an average particle size of 9.54 nm.

The 2θ peak at 76° indicates the presence of a crystal structure of FCC (220) which is a change in orientation from the crystal structure of FCC (111). The main factors that cause the change in the crystal structure orientation are the increase in current and the high percentage of Ni in the deposit [12]. Therefore, the peak at 2θ around 76° was only found in the diffraction pattern of the Fe-Ni alloy with a current of 50 mA and in solution 2, which had a very high percentage of Ni atoms.

The observation of the effect of current on the crystallite size conducted previously [10] showed that the crystallite size decreased with the increase in the current. The size of the Fe-Ni alloy crystallites from solution 2 did not have a significant difference. This is due to the effect of the percentage of Fe on the crystallite size, where an increase in the percentage of Fe in the deposit reduce the crystallite size [19]. The effect of current and percentage

of Fe on the crystallite size of Fe-Ni alloy is seen in solution 1. At a current of 10 mA, the crystallite size obtained at 2θ around 44° is relatively low (16.32 nm) because it has a very high Fe content, while at 2θ around 53° , Fe content is lower so that the crystallite size increases [20]. At the same 2θ , the crystallite size with a current of 25 mA increased (22.27 nm) because the percentage of Fe in the Fe-Ni alloy was reduced. The crystallite size of the Fe-Ni alloy with a current of 50 mA decreased again due to an increase in the current and the percentage of Fe in the deposit.

4. Conclusions

In summary, the increase in electrodeposition current of Fe-Ni alloys increase the deposits thickness and Fe composition in the deposits. The ratio of Fe and Ni metal concentrations in solution 1 is only 2:3. In comparison, the effect of the ratio of the concentration of metals in solution on the composition of metals in the deposit is shown by the AAS data in solution 2, where the ratio of Fe and Ni is 1:9. An increase in the concentration of Ni in the solution led to an increase in the percent Ni. From the XRD analysis, it can be concluded that the FCC structures of Fe-Ni deposits were obtained from different electro-deposition currents and electrolyte compositions.

Acknowledgements

The authors thank Direktur Riset dan Pengabdian Masyarakat Kementerian Riset dan Teknologi Republik Indonesia for supporting this work with research grant No. 11/SP2H/DRPM/LP-UNJ/HF/III/2016.

References

- [1] Kefeni, K.K, *et al.* (2015). Synthesis and Characterization of Magnetic Nanoparticles and Study Their Removal Capacity of Trace Metals from Acid Mine Drainage. *Chemical Engineering Journal*, 276, 222-231.
- [2] Zadeh S.A, Raeissi K, Saidi A. (2009). Properties of Nanocrystalline Iron-Nickel Alloys Fabricated by Galvanostatic Electrodeposition. *Journal of Alloys and Compounds*, 485, 402-407.
- [3] Hanafi I, RazakDaud A, Radiman S. (2015). Electro-deposition of CoNiFe Alloy Thin Films in a Sulfate Solution. *Journal of Applied Science and Agriculture*, 10 (5), 58-62.
- [4] Sutrisno M G, Fauziyyah N, Paristiowati M. The Effects of Aniline Concentration and Deposition Time on Polyaniline Conductance as Substrate in Fe/Ni Electrodeposition. *The Center for Science Innovation*, 1, 12-17.

- [5] Nakano, H, *et al.* (2004). Mechanism of Anomalous Type Electrodeposition of Fe-Ni Alloys from Sulfate Solutions. *Materials Transactions*, 45 (11), 3130-3135
- [6] Ullal Y dan Hedge A.C. (2014). Electrodeposition and Electro-Catalytic Study of Nanocrystalline Ni-Fe Alloy. *International Journal of Hydrogen Energy*, 1-8.
- [7] Spain, Elain. (2011). High Sensitivity DNA Detection Using Gold Nanoparticles and Conducting Polymers. Ph.D. thesis. Dublin City University.
- [8] Tallman D.E, *et al.* (2002). Direct Electrodeposition of Polypyrrole on Aluminum and Aluminum Alloy by Electron Transfer Mediation. *Journal of The Electrochemical Society*, 149 (3), 173-179.
- [9] Frey A, *et al.* (2011). Analysis of Electrodeposited Nickel-Iron Alloy Film Composition Using Particle-Induced X-Ray Emission. Hope College.
- [10] Tripathi M. K dan Singh V.B. (2015). Properties of Electrodeposited Functional Ni-Fe/AlN Nanocomposite Coatings. *Arabian Journal of Chemistry*, Elsevier.
- [11] Deepthi K.A, *et al.* (2015). Physical and Electrical Characteristics of NiFe Thin Films Using Ultrasonic Assisted Pulse Electrodeposition. Elsevier.
- [12] Pavithra G.P dan Hedge A.C. (2012). Magnetic Property and Corrosion Resistance of Electrodeposited Nanocrystalline Iron-Nickel Alloys. *Applied Surface Science*, 258, 6884-6890.
- [13] Hanafi I, *et al.* (2013). Surfactant Assisted Electrodeposition of Nanostructured CoNiCu Alloys. *Journal of Physics*, 431.
- [14] Hanafi I, RazakDaud A, Radiman S. (2016). Potentiostatic Electrodeposition of Co-Ni-Fe Thin Films from Sulfate Medium. *Journal of Chemical Technology and Metallurgy*, 51 (5), 547-555.
- [15] Mah C.F, Yam F.K, Hassan Z. (2016). Investigation and Characterization of ZnO Nanostructures Synthesized by Electrochemical Deposition. *Procedia Chemistry*, 19, 83-90.
- [16] Budi S, *et al.* (2010). Effective electrodeposition of Co-Ni-Cu alloys nanoparticles in the presence of alkyl polyglucoside surfactant. *Applied Surface Science*, 257, 1027-1033.
- [17] Izaki M. (2010). Electrodeposition of Iron and Iron Alloys. *Modern Electroplating*, 5, John Wiley & Sons, Inc.
- [18] Jiraskova Y, *et al.* (2014), Phase and Magnetic Studies of the High-Energy Alloyed Ni-Fe. *Journal of Alloys and Compounds*, 594, 133-140.
- [19] Li Hongqi, Ebrahimi Fereshteh. (2003). An Investigation of Thermal Stability and Microhardness of Electrodeposited Nanocrystalline Nickel-21% Iron Alloys. *Acta Materialia*, 51, 3905-3913.
- [20] Li Hongqi, Ebrahimi Fereshteh. (2003). Synthesis and Characterization of Electrodeposited Nanocrystalline Nickel-Iron Alloys. *Material Science and Engineering*, A347, 93-101.