Microstructure And Resistivity Of Cu/Ni Thin Film Prepared By Magnetized Electroplating On Various Electrolyte Temperature

By M. TOIFUR

Microstructure And Resistivity Of Cu/Ni Thin Film Prepared By Magnetized Electroplating On Various Electrolyte Temperature

Jumratul Wustha, Moh. Toifur, Azmi Khusnani, Oki mustava

Abstract: Plating of Ni has been carried out on Cu assisted by a 200 gauss magnetic field at an electrolyte temperature variation of 30 °C-70 °C. The research aims to study the influence of the temperature of the electrolyte solution on the microstructure and resistivity. The results show that in the influence of the magnetic field, the increase in the electrolyte temperature strongly affects the microstructure of the Cu/Ni layer. Temperature of 60 °C is the best temperature to produce a layer that is not dull and does not burn. On the other hand, the increase in the electrolyte temperature can increase the intensity of Ni diffraction, d-spacing and grain size. The sheet resistivity is very dependent on the three microstructure parameters in accordance with the equation of R_s = 1.26×10-3 T +1.38 (Ω/sq).

Index Terms: Cu/Ni layer, microstructure, electrolyte temperature, diffraction peak intensity, d-spacing, grain size, sheet resistivity

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1. INTRODUCTION

low temperature sensors are very needed in various fields, especially in the medical field for cryonics techniques [1], [2]. One type of sensor that is being developed today is a temperature sensor based on RTD (Resistance Temperature Detector) based on combination of Cu and Ni [3], [4], [5], [6], which is made by electroplating method. Electroplating is used in various application [7]. It is comparable to other methods such as sputtering [8]. The material commonly used as a temperature sensor is platinum, copper and nickel [10]. Copper is more preferable than other by considering about the abondance and not expensive. However, the resistivity of copper is low, so need to be increased. Combination of Cu by Ni that has high resistivity is expected can solve this problem. Resistivity is an important parameter because it plays a role in determining sensor sensitivity. Many factors that determine the success of electroplating include current density, voltage, pH of the electrolyte solution, electrolyte temperature, deposition time, type of metal ions, electrolyte concentration, and stirring [11]. The temperature of the solution affects the electrolyte viscosity. At high temperatures the salt solution will decompose (as example NiCl → $Ni2^+$ + Cl_2-) so that the amount of Ni^{2+} ion increases and moves faster because the solution becomes runny. Both of these will accelerate the rate of reaction to form deposit at the cathode. The advantage is that it does not give the opportunity for the polarization of electrolyte molecules which will increase the voltage of process due to the occurrence of electrical double layer (EDL), which can avoid the formation of hydrogen gas which can prevent the attachment of deposits or form a layer that contains many bubbles so that the layers become brittle. Hydrogen gas arises when electroplating was carried out at low voltage. According to Kumar, the higher the temperature of the electrolyte solution, the greater the grain size [11]. The disadvantage is that the formation of these deposits makes the process of arranging atoms on the surface of the cathode becomes slow so that the layers tends to rough. In this study the synthesis of Cu/Ni layers was carried out by varying the temperature of the electrolyte from 30 °C – 70 °C. Plating is assisted by a magnetic field in the transverse direction with the aim of generating Lorentz forces on Ni2+ ions that lead to the cathode. It is hoped that this will reduce the hydrogen gas [12] because the deflection of the ion path is similar to stirring in an electrolyte solution so that it will also reduce the incidence of EDL. In addition, of course this deflection can increase the temperature of the solution, but according to Chiba's research, the rise in temperature does not reach 1 °C. The use of magnetic deposition can also smooth the grain size. Ganesh et al. have done the coating of nickel with a magnetic field forming a 45° angle to the cathode [12], [14]. As a result the grain size becomes smoother in the order of tens of angstroms, the effect of polarization decreases, so that the increase in stress during the deposition process does not occur. This is a solution to the problem of using the temperature of the solution which causes the grain size to be larger as revealed by Anand and Kumar [11]. It is expected that by varying the temperature the optimum temperature is obtained which results in a good microstructure and good electrical.

2 EXPERIMENT METHODS

In the electroplating process, electrolyte solution is prepared with the composition of H_3BO_3 (30 g), $NiSO_4$ (195 g), $NiCl_2$ (450 g) and H_2O (750 ml). The solution is heated to a temperature of 30 °C, then the equipment is assembled according to Figure 1. The coating process is carried out at a voltage of 1.5 volts for 60 seconds with a 4 cm electrode distance, and a magnetic field of 200 gauss. The coating process is repeated by varying the temperature of 30 °C, 40 °C, 50 °C, 60 °C, and 70 °C.

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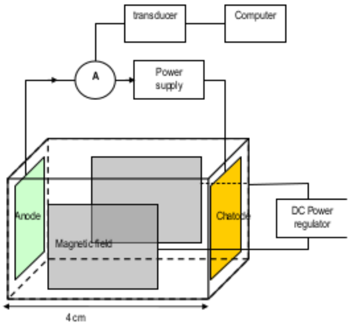


***Fig. 1**. Schematic experiment in the electroplating process of Cu/Ni.*

Some of the characterization done include SEM-EDS, XRD, and sheet resistivity. SEM tests are carried out to determine the formation of grains in Ni deposits and changes in size with changes in solution temperature. From the EDS spectrum we can know the Ni content in the layers as well as changes in Ni content to changes in electrolyte temperature XRD test to determine the formation of the crystalline phase on the deposit, as well as the effect of electrolyte temperature on the orderly level of the crystal structure, d-spacing, and grain size. All these parameters affect the resistivity of the layer.

Sheet resistivity is determined by a four-point probe by varying the current I and adding the final voltage V value [6]

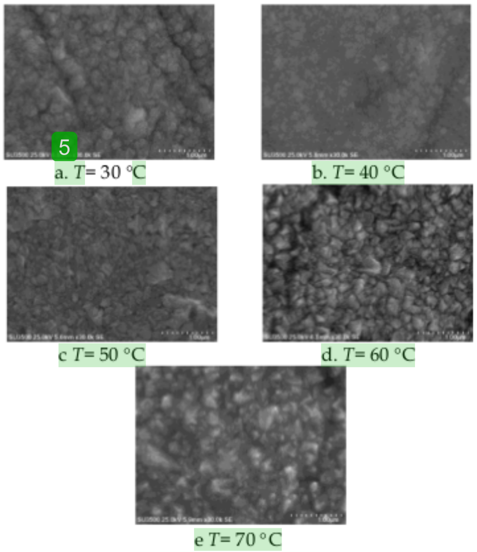
$$R_s = a\pi\left(\ln 2\right)^{-1} \qquad (1)$$

Where a is slope graph V-I.

3 RESULT AND DISCUSSION

3.1 Characterization of Cu/Ni using SEM-EDS

In Fig. 2 a SEM photo of the surface is displayed with a magnification of 30.000 times.



a. T= 30 °C b. T= 40 °C

c T= 50 °C d. T= 60 °C

e T= 70 °C

***Fig. 2**. SEM Photo of Cu/Ni specimen surface resulted from deposition at a electrolyte temperature of 30 °C-70 °C.*

From the figure it appears that the temperature of the solution affects the grain size. Some are dull, and some are clear. In the deposition specimens at 30 °C and 40 °C electrolyte temperatures the surface of the layer is dull colored then in line with the rise in color temperature electrolyte the surface layer becomes clearer, and the clearest color corresponds to temperatures up to 60 °C. Sadiku et al. [15] found that the brightest nickel color occurred at 56C electrolyte temperature, so what the researchers found was close to Sadiku's discovery. When the temperature of the solution is raised again to 70 °C the surface color of the layer becomes dull again. The dull color of the specimens resulting from deposition at temperatures of 30 °C and 40 °C is due to the temperature at which the solution has not been able to generate enough dense ion currents so that the layers appear dull [16], [17]. Usually, this kind of thing happens to plating using a weak electrolyte solution. But it is essentially the same, that is because of the minimal number of ions flowing into the cathode. Similarly, at a temperature of 70 °C the color of the Ni layer appears black because the solution temperature is too high so the surface of film is burned.

Next in Figure 3 an analysis of Ni content in the form of weight percent is shown on the rise in electrolyte temperature.

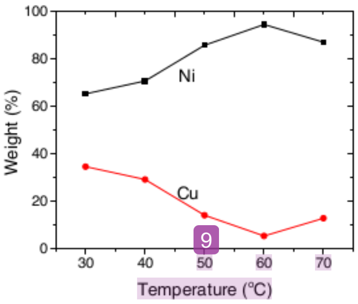


***Fig. 3**. Results of elemental analysis on Cu/Ni surfaces*

From figure 3, it is found that there is a tendency for the rising Ni content due to an increasing electrolyte temperature, which is 65.4% at 30 °C to 94.8% at 60 °C, except for 70 °C, the level decreases to 87.2% [18]. The increasing in Ni content indicates that an increase in electrolyte temperature can reduce the viscosity of the solution thereby increasing the ease of mass transport of Ni ions to the Cu cathode. As a result, the rate of formation of Ni deposits at the cathode becomes greater so that the levels increase. In contrast to the specimens resulted from deposition in 70 °C electrolyte, because the temperature is too high, the Ni layer is burned and less attached so that Ni levels become decrese. The other thing that happens to Cu, is that because the X-ray energy dispersed has a limited range, the increase in Ni levels will reduce the Cu level too. Therefore 60 °C is the best electrolyte temperature for Cu/Ni plating.

3.2 Study on XRD pattern

From the diffraction pattern for all specimens as presented in Figure 4, it is known that the Cu/Ni layer is in the form of crystals with two dominant peaks (preferred orientation) that occur at around angles 2-theta 43° and 44°. This peak corresponds to the Cu (111) and Ni (111) phases. This is consistent with the results that was done by Ibrahim. He investigate that black nickel is a pure nickel in the direction (111) [19]. Here, the source of X-rays comes from Cu-Kα radiation, with a wavelength λ = 1.5419 Å.

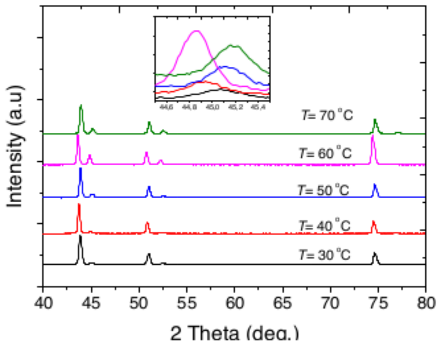


***Fig.** 4. X-ray diffraction for sample Cu/Ni films in various electrolyte temperature.*

From the graph subplot it is seen that the angle of peak diffraction of Ni has shifted for all specimens compared to the position of the peak angle of the diffraction of the previous specimen According to the Baragg diffraction formula, the diffraction peak shift affects the distance between the crystal planes (d-spacing). Besides that the peak movement together with FWHM according to the Scherrer formula will affect the size of the crystal grains. In addition, the intensity of the Ni diffraction peak also changes due to changes in electrolyte temperature.

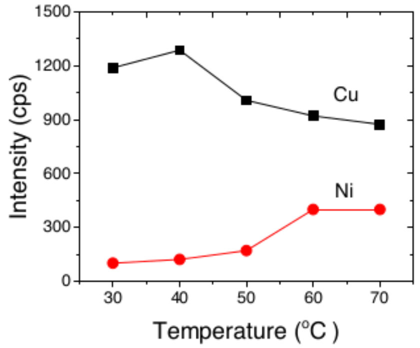


***Fig.** 5. The peak intensity of diffraction from Cu and Ni crystals in the various of the electrolyte temperature.*

Fig. 5 shows the diffraction peak intensity of Cu and Ni as a function of electrolyte temperature. From the picture it appears that the increase in the temperature of the solution can make the level of regularity of the Ni crystal structure increase. This is contrary to the intensity of Cu crystals which decreases with increasing temperature of the solution. Because the surface of Cu is a substrate for Ni, then with the condition of the formation of Cu crystals such as this that the level of regularity of the crystal structure decreases, the growth of Ni atoms does not depend on Cu substrate. The level of regularity if related to the electrical properties of the material, the resistivity of the layer tends to decrease. Fig. 6 shows the d-spacing at the electrolyte temperature variation. From the figure it appears that the d-spacing is influenced by electrolyte temperature. The relationship between d-spacing and the diffraction angle shift is the right shifting will reduce d-spacing while the left shifting will add d-spacing. The consequence of this is that if in this direction the electric current is flowed then the size of the d-spacing will influence the electrical conductivity (inverse of the electrical resistivity). For specimens that have small d-spacing in the current is flowed in direction (111), the layer is more conductive than samples that have large d-spacing. But the total conductivity of the specimen does not only depend on d-spacing.

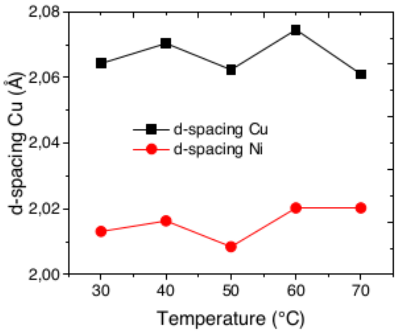


***Fig.** 6. Interplanar spacing of Ni and Cu in various temperature electrolyte.*

Next in Fig. 7 the grain size of Cu and Ni deposits varies with electrolyte temperature. In general there is a tendency as expressed by Anand [11] that an increase in electrolyte temperature will make the grain size bigger. This is due to the increase in temperature which will further reduce the the rising of H_2 gas bubbles.

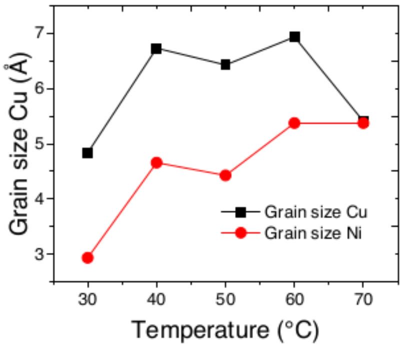


***Fig.** 7. Grain size of Cu and Ni*

At 30 °C the grain size of Ni 2.01Å is the smallest among the others and at the electrolyte temperature 70 °C grain size increased to 2.02Å. The increase is little, that is 0.01 Å but this is a contributor to the electrical properties of the coating which will increase the conductivity of the material.

3.3 Sheet Resistivity of Cu/Ni

The effect of electrolyte temperature on sheet resistivity of Ni and Cu at various electrolyte temperatures is shown in Fig. 8.

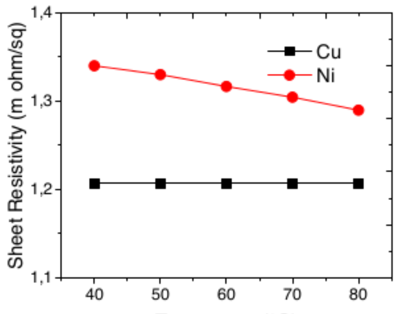


***Fig. 8**. Relationship bertween sheet resistivity of Cu and Ni to electrolyte temperature.*

Based on the data above it is known that the resistivity of the Cu layer increases when coated with Ni, where previously the resistivity of the Cu layer was 1.21×10-3 Ω/sq. As said by Afsarimanesch [20] if Cu is combined with Ni, the resistivity increases. Furthermore, with the increase in electrolyte temperature the thickness of the Ni layer increases, but as is well known, in metals if the thickness of the layer increases the resistivity decreases, but the decreasing resistivity not to be lower than the Cu sheet resistivity. The equation that states the relationship between Cu/Ni sheet resisitivity and electrolyte temperature:

$$R_s = 1.26\times 10\text{-}3\ T + 1.38 \quad (\Omega sq) \qquad (2)$$

with the determination index $R^2 = 0.99$. The sheet resistivity contributed by the regularity of the Cu/Ni crystal structure, d-spacing, and grain size. With the tendency of the peak intensity of Ni diffraction to in crease with increasing temperature of the electrolyte solution, interplanar distances which are increasing, and grain size that is getting bigger, those will make R_s getting decrease. This happen has been confirmed according to the profile of the R_s value which decreases with increasing the temperature of the solution. The dependence of the sheet resistivity on the electrolyte temperature is strong. This is indicated by the determination index that close to 1.

4 CONCLUSION

The following conclusions are a resume of the results of the research and discussion described above. The various electrolyte temperature can affects the microstructure and resistivity of Cu/Ni. The 60 °C is the best temperature to produce a layer that looks does not dull and does not burn. An increase in electrolyte temperature can increase the regularity of the crystal structure, the interplanar distance and grain size. All components the contribution to the reduction in sheet resistivity. In addition, sheet resistivity has a strong negative linear relationship to the electrolyte temperature.

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REFERENCES

[1] D. Miguel-Soto, D. Leandro, A. Lopez-Aldaba, J. Beato-López, J. Pérez-Landazábal, J.L. Auguste, R. Jamier, P. Roy, and M. Lopez-Amo, “Study of Optical Fiber Sensors for Cryogenic Temperature Measurements”, Sensors, 17(12), pp.2773, 2017.
[2] S. Kar and R.G., Sharma, “Cryogenic Temperature Sensors”, DEF SCI J, Vol. 57, No.3, pp. 195-208, 2007.
[3] E.I. Ionete, S.M. Iordache, A.M. Iordache, R.E. Ionete, A. Salceanu, C. Nichita, B. Dobrica, and I. Stamatin, “Cryogenic Sensor With Carbon Nanotubes”, Digest Journal of Nanomaterials & Biostructures (DJNB), 9(2), 2014.
[4] M. D. P. Khant, S. T. Shah, M.E, “Selection of sensor for Cryogenic Temperature Measurement”, IJSRD - International Journal for Scientific Research & Development, Vol. 1, Issue 2, ISSN (online): pp.2321-0613, 2013.
[5] M. Toifur, Y. Yuningsih, and A. Khusnani, “Microstructure, Thickness and Sheet Resistivity Of Cu/Ni Thin Film Produced By Electroplating Technique on The Variation of Electrolyte Temperature”. In Journal of Physics: Conference Series, IOP Publishing, Vol. 997, No. 1, Pp. 012053, 2018.
[6] R.G. Delatorre, M.L. Sartorelli, A.Q. Schervenski, A.A. Pasa, and S. Güths, “Thermoelectric Properties of Electrodeposited CuNi Alloys on Si”, Journal of applied physics, 93(10), pp.6154-6158, 2003.
[7] M. Sajjadnejad, H. Omidvar, M. Javanbakht, and A. Mozafari, “Characterization of Pure Nickel Coatings Fabricated under Pulse Current Conditions”, International Journal of Chemical, Molecular, Nuclear, Materials and Metallurgical Engineering, Vol:9, No:8, pp.1061-1065, 2015.
[8] A.M El-Sherik and U. Erb, “Synthesis of bulk nanocrystalline nickel by pulsed Electrodeposition”, Journal of Materials Science, 30, pp.5743-5749, 1995.
[9] H. Mu, J. Seok, and R.Y. Lin, “Nickel Thin Film Coatings on Steels With Electroless Plating and Sputter Deposition”. Journal of The Electrochemical Society, 150(2), pp.C67-C72, 2003.
[10] E.J.P. Santos, I.B. Vasconcelos, “RTD-Based Smart Temperature Sensor: Process Development and Circuit Design”, Proceedings in 2008 26th International Conference on Microelectronics, Vol. 1 and 2. pp. 333-336, 2008
[11] A. Anand, A. Aggarwal, J. Kumar, “Resistance Temperature Detector (RTD)”, International Journal of Innovative Research In Technology, Vol.1, Issue 12, pp.15-19, 2015.
[12] A. Chiba, K. Kitamura, and T. Ogawa, “Magnetic Field Effects on The Electrodeposition of Nickel From A High pH Watt’s Bath”. Surface and Coatings Technology, 27(1), pp. 83–88, 1986.
[13] V. Ganesh, D. Vijayaraghavan and V. Lakshminarayanan, “Fine Grain Growth Of Nickel Electrodeposit: Effect Of Applied Magnetic Field During Deposition”, Applied Surface Science, 240(1-4), pp. 286–295, 2005
[14] S. Kumar, S. Pande, and P. Verma, “Factor Effecting

Electro-Deposition Process", International Journal of Current Engineering and Technology, 5(2), pp.700-703, 2015.

[15] O. Sadiku-Agboola, E.R. Sadiku, O.I. Ojo, O.L Akanji, and O.F. Biotidara, "Influence of Operation Parameters on Metal Deposition in Bright Nickel-Plating Process", Portugaliae Electrochimica Acta, 29(2), pp.91-100, 2011.

[16] A.O. Gezerman, and B.D. Corbacioglu, "Analysis of The Characteristics of Nickel-Plating Baths", International Journal of Chemistry, 2(2), p.124, 2010.

[17] W. Giurlani, G. Zangari, F. Gambinossi, M. Passaponti, E. Salvietti, F. Di Benedetto, S. Caporali, and M. Innocenti, "Electroplating for Decorative Applications: Recent Trends In Research and Development". Coatings, 8(8), pp.260, 2018.

[18] A.O. Gezerman, and B.D. Corbacioglu, "Analysis of The Characteristics of Nickel-Plating Baths", International Journal of Chemistry, 2(2), pp.124, 2010.

[19] M.A. Ibrahim, "Black Nickel Electrodeposition from a Modified Watts Bath", Journal of Applied Electrochemistry, 36(3), pp.295-301, 2006.

[20] Afsarimanesh, Nasrin and A.P. Zaheer, "LabVIEW Based Characterization and Optimization of Thermal Sensors", International Journal on Smart Sensing and Intelligent Systems, Vol. 4 No. 4, ISSN: 1178-5608. pmp. 726-739, 2011.

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