Advanced Ceramics Progress: Vol. 7, No. 1, (Winter 2021) 46-51

Advanced Ceramics Progress

Research Article

Journal Homepage: www.acerp.ir

Heat Treatment of Pulsed Electroplated Nickel Deposited on AA2024 Aluminum

A. Khiabani [a], Z. S. Seyedraoufi [a]*, M. Heydarzadeh Sohi [b]

[a] *Department of Metallurgy and Materials Engineering, Karaj Branch, Islamic Azad University, Karaj, Alborz, Iran*
[b] *School of Metallurgy and Materials, College of Engineering, University of Tehran, Tehran, Tehran, Iran*

ARTICLE INFO

Article History:

Received 14 May 2021
Received in revised form 16 June 2021
Accepted 29 June 2021

Keywords:

AA2024 A luminum
Pulse Electroplating
Heat Treatment
Al_3Ni Intermetallic Compound
Corrosion

ABSTRACT

In this study, pure nickel was deposited on AA2024 aluminum via pulsed electrodeposition using two duty cycles (25 % and 50 %) at two different frequencies (50 and 100 Hz). The coated specimens were then heat-treated in an argon atmosphere at 500 °C for one hour. The treated specimens were characterized using Optical Microscope (OM), Scanning Electron Microscope (SEM) equipped with energy dispersive spectrometer (EDS), and X-Ray Diffraction (XRD) analysis. According to the results, the coatings were thickened upon increasing the duty cycle and frequency. Heat treatment of the nickel-coated specimen under the duty cycle of 25 % and frequency of 50 Hz experienced formation of Al_3Ni intermetallic compound at the interface of the nickel coating and aluminum-based substrate. Heat treatment adversely affected the corrosion resistance of the nickel-coated specimens. Nevertheless, the specimens with Al_3Ni intermetallic compound at their interface demonstrated better corrosion resistance among heat-treated materials.

https://doi.org/10.30501/ACP.2021.252211.1048

1. INTRODUCTION

Aluminum alloys have a wide range of applications, especially in automotive and aerospace industries, due to their superior properties such as high strength-to-weight ratio, corrosion resistance, and electrical and thermal conductivities [1]. However, these alloys suffer from poor tribological properties and inadequate corrosion resistance in some applications [2]. Different surface modifications and coating technologies have been assessed and applied on these alloys to overcome the aforementioned deficiencies.

Anodizing [3], electroplating [4], plasma nitriding [5], ion implantation [6], physical vapor deposition [7], diffusional coatings [8], thermal spray coatings [9], and liquid phase surface treatments [10] are among the different surface engineering processes that have been applied on these alloys. Electroplating is among the cheapest coating technologies. A number of metallic elements including cupper, chromium, and nickel have been electrodeposited on aluminum alloys using both direct and pulse currents so far [4,11]. In addition, a number of composite coatings have been deposited on these alloys through electroplating [4,12,13]. Diffusion coatings by means of electroplating followed by heat treatment have also been investigated by a number of researchers. These coatings might enjoy better adhesion to the substrate as well as higher hardness. Therefore, diffusional coatings could affect both corrosion and wear resistances of the treated materials [14-16].

Amadeh [17] and Lee [18] et al. studied the effects of heat treatment on electrodeposited nickel coating on 6061 aluminum alloy and reported the formation of Al_3Ni_2 and Al_3Ni, the intermetallic compounds on the treated layers, which could reduce corrosion resistance. Presd et al. also applied the above process on A352 aluminum and reported similar results [19]. There is little or no information on the application of pulse current for electrodeposition metallic elements prior to heat treatment in diffusional coatings. Therefore, in this study, nickel was pulse-electrodeposited on 2024 aluminum alloy before being heat treated. In addition, the effect of the duty cycle and frequency was evaluated.

* Corresponding Author Email: z.seyedraoufi@kiau.ac.ir (Z. S. Seyedraoufi)
https://www.acerp.ir/article_132766.html

Please cite this article as: Khiabani, A., Seyedraoufi, Z. S., Heydarzadeh Sohi, M., "Heat Treatment of Pulsed Electroplated Nickel Deposited on AA2024 Aluminum", ***Advanced Ceramics Progress***, Vol. 7, No. 1, (2021), 46-51. https://doi.org/10.30501/ACP.2021.252211.1048

2. MATERIALS AND METHODS

In this study, 20×20×4 mm specimens were cut out from AA2024 aluminum alloy (with a composition containing 4.41 wt.% Cu, 1.52 wt.% Mg, 0.50 wt.% Mn, 0.09 wt.% Si, 0.02 wt.% Fe, 0.02 wt.% Cr, and 0.20 wt.% Zn), and they were used as the substrates. Figure 1 shows the microstructure of the received substrate. Based on the studies by Goli et al. and Ghorbanzadeh et al., the precipitates identified by the red circle appear to be copper-rich compounds [20,21].

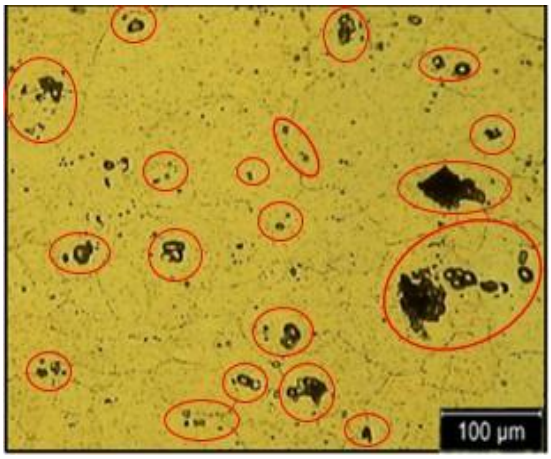

Figure 1. Optical micrograph of the AA2024 aluminum substrate

The specimens were then cleaned and polished with Emery paper up to 1200 grades. After washing the specimens with acetone, they were etched in a solution containing 50 g/L sodium hydroxide at 70 °C for 15 seconds and then washed in water. This step was repeated with a 65 % nitric acid solution for 5 seconds. At the final stage of the substrate preparation, the samples were immersed in zincate solution for 20 seconds (containing 2 g/L $FeCl_2$, 5 g/L $CuSO_4$, 40 g/L $KHC_4H_4O_6$, 10 g/L KCN, 106 g/L NaOH, 40 g/L $ZnSO_4$, and 30 g/L $Ni(SO_4)_2$).

Pulse electroplating was applied to nickel plating using Watts bath. The chemical composition of the bath is given in Table 1. A pure nickel sheet was used as anode, and the distance between AA2024 cathode and anode was fixed at 4 cm. The pH of the bath was 4 and its temperature was kept in the range of 45-55 °C. The current density was 2 A/dm^2 and the plating time was 15 minutes. Electroplating was performed at two pulse frequencies of 50 and 100 Hz and in two duty cycles of 25 and 50 %. After plating, the samples were heat-treated in a furnace under an argon gas atmosphere with a purity of 99.99 % at a temperature of 500 °C for 60 minutes. Table 2 shows the coding of the specimens according to their process conditions.

TABLE 1. Composition of Watts nickel plating bath

Brightener	50 g/L
Nickel sulfate ($NiSO_4·6H_2O$)	400 g/L
Boric acid (H_3BO_3)	50 g/L
Nickel chloride ($NiCl_2·6H_2O$)	100 g/L

TABLE 2. Specimen's coding based on process parameters

Sample's Code	Frequency (Hz)	Duty Cycle (%)	Heat treatment
P1	50	25	None
P2	100	50	None
PH1	50	25	Yes
PH2	100	50	Yes

The surface morphology and cross-sectional area of the electroplated and heat-treated specimens were examined by a VEGA/TESCAN Scanning Electron Microscope (SEM) equipped with Energy Dispersive Spectroscopy (EDS) analyzer. Phase analysis of the specimens was performed by PHILIPS PW1730 X-Ray Diffractometer (XRD) using a Cu K_α with a wave length of 1.54056 angstrom and a scanning speed of 0.05 degrees per second.

Polarization test was used to study the corrosion behavior of the coatings. Corrosion tests were performed in a 3.5 % NaCl solution. All potentials in this study were measured relative to Ag/AgCl. In all experiments, the solution temperature was 25 °C, and the sweeping speed was 1 mV/s. Before starting the test, the samples were immersed in the solutions for 1 hour.

3. RESULTS AND DISCUSSION

SEM images in Figure 2A-D show surface morphologies of the nickel-plated specimens before (P1 and P2) and after heat treatment (PH1 & PH2), respectively. Colonies of deposited nickel are observed in these images. In Figure 2C and D, the distances between the colonies are reduced, which may be due to the growth of the colonies. Heat treatment at high temperature and sufficient time results in a more or less proper diffusion and crystallites growth in the coating that leads to the agglomeration and enlargement of the colonies [22].

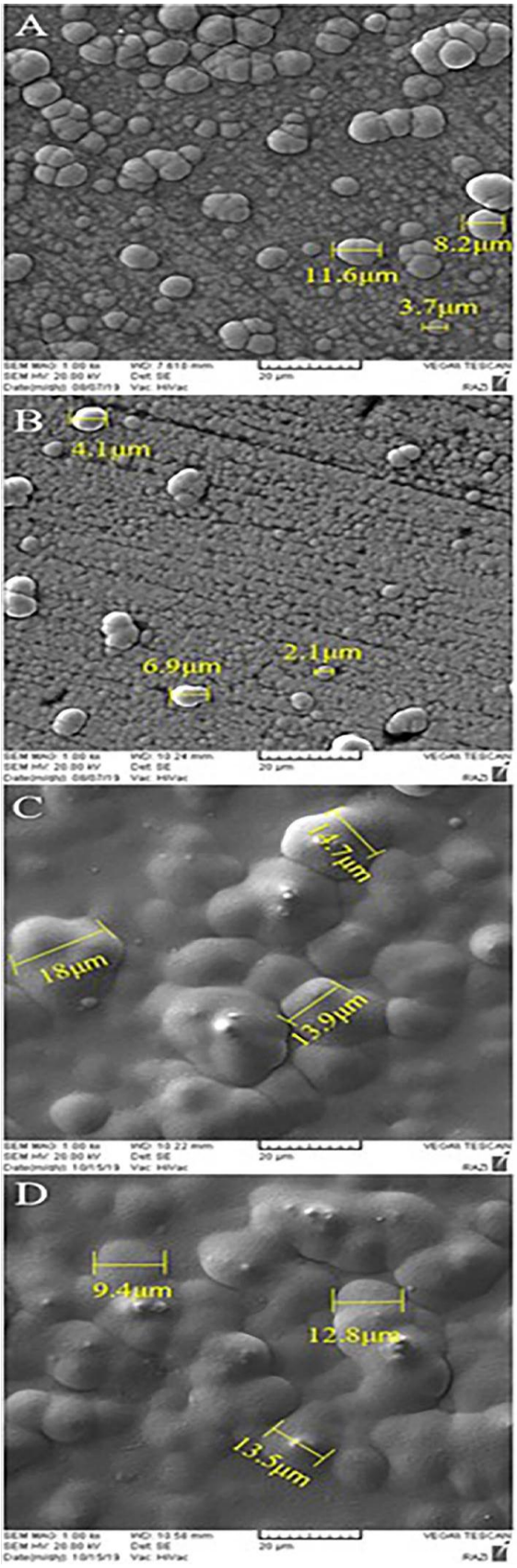

Figure 2. SEM images after electroplating and heat treatment: A) P1, B) P2, C) PH1, and D) PH2

Figure 3A-B shows SEM cross-sectional micrographs of the nickel electrodeposited specimens achieved in two different conditions of P1 and P2, indicating that the coating is thickened by increasing frequency and duty cycle. On the whole, as the duty cycle increases, the current on time increases and the off-time reduces. This situation leads to the formation of a thicker layer. On the other hand, increasing frequency results in a shorter pulse duration that leads to a thinner diffusion layer. Thus, transformation and diffusion of metal ions from the electrolyte to the surface of the cathode become easier. Therefore, increasing the pulse frequency can result in the thickening of the deposited layer [25]. The type of the frequency applied in pulse electrodeposition also affects the thermodynamics and kinetics of electrochemical reactions, consequently influencing the deposit's characteristics and properties such as wear and corrosion resistances [23-24].

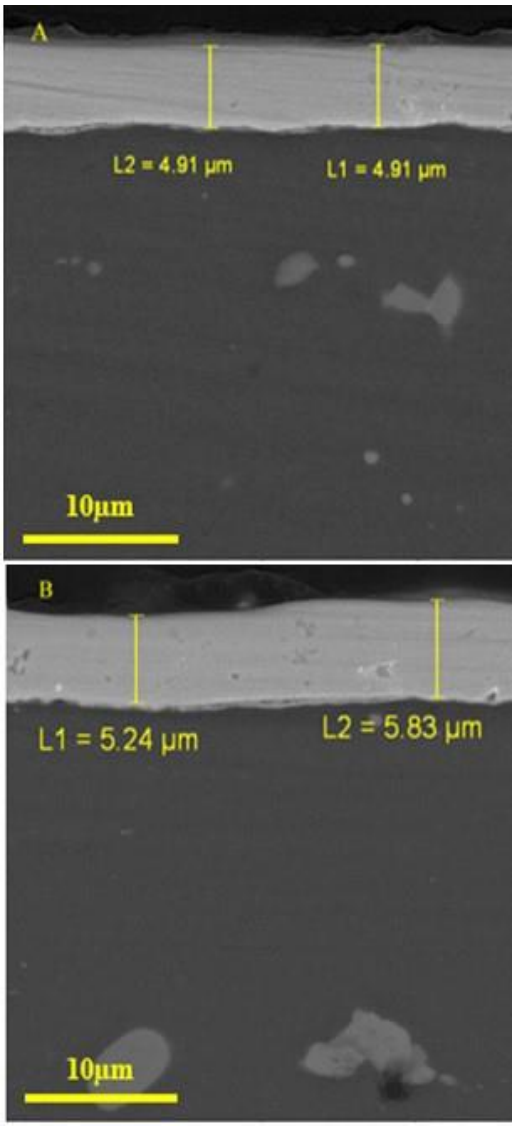

Figure 3. Cross-sectional SEM images of A) P1 and B) P2

Figure 4 illustrates the cross-sectional SEM images of the PH1, PH2 specimens and the result of the EDS analysis at the interface of the heat-treated PH2 specimen. As shown in Figure 4, a relatively thin layer has been formed at the interface between the coating and the substrate in the PH1 specimen, while this layer did not develop in the PH2 specimen. The reason for these structural differences is mainly related to the differences in the process parameters. By reducing the duty cycle, the sizes of the crystallites are reduced, and coating with a fine structure is formed on the substrate [17,27,28]. As the size of the crystallites decreases, the density of the grain boundary increases and, as a result, the paths prone to diffusion increase [17,27,28]. Consequently, diffusion facilitates and happens easier during heat treatment of the specimen which is electroplated in a lower duty cycle. As a result, a thin layer of about 300 nm consisting of intermetallic compounds was formed at the interface of PH1. The EDS results of this layer show that in addition to the nickel and aluminum substrates, magnesium from the substrate, zinc, and sulfur from the zincate layer and oxygen are also present in the EDS analysis. The high input of substrate elements in the EDS result results from the low thickness of the coating. Therefore, to accurately investigate this phase, the XRD test is required.

XRD patterns of the coated specimens before and after heat treatments are given in Figure 5. The main peak for nickel plated specimens (P1 & P2) in the range of 45 to 46 degrees appears to be a little bit broadened that could be an indication of some degrees of amorphism in the coating structure. Numerous studies have suggested that in electroplating and electroless plating, nickel coatings might be amorphous or semi-amorphous [29]. When the plating process is applied at more or less high speeds, the formation of coating in an amorphous manner is possible. Random placement of nickel atoms on the surface of the substrate deprives them of the opportunity to crystallize, and the atoms take a shorter order of domain [30,31]. Nevertheless, when the duty cycle increases, the intensity of the crystalline nickel peak in (111) increases. Similar results were reported by Borkar et al. [32].

After heat treatment, the amorphicity of the coating was entirely vanished and the coating turned to become crystalized. XRD pattern of the PH1 specimen indicates that the Al_3Ni intermetallic compound has been formed in the coating (Figure 5). This peak corresponds to the thin layer identified in the coating-sublayer interface in Figure 4-A. Since the coating was formed with a much smaller grain size in the lower duty cycles, the density of grain boundaries in this case was higher than that in the specimen coated with a higher duty cycle. The grain boundaries are highly suitable pathways for diffusion that is activated at high temperatures with sufficient time. Therefore, diffusion via grain boundaries leads to the formation of a thin Al_3Ni intermetallic layer.

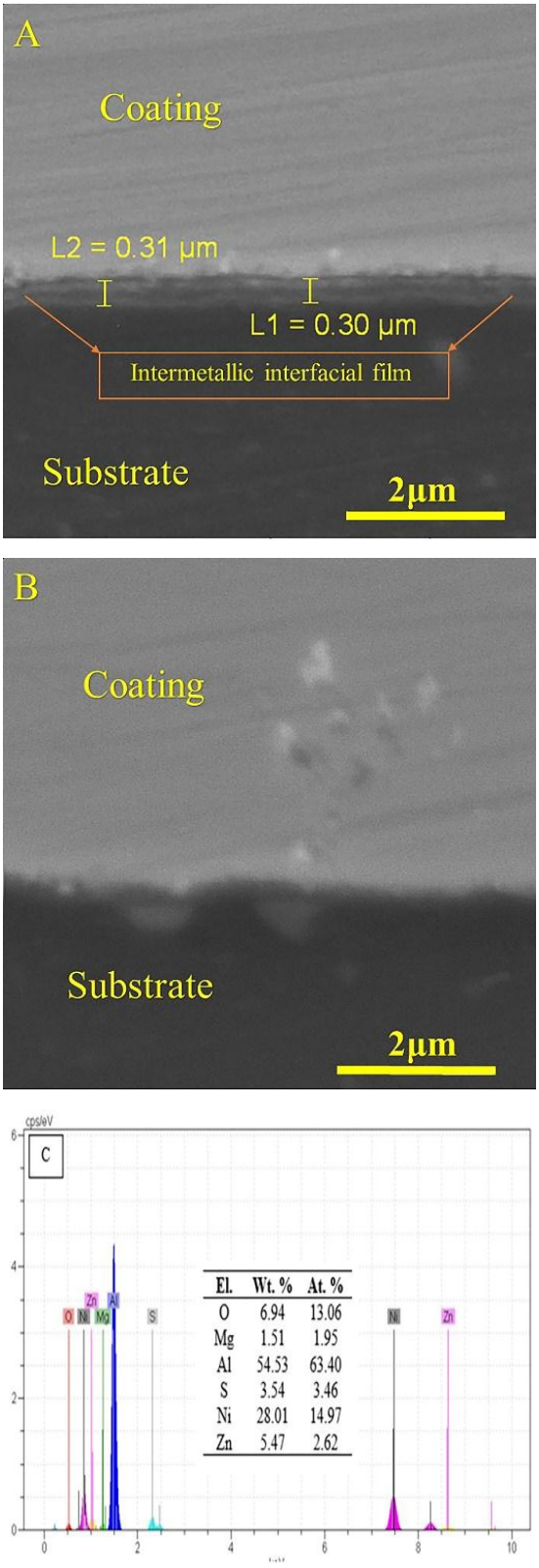

Figure 4. Cross-sectional SEM images of A) PH1, B) PH2, and C) EDS analysis of PH1 near the interface

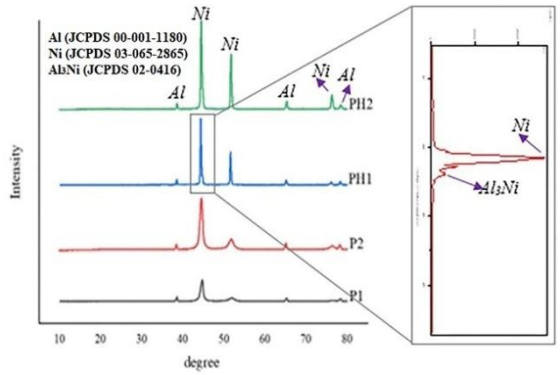

Figure 5. XRD patterns of the electrodeposited nickel before (P1 & P2) and after heat treatment PH1 & PH2)

Figure 6 shows the polarization curves achieved from corrosion tests of the specimens. These results show that the corrosion current density of the coating achieved at higher duty cycle and higher frequency is slightly higher than the one obtained in the lower duty cycle and frequency by around 10 %. In other words, corrosion resistance is slightly reduced when the duty cycle and frequency are doubled. These results also show that heat treatment adversely affects the corrosion resistance of the specimens. This might be related to the amorphicity of the electrodeposited nickel which disappears by heat treatment [33]. As shown in Figure 6, the PH1 specimen has higher corrosion resistance than the PH2 specimen. This might be due to the formation of Al_3Ni thin film that could create a new barrier against the corrosive agent. Previous researches have also shown that Al_3Ni intermetallic compound has high corrosion resistance [34].

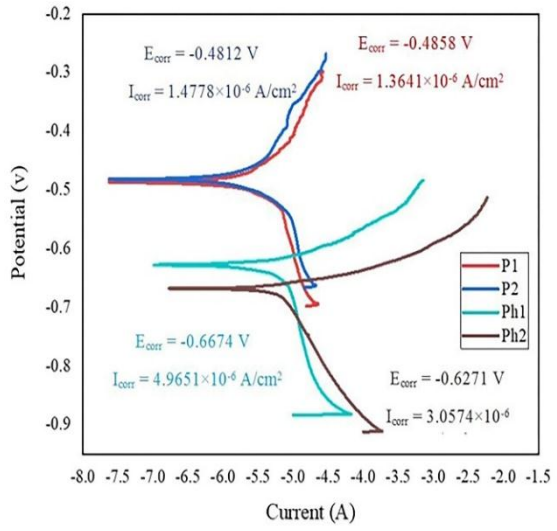

Figure 6. Polarization curves of the electrodeposited nickel before (P1 & P2) and after heat treatment PH1 & PH2)

4. CONCLUSIONS

1- Increase in the frequency and duty cycle resulted in the thickening of the nickel coating achieved from pulsed electrodeposition.
2- Heat treatment improved the crystallinity of the nickel coating.
3- Heat treatment of the nickel coated specimen achieved in the duty cycle of 25 % and frequency of 50 Hz resulted in the formation of thin Al_3Ni intermetallic layer at the interface of the coating and substrate.
4- After heat treatment, the corrosion resistance of the coating deteriorated. Among the heat-treated specimens, the one with the intermetallic compounds at its interface exhibited better corrosion resistance than the specimen without the intermetallic compounds.

5. ACKNOWLEDGMENTS

The authors thank Mr. Yazdan Shajari for his invaluable suggestions during experimental design.

REFERENCES

1. Davis, J. R., Ed., ***Aluminum and Aluminum Alloys, Alloying: Understanding the Basics***, ASM International, (2001), 351-416. https://doi.org/10.31399/asm.tb.aub.t61170351
2. Dorward, R. C., Pritchett, T. R., "Adanced aluminium alloys for aircraft and aerospace applications", ***Materials & Design***, Vol. 9, No. 2, (1988), 63-69. https://doi.org/10.1016/0261-3069(88)90076-3
3. Thompson, G. E., Habazaki, H., Shimizu, K., Sakairi, M., Skeldon, P., Zhou, X., Wood, G. C., "Anodizing of aluminium alloys", ***Aircraft Engineering and Aerospace Technology***, Vol. 71, No. 3, (1999), 228-238. https://doi.org/10.1108/00022669910270709
4. Rashidi, A. M., Amadeh, A., "Effect of Electroplating Parameters on Microstructure of Nanocrystalline Nickel Coatings", ***Journal of Materials Science & Technology***", Vol. 26, No. 1, (2010), 82-86. https://doi.org/10.1016/s1005-0302(10)60013-8
5. Naeem, M., Díaz-Guillén, J. C., Akram, M., Iqbal, J., Naz, M. Y., Shafiq, M., "Novel Active Screen Plasma Nitriding Of Aluminum Using Aluminum Cathodic CAGE", ***Surface Review and Letters***, Vol. 27, No. 09, (2020), 1950205. https://doi.org/10.1142/s0218625x19502056
6. Figueroa, R., Abreu, C. M., Cristóbal, M. J., Pena, G., "Effect of nitrogen and molybdenum ion implantation in the tribological behavior of AA7075 aluminum alloy", ***Wear***, Vol. 276, (2012), 53-60. https://doi.org/10.1016/j.wear.2011.12.005
7. Li, Y., Zhaob, J., Zenga, G., Guana, C., He, X., "Ni/Ni_3Al microlaminate composite produced by EB-PVD and themechanical properties", ***Materials Letters***, Vol. 58, No. 10, (2004), 1629–1633. https://doi.org/10.1016/j.matlet.2003.09.052
8. Rezaei, M., Jeshvaghani, R. A., Shahverdi, H. R., Mojaver, R., Torkamany, M. J., "Formation of Ni-rich aluminide layers on an A356 aluminum alloy by a combined electroplating/laser alloying treatment: Microstructure and tribological characteristics", ***Journal of Manufacturing Processes***, Vol. 29, (2017), 310-319. https://doi.org/10.1016/j.jmapro.2017.07.028

9. Naeimianm, H., Mofid, M. A., "TLP bonding of Ti−6Al−4V to Al 2024 using thermal spray Babbitt alloy interlayer", ***Transactions of Nonferrous Metals Society of China***, Vol. 30, No. 5, (2020), 1267-1276. https://doi.org/10.1016/s1003-6326(20)65294-3
10. Heydarzadeh Sohi, M., Ansari, M., Ghazizadeh, M., Zebardast, H., "Liquid phase surface nitriding of aluminium using TIG process", ***Surface Engineering***, Vol. 31, No. 8, (2015), 598-604. https://doi.org/10.1179/1743294414y.0000000372
11. Carvalho, A. L. M.,Voorwald, H. J. C., "Influence of shot peening and hard chromium electroplating on the fatigue strength of 7050-T7451 aluminum alloy", ***International Journal of Fatigue***, Vol. 29, No. 7, (2007), 1282-1291. https://doi.org/10.1016/j.ijfatigue.2006.10.003
12. Taye, D., Mohanty, S., Das, A. K., Singh, K. N., "Electroless Ni–Al_2O_3–WS_2 Composite Coating on Aluminum Substrate", ***Transactions of the Indian Institute of Metals***, Vol. 72, No. 9, (2019), 2281-2292. https://doi.org/10.1007/s12666-019-01677-1
13. He, L., Tan, Y., Wang, X., Xu, T., Hong, X., "Microstructure and wear properties of Al_2O_3-CeO_2/Ni-base alloy composite coatings on aluminum alloys by plasma spray", ***Applied Surface Science***, Vol. 314, (2014), 760-767. https://doi.org/10.1016/j.apsusc.2014.07.047
14. Hughes, A. E., Parvizi, R., Forsyth, M., "Microstructure and corrosion of AA2024", ***Corrosion Reviews***, Vol. 33, No. 1-2, (2015), 1-30. https://doi.org/10.1515/corrrev-2014-0039
15. Jung, J., Oak, J., Kim, Y. H., Cho, Y. J., Park, Y. H., "Wear behaviors of pure aluminum and extruded aluminum alloy (AA2024-T4) under variable vertical loads and linear speeds", ***Metals and Materials International***, Vol. 23, No. 6, (2017), 1097-1105. https://doi.org/10.1007/s12540-017-7158-y
16. Bahri, H., Danaee, I., Rashed, G. R., Dabiri, A. R., "Scratch and wear resistance of nano-silica-modified silicate conversion coating on aluminium", ***Materials Science and Technology***, Vol. 32, No. 13, (2016), 1346-1353. https://doi.org/10.1080/02670836.2015.1124192
17. Adabi, M., Amadeh, A. A., "Formation mechanisms of Ni–Al intermetallics during heat treatment of Ni coating on 6061 Al substrate", ***Transactions of Nonferrous Metals Society of China***, Vol. 25, No. 12, (2015) 3959-3966. https://doi.org/10.1016/s1003-6326(15)64073-0
18. Li, C., Yuan, Z., Guo, R., Xuan, W., Ren, Z., Zhong, Y., Li, X., Wang, H., Wang, Q., "Reaction diffusion in Ni–Al diffusion couples in steady magnetic fields", ***Journal of Alloys and Compounds***, Vol. 641, (2015), 7-13. https://doi.org/10.1016/j.jallcom.2015.04.061
19. Prasad, D. S., Ebenezer, N. S., Shoba, C., Raju, P., Rao, P. S., "The Effect of T6 Heat Treatment on Corrosion Behavior of Nickel Electroplated Metal Matrix Composites", ***Silicon***, Vol. 11, No. 4, (2019), 2025–2032. https://doi.org/10.1007/s12633-018-0023-x
20. Goli, F., Jamaati, R., "Effect of strain path during cold rolling on the microstructure, texture, and mechanical properties of AA2024 aluminum alloy", ***Materials Research Express***, Vol. 6, No. 6, (2019), 066514. https://doi.org/10.1088/2053-1591/ab0a1f
21. Ghorbanzade, T., Soltanipour, A., Dehghani, K., Chabok, A., "Microstructural evolutions and mechanical properties of friction stir welded AA2024-3", ***Proceedings of the Institution of Mechanical Engineers, Part L: Journal of Materials: Design and Applications***, Vol. 230, No. 1, (2014), 75-87. https://doi.org/10.1177/1464420714545369
22. Hanachi, M., Seyedraoufi, Z. S., Abouei, V., "Investigation of Microstructure, Hardness, and Corrosion Resistance of Ni-P-GO Electroless Nanocomposite Coating on AZ31D Alloy Surface", ***Advanced Ceramics Progress***, Vol. 6, No. 3, (2020), 55-62. https://doi.org/10.30501/ACP.2020.233518.1038
23. Lajevardi, S. A., Shahrabi, T., "Effects of pulse electrodeposition parameters on the properties of Ni–TiO_2 nanocomposite coatings", ***Applied Surface Science***, Vol. 256, No. 22, (2010), 6775-6781. https://doi.org/10.1016/j.apsusc.2010.04.088
24. Kumar, K. A, Kalaignan, G. P., Muralidharan, V. S., "Direct and pulse frequency electrodeposition of Ni–W–TiO_2 nanocomposite coatings", ***Ceramics Internatinal***, Vol. 39, No. 3, (2013), 2827-2834. https://doi.org/10.1016/j.ceramint.2012.09.054
25. Seyedraoufi, Z. S., Mirdamadi, S., "Effects of pulse electrodeposition parameters and alkali treatment on the properties of nano hydroxyapatite coating on porous Mg–Zn scaffold for bone tissue engineering application", ***Materials Chemistry and Physics***, Vol. 148, No. 3, (2014), 519-527. https://doi.org/10.1016/j.matchemphys.2014.06.067
26. Moradi, E. H., Jafarzadeh, K., Borji, S., Abbaszadeh, H., "Pulse electrodeposition as a new approach in electrowinning of high purity cobalt from WC–Co scraps. Part I: The effect of frequency and duty cycle", ***Minerals Engineering***, Vol. 77, (2015), 10-16. https://doi.org/10.1016/j.mineng.2015.02.012
27. Adabi, M., Amadeh, A. A., "Electrodeposition mechanism of Ni–Al composite coating", ***Transactions of Nonferrous Metals Society of China***, Vol. 24, No. 10, (2014), 3189-3195. https://doi.org/10.1016/s1003-6326(14)63459-2
28. Adabi, M., Amadeh, A., "Effect of electrodeposition conditions on properties of Ni–Al composite coatings", ***Surface Engineering***, Vol. 31, No. 9, (2015) 650-658. https://doi.org/10.1179/1743294414y.0000000449
29. Zhang, P., Lv, Z., Liu, X., Xie, G., Zhang, B., "Electroless nickel plating on alumina ceramic activated by metallic nickel as electrocatalyst for oxygen evolution reaction", ***Catalysis Communications***, Vol. 149, (2021), 106238. https://doi.org/10.1016/j.catcom.2020.106238
30. Li, H., He, Y., Fan, Y., Xu, W., Yang, Q., "Pulse electrodeposition and corrosion behavior of Ni–W/MWCNT nanocomposite coatings", ***RSC Advances***, Vol. 5, No. 84, (2015), 68890-68899. https://doi.org/10.1039/c5ra09462c
31. Atarchi, M., Sadrnezhad, S., "Pulse reverse electrodeposition of spherical Ni-MWCNT composite skein", ***International Journal of Engineering, Transactions B: Applications***, Vol. 22, No. 2, (2009), 161-168. https://www.ije.ir/article_71780.html
32. Borkar, T., Harimkar, S. P., "Effect of electrodeposition conditions and reinforcement content on microstructure and tribological properties of nickel composite coatings", ***Surface and Coatings Technology***, Vol. 205, No. 17-18, (2011), 4124–4134. https://doi.org/10.1016/j.surfcoat.2011.02.057
33. Shajari, Y., Porhonar, M., Seyedraoufi, Z. S., Razavi, S. H., Baghdadabad, D. M., Yousefnia, H., Farahani, M., "Improvement of the NiBrAl Casting Alloy Surface Properties by Electroless Ni-B Plating for Dynamic Marine Applications", ***Physical Mesomechanics***, Vol. 23, No. 1, (2020), 81-88. https://doi.org/10.1134/s1029959920010087
34. Mohammed, A., Yaro, S. A., Abdulwahab, M., "Influence of Ni additions and age-hardening treatment on the corrosion resistance of Al-Cu-Si alloy", ***Journal of Materials and Environmental Science***, Vol. 7, No. 5, (2016), 1549-1555. https://www.jmaterenvironsci.com/Document/vol7/vol7_N5/170-JMES-948-Mohammed.pdf