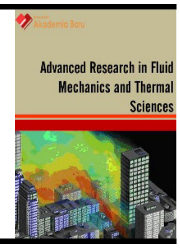
Penerbit Akademia Baru

Journal of Advanced Research in Fluid Mechanics and Thermal Sciences

Journal homepage: www.akademiabaru.com/arfmts.html

ISSN: 2289-7879



Open Access

Effect of Current Density and Fly Ash Composition on Nickel Grain Size and Hardness of Nickel-Fly Ash Composite Coating Deposited on Aluminum Alloy 6061

Intan Sharhida Othman[1,*], Muhamad Khaizaki Ahmad[1], Muhammad Zaimi Zainal Abidin[1], Qumrul Ahsan[1], Noraiham Mohamad[1], Syahrul Azwan Sundi[2]

[1] Faculty of Manufacturing Engineering, Universiti Teknikal Malaysia Melaka, Hang Tuah Jaya, 76100 Durian Tunggal, Melaka, Malaysia
[2] Faculty of Engineering Technology, Universiti Teknikal Malaysia Melaka, Hang Tuah Jaya, 76100 Durian Tunggal, Melaka, Malaysia

ARTICLE INFO

Article history:
Received 3 May 2018
Received in revised form 19 August 2018
Accepted 22 September 2018
Available online 26 September 2018

Keywords:
Current Density, fly ash composition, grain size, Ni-FA composite coating

ABSTRACT

The nickel- fly ash (Ni-FA) composite coatings were deposited on zincated aluminium alloy 6061 (AA6061) substrate by using electrodeposition technique. The electrodeposition process was carried out for 1 hour at 40º C. The current density used was extended from 1, 5 to 9 A/dm2. Fly ash particles was added to nickel citrate bath at 0, 10, 50 and 90 g/l for electrodeposition of Ni-FA composite coating. The produced composite coatings were characterized using X-ray Fluorescence (XRF) and X-ray Diffraction (XRD). Microhardness of Ni-FA composite coatings were also investigated in this study. As the current density increased from 5 to 9 A/dm2. The nickel grain size in the Ni-FA composite coating were decreased. The decreasing in nickel grain size resulted in increasing of hardness value. Besides, the hardness results show a clear increasing trend with an increase of fly ash composition from 10 to 90 g/l, due to the presence of hard oxide in the fly ash particles.

1. Introduction

Aluminium alloy 6061 (AA6061) is the most versatile of the heat treatable alloy which is commonly used for medium to high strength requirements and has good toughness characteristics. Based on Leon [1], the aluminium alloy 6061 has excellent strength to weight ratio, corrosion resistance, good ductility and cracking resistance in adverse environment. These AA6061 is extensively employed in marine frames, pipelines, storage tanks, automotive parts and aircraft application [2]. However, according to Bocking and Reynolds [3] and Visser [4], it has low hardness and wears resistance, as well as susceptible to surface degradation when exposed to elevated temperatures.

Therefore, nickel composite coating is introduced to AA6061 to improve its properties. According to Nguyen *et al.*, [5], the composite coating is a helpful and minimal effort approach to

* *Corresponding author.*
E-mail address: intan_sharhida@utem.edu.my (Intan Sharhida Othman)

enhance the substrates' properties. Based on Boonyongmaneerat *et al.*, [6] and Lins *et al.*, [7], the composite coatings produced from the electrodeposition process have excellent surface properties, such as high hardness, high abrasion, high corrosion resistance and low friction coefficient. Fly ash is residue derived from combustion of coal which widely available in worldwide and lead to waste management proposal [8]. The fly ash can be employed as inexpensive strengthening particles which can increase wear resistance and enhanced micro-hardness and have low density [9,10].

According to Rashidi and Amadeh [11], current density plays an important role to the grain size of the electrodeposited coatings. Grain size decrease due to higher nucleation rate associated with higher current density [12]. Increasing current density will increase the incorporation of particles content in the composite. The deposition of matrix metal with increasing current density is fast enough to entrap and occlude some of the particle and incorporate them into deposits [13].

However, the effect of current density and fly ash composition on nickel composite coating deposited on AA6061 substrate are still less known. Thus, the present work is aimed to investigate the influence of various current density and fly ash composition on the nickel grain size and hardness properties of Ni-FA composite coating.

2. Methodology

The aluminium alloy 6061 (AA6061) substrate with dimension 40 mm x 30 mm x 3 mm were mechanically ground using silicon carbide papers of 180, 600, 800 and 1200 grit. By using XRF technique, the aluminium alloy contains 98.884 wt % of Al, 0.498 wt % of Si and 0.319 wt % of Cu and the rest are shown in Table 1. The substrates were first cleaned with acetone, and then followed by immersion in 10 wt. % of sodium hydroxide (NaOH) solution for 10 seconds and immersion in 50 vol. % of nitric acid (HNO_3) for 20 seconds. The zincating process was carried out by dipping the pre- cleaned substrate vertically in a small glass beaker containing a zincating solution for 5 minutes at room temperature.

Table 1
Element of Aluminium alloy 6061, as found by XRF technique

Element	Concentration (wt. %)
Al	98.884
Si	0.498
Cu	0.319
Mn	0.132
Cr	0.131
S	0.017
Ta	0.008
Ni	0.006
P	0.002
Pb	0.002

The depositions of Ni-FA composite coating on zincated substrates were performed by applying various current density at 1, 5 and 9 A/dm^2. The chemical composition and operating condition of electrodeposition of Ni-FA composite coating on AA6061 substrate were summarized in Table 2. The electrodeposition process was carried out by using three electrode systems consisting of working electrode of AA6061, counter electrode of nickel plate and silver-silver chloride potassium chloride as a reference electrode. These three electrodes were connected to the Autolab Potentiostat/Galvanostat 302. The schematic diagram of electrodeposition set-up is shown in Figure 1.

Table 2
Composition of nickel citrate solution and electrodeposition operating condition

Composition	Concentration (g/l)
Nickel sulphate hexahydrate	200
Nickel chloride	20
Sodium citrate	30
Operating Condition	
Temperature (°C)	40
Deposition time (min)	60
Current density (A/dm^2)	1, 5, 9
Composition of fly ash (g/l)	0, 10, 50, 90

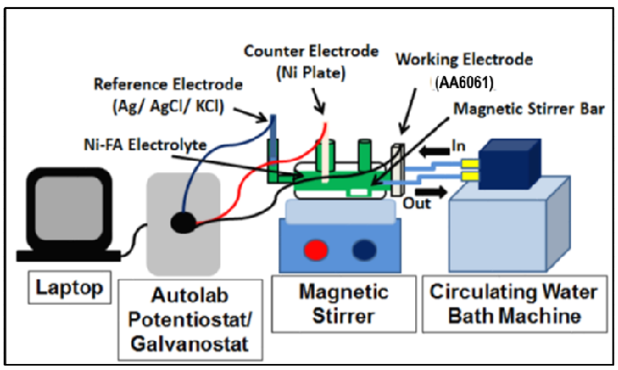


Fig. 1. Schematic diagrams of electrodeposition process

The grain size of nickel was evaluated using the Scherrer Equation (Equation 1) from XRD patterns. This might fail to give accurate result, but many comparison experiments showed that the XRD result was very close to TEM result [14,15].

$$\mathrm{d}p = \frac{k\lambda}{B \cos \theta} \tag{1}$$

Where k is a constant (0.94), λ is the wave number of the Cu Ka, 1.5405A, ϴ (rad) is the scattering angle and β is FWHM (rad) which is the width at the half way mark of the peak analyzed. The micro-hardness testing was performed using micro Vickers hardness tester at 0.5 kg/f load and 10 times indentations for 15s in each indentation.

3. Results and Discussion

3.1 Characterization of Fly Ash

The chemical composition for fly ash particles are found by using X-Ray Fluorescence (XRF) technique and shown in Table 3. The FA particles contain 44.1% of silicon dioxide (SiO_2), 19.1% of aluminium dioxide (Al_2O_3) and 12.4% of ferum trioxide (Fe_2O_3). According to ASTM C618 [16], the FA particles used is classified as Class F due to the calcium oxide (CaO) (12.4%) in the fly ash particles is less than 20%.

The morphology of fly ash particles was studied by Scanning Electron Microscope (SEM). It can be seen clearly from Figure 2 that the fly ash particles are in spherical shape with various diameter ranging from 1.226 to 7.655 µm, due to various elements in fly ash as shown in Table 1.

Table 3
Composition of fly ash particles, using XRF technique

Composition	Concentration (wt %)
SiO_2	44.1
Al_2O_3	19.1
CaO	13.5
Fe_2O_3	12.4
MgO	4.7
Na_2O	2.9
K_2O	1.4
SO_3	1.0
TiO_2	0.7
Mn_2O_3	0.1
P_2O_5	0.1

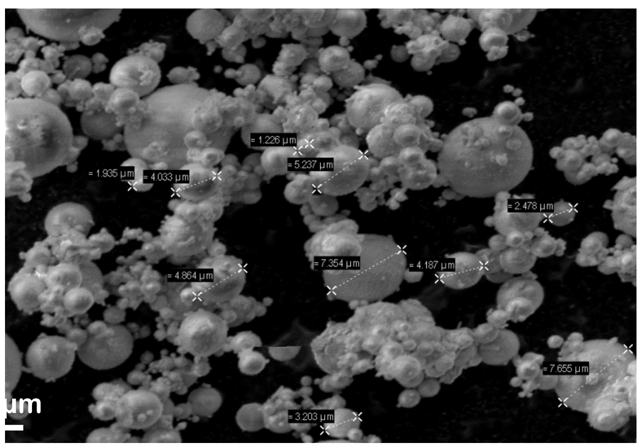


Fig. 2. SEM images of fly ash particles

3.2 Effect of Current Density and Fly Ash Composition on the Surface Morphology of Electrodeposited Ni-FA Composite Coating

The SEM images of the effect of current density on Ni-FA composite coating are shown in Figure 3. The SEM micrograph images show a colony like morphology that consist lots of nickel grains which have various sizes. For current density 1 A/dm^2, the SEM morphology show more compact structure (Figure 3 (a)). However, when current density increased to 5 A/dm^2, the structure starts to become larger and less compact (Figure 3 (b)). Then, at current density of 9 A/dm^2, the image shows that the number of colonies like morphology increased and the structure become larger (Figure 3 (c)). Thus, the deposition of the Ni-FA composite coating at high current density results in rough surface coating compared to deposit at low current density. The SEM morphology points that the crystal shape change from smaller to larger crystal shape due to increasing current density which resulted the shaped of the crystal become larger and denser [11].

According to Baghal *et al.*, [17], increasing current density results in higher over-potential which in turn increases the density of atomic clusters and so enhances the nucleation rate of the coating.

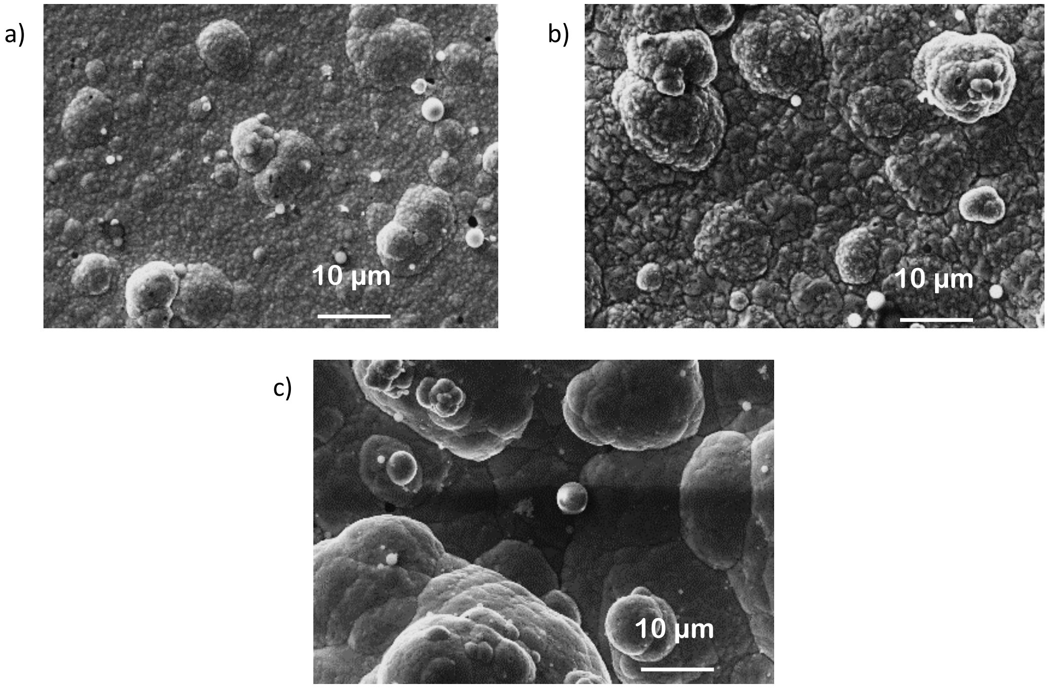


Fig. 3. SEM micrographs on the effect of current density on Ni-FA composite coating with 50 g/l of fly ash and current density (a) 1 A/dm^2 (b) 5 A/dm^2 (c) 9 A/dm^2

Figure 4 shows the comparison of SEM images of pure nickel coating (Figure 4 (a)) between Ni-FA composite coatings at various fly ash composition (Figure 4 (b-d)). The co-deposition of fly ash particles at 10 g/l shows a rough surface morphology (Figure 3 (b)), compared to pure nickel coating (Figure 3(a)). As the fly ash composition increased from 10 g/l to 50 g/l the image shows the agglomeration of colonies like morphology. This is due to the hindering of nickel grain growth during the deposition process. This was confirmed by another researcher which found that the fly ash particles were trapped within the matrix of nickel which hinder the growth of nickel grains [18]. Thus, the Ni-FA composite coating tends to become agglomerate and form more layers on the surface of the coating. When the composition of fly ash at 90 g/l, the images shows smoother surface although there have agglomeration. This is because the concentration of fly ash is too high thus the fly ash particles are fill up the gap between the nickel grains during the deposition process. The reinforcement of fly ash particles in the nickel matrix was confirmed and the size of fly ash particles were measured and it co-inside with the range of FA particles blend in the plating solution (Figure 5).

3.3 Effect of Current Density and Fly Ash composition on the Grain Size of Electrodeposited Ni-FA Composite Coating

Figure 6 shows the effect of current density and fly ash composition on nickel grain size of Ni-FA composite coating. As seen in Figure 6, the co-deposition of FA particles at various composition in the Ni-FA composite coating under 1 A/dm^2 has a significant effect on the grain size of nickel. At current density 1 A/dm^2 and 90 g/l of fly ash composition, the nickel grain size suddenly decreased. For both current density of 5 and 9 A/dm^2, there were no significant influence on the nickel grain size as the fly ash composition increased from 10 to 90 g/l.

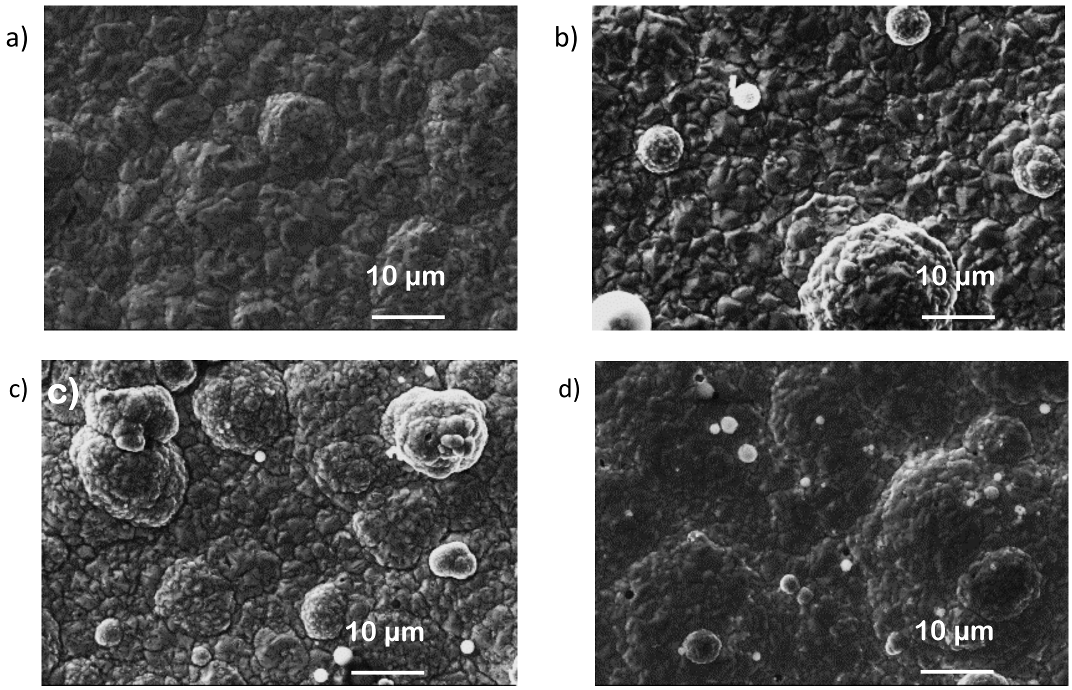


Fig. 4. SEM micrographs on the effect of fly ash composition on the Ni-FA composite coating with current density of 5 A/dm^2 and fly ash composition of (a) without FA particles (b) 10 g/l (c) 50 g/l (d) 90 g/l

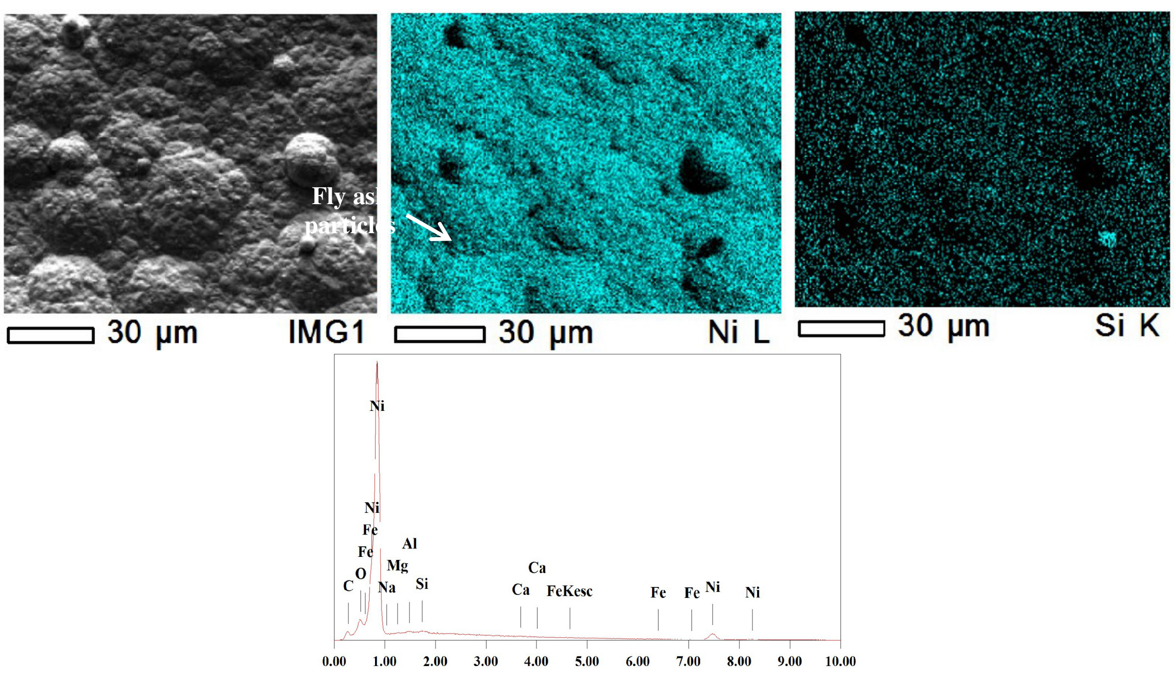


Fig. 5. EDX analysis of Ni-FA composite coating produced at 5 A/dm^2 and 50 g/l fly ash, confirming the evidence of fly ash particles in the coating

However, when the current density increased from 5 to 9 A/dm^2, the nickel grain size is decreased. This is due to the grain refining at high current density during the deposition process. Based on the electrochemical theory, the electrodeposition will produce nanostructures when deposition parameter is chosen so that the nucleation of new grains is formed rather than growth of the existing grains [19]. This is due to low current density of deposition process which the nucleation rate is slow, thus nickel grain has sufficient time to growth.

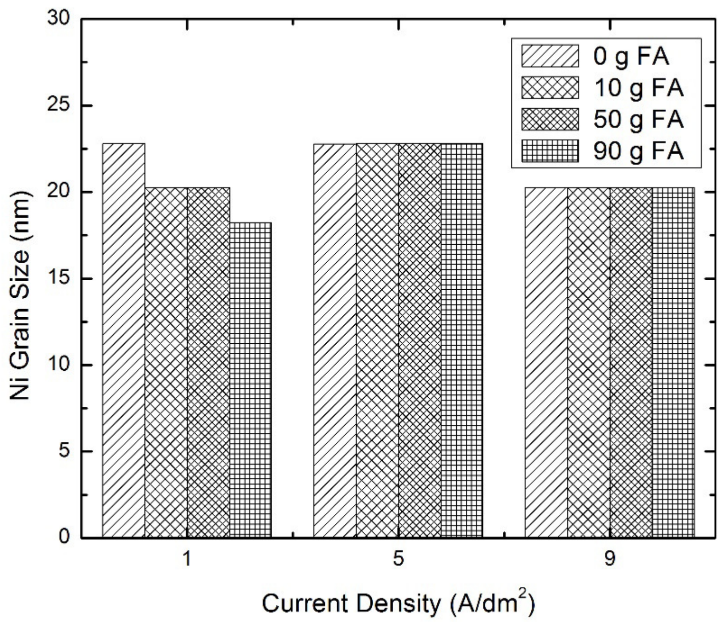


Fig. 6. The effect of current density and fly ash composition on the grain size of nickel of Ni-FA composite coating

3.4 Effect of Current Density and Fly Ash composition on Microhardness of Electrodeposited Ni-FA Composite Coating

The microhardness of Ni-FA composite coating increased as the current density of electrodeposition increased (Figure 7). The hardness value gradually increases from current density of 1 A to 9 A/dm^2. According to Yundong *et al.*, [12], the increase in hardness value due to the grain refinement effect by increased in current density. Besides that, by increasing current density also result in higher over-potential which then turn to increases the density of atomic cluster thus, enhances the nucleation rate of the Ni matrix [17]. As current density increased the deposition of matrix metal is fast enough to entrap and occlude some of the particles and incorporate them into deposits [13].

The hardness value also increased due the incorporated of fly ash particles in the Ni matrix solution. This is because of the presence of hard oxide, such as SiO_2 and Al_2O_3 particles in the FA. Besides, the modification of microstructures of the composite coating due to the co-deposition of FA particles to the coatings, which results in refining the nickel grains. Other than that, the dispersion hardening of small fly ash particles is deposited along the grain boundaries of the crystalline Ni matrix [5,18].

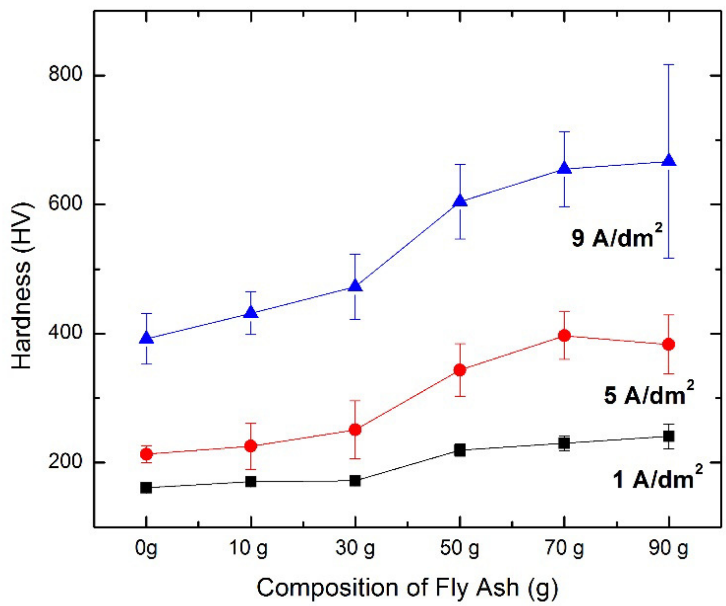


Fig. 7. Microhardness of Ni-FA composite coatings at various current density

4. Conclusion

From the experiment investigation given, the following deduction can be made:

- Ni-FA composite coatings were deposited on zincated AA6061 substrate at various current density. As current density increased, the colonies like morphology become larger and denser.
- As the concentration of fly ash particle increase in the nickel solution the Ni-FA composite coating tends to become agglomerate and form more layer on the surface of the coating.
- The nickel grain size decrease as the current density of the electrodeposition of Ni-FA composite coating increased.
- Micro-hardness of Ni-FA composite coatings shows that as current density increases, the hardness value also increases. Besides, the present of fly ash particles also increased the hardness value due to the presences of hard oxide in the FA particles.

Acknowledgement
We greatly acknowledge the research funding from Research Acculturation Grants Scheme code RAGS/1/2014/TK04/FKP/B00074-Universiti Teknikal Malaysia Melaka (UTeM) for sponsoring this work.

References

[1] Leon, J. Stephen, and V. Jayakumar. "Investigation of mechanical properties of aluminium 6061 alloy friction stir welding." *American Journal of Mechanical Engineering and Automation* 1, no. 1 (2014): 6.

[2] Wang, Yongjun, Ke Zhang, Weichao Wu, Wang Wang, and Junbiao Wang. "Effect of electrical pulse treatment on the retrogression and re-aging behavior of 6061 aluminum alloy." *Materials Science and Engineering: A* 703 (2017): 559-566.

[3] Bocking, C., and A. Reynolds. "Mechanism of adhesion failure of anodised coatings on 7075 aluminium alloy." *Transactions of the IMF* 89, no. 6 (2011): 298-302.

[4] Visser, P. "Novel totally chrome free corrosion inhibiting coating technology for protection of aluminium

alloys." *Transactions of the IMF* 89, no. 6 (2011): 291-294.

[5] Nguyen, Viet Hue, Thi Anh Tuyet Ngo, Hong Hanh Pham, and Ngoc Phong Nguyen. "Nickel composite plating with fly ash as inert particle." *Transactions of Nonferrous Metals Society of China* 23, no. 8 (2013): 2348-2353.

[6] Boonyongmaneerat, Yuttanant, Kanokwan Saengkiettiyut, Sawalee Saenapitak, and Supin Sangsuk. "Effects of WC addition on structure and hardness of electrodeposited Ni–W." *Surface and Coatings Technology* 203, no. 23 (2009): 3590-3594.

[7] Lins, Vanessa FC, Erik S. Cecconello, and Tulio Matencio. "Effect of the current density on morphology, porosity, and tribological properties of electrodeposited nickel on copper." *Journal of Materials Engineering and Performance* 17, no. 5 (2008): 741-745.

[8] Khairul Nizar, I., Abdullah Mohd Mustafa Al Bakri, A. R. Rafiza, Hussin Kamarudin, Alida Abdullah, and Zarina Yahya. "Study on physical and chemical properties of fly ash from different area in Malaysia." In *Key Engineering Materials*, vol. 594, pp. 985-989. Trans Tech Publications, 2014.

[9] Obi, E. R., I. N. A. Oguocha, and R. W. Evitts. "Effect of fly ash reinforcement on the corrosion NaCl solution." *Computational Methods and Experiments in Materials Characterization III* 57 (2007): 11121.

[10] Marin, Elia, Maria Lekka, Francesco Andreatta, Lorenzo Fedrizzi, Grigorios Itskos, Aggeliki Moutsatsou, Nikolaos Koukouzas, and Niki Kouloumbi. "Electrochemical study of Aluminum-Fly Ash composites obtained by powder metallurgy." *Materials Characterization* 69 (2012): 16-30.

[11] Rashidi, A. M., and A. Amadeh. "The effect of current density on the grain size of electrodeposited nanocrystalline nickel coatings." *Surface and Coatings Technology* 202, no. 16 (2008): 3772-3776.

[12] Li, Yundong, Hui Jiang, Weihua Huang, and Hui Tian. "Effects of peak current density on the mechanical properties of nanocrystalline Ni–Co alloys produced by pulse electrodeposition." *Applied Surface Science* 254, no. 21 (2008): 6865-6869.

[13] kumar Singh, Dhananjay, Manoj Kumar Tripathi, and V. B. Singh. "Electrocodeposition and characterization of Ni-WC composite coating from non–aqueous bath." (2013).

[14] NabiRahni, D. M. A., Peter Torben Tang, and Peter Leisner. "The electrolytic plating of compositionally modulated alloys and laminated metal nano-structures based on an automated computer-controlled dual-bath system." *Nanotechnology* 7, no. 2 (1996): 134.

[15] Yasaka, M. (2010). "X-ray thin-film measurement techniques. V. X-Ray Reflectivity Measurement." *The Rigaku Journal* 26, no. 2 (2010): 1-9.

[16] ASTM Committee C-09 on Concrete and Concrete Aggregates. *Standard specification for coal fly ash and raw or calcined natural pozzolan for use in concrete*. ASTM International, 2013.

[17] Baghal, SM Lari, M. Heydarzadeh Sohi, and A. Amadeh. "A functionally gradient nano-Ni–Co/SiC composite coating on aluminum and its tribological properties." *Surface and coatings Technology* 206, no. 19-20 (2012): 4032-4039.

[18] Panagopoulos, C. N., and E. P. Georgiou. "Surface mechanical behaviour of composite Ni–P–fly ash/zincate coated aluminium alloy." *Applied Surface Science* 255, no. 13-14 (2009): 6499-6503.

[19] Li, Yundong, Hui Jiang, Lijuan Pang, Bao'an Wang, and Xiaohui Liang. "Novel application of nanocrystalline nickel electrodeposit: Making good diamond tools easily, efficiently and economically." *Surface and Coatings technology* 201, no. 12 (2007): 5925-5930.