

Influence of ball milling duration of quarry dust on the properties of nickel-quarry dust composite coating

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ABSTRACT

The research focuses on the effect of various ball milling duration on the surface morphology, hardness and wear properties of nickel-quarry dust (Ni-QD) composite coating electrodeposited on aluminium alloy 6061 (AA6061) substrate. The quarry dust particles were prepared by ball milling process at 5, 10, 15 and 20 hours. X-ray Fluorescence (XRF), X-ray Diffraction (XRD) and Scanning Electron Microscope (SEM) analyses were carried out in order to investigate the influence of ball milling duration on the prepared quarry dust as well as the produced composite coating. In addition, microhardness Vickers and wear testing of Ni-QD composite coatings were also performed in this study. The results obtained indicate that the quarry dust particle size is influenced by increasing the ball milling duration. Increase in ball milling duration from 5 to 20 hours results to finer particle size, which improves the microhardness of the Ni-QD composite coatings from 190.6 HV to 282.2 HV. The increase in microhardness values is due to the increase incorporation of fine quarry dust particle in the Ni coating. Furthermore, the wear resistance of the Ni-QD composite coatings were improved, as the ball milling duration increased from 5 to 20 hours.

Keywords: Nickel; quarry dust; composite coating; aluminium alloy 6061; wear; hardness.

INTRODUCTION

Quarry industry in Malaysia plays a significant and essential role in the development of the country. However, the environmental concern is currently rising as one of the main challenging issues affecting the natural aggregate and limestone production. According to Sridharan et al. (2006), about 20- 25% of total production in each crusher unit has left out the quarry dust as waste material [1]. Therefore, efforts have been taken to control environmental pollution arising due to disposal of these industrial wastes by converting them into utilizable raw materials for usable application. Quarry dust is one of the by-product from the crushing process during quarrying activities, which has gained attention to be used for various applications, such as construction industry and manufacturing of building

material industry, due to the high percentage of ceramic particles, SiO_2 and Al_2O_3 . A considerable amount of literature has been published on the utilization of the quarry dust as an alternative material to replace natural river sand in concrete manufacturing industry [2-5]. These studies focus on the improvement of the mechanical properties with the addition of quarry dust in conventional concrete. As noted by Mustafa Al Bakri (2013), compressive strength of concrete with quarry dust as aggregates is higher than concrete without quarry dust. Therefore, quarry dust can be the alternative for natural river sand in concrete manufacturing industry [3]. Aluminium matrix reinforced with ceramic particles has already drawn attention to be used in automobile and aerospace applications [6-8]. This is because of many admirable properties such as, high strength, high stiffness, high thermal conductivity and combined properties like wear resistance with fracture toughness and high strength with corrosion resistance. There is a significant improvement in the mechanical properties and wear resistance of the composites with addition of quarry dust particulates than the base alloy. By incorporating 7.5% of quarry dust to the metal matrix, the tensile strength of the composite is 15% greater than the base alloy [4]. According to Ramesh et al. (2014), the maximum hardness of 82 BHN is found at 10% of quarry dust reinforcement in aluminium-quarry dust composites which exhibits 13.7% improvement of hardness compared to base matrix alloy [4]. Other than that, increase in the quarry dust content from 10 to 20 wt% increases the resistance to wear [7]. Based on the great performance of the quarry dust in improving the mechanical properties and wear resistance of conventional concrete and MMCs, the utilization of the quarry dust has been extended to be used as a reinforcement in MMCs for coating application in micro channel reactor [9]. There is a significant improvement in the mechanical properties, wear resistance and corrosion resistance of the composite's coatings due to the addition of ceramic particles into the metal matrix. Even though previous investigators have examined the effects of ceramic particles like SiC [10], Al_2O_3 [11], TiC, B4C, MgO, Si_3N_4 and SiO_2 [12] to the properties of MMCs coatings, no studies have been found to be using the recycled quarry dust as a source for the ceramic particles. In producing nickel- quarry dust composite coating, the preparation of quarry dust particles is essential. The quarry dust characteristic, such as the particle size influences the properties of the composite. The quarry dust particles are prepared by using ball milling process. The duration of the milling process affects the characteristics of quarry dust, such as the particle size and dispersion of the quarry dust particles, which will consequently affect the nickel- quarry dust composite coating properties. Thus, this paper focuses on the effect of ball milling duration on the characteristics, hardness and wear properties of nickel- quarry dust composite coating.

METHODS AND MATERIALS

Size Modification on the Quarry Dust

The quarry dust was obtained from a quarry in Negeri Sembilan. Quarry dust was milled using a mixer process for 2 hours to ensure the particles were mixed well during the process. After that, the as-received quarry dust was crushed using a disc milling machine (Retsch, DM200). Then, the quarry dust was ball- milled for 5, 10, 15 and 20 hours using a conventional ball milling equipment. In this study, the ball- milled quarry dust produced was compared using the disc milling equipment and conventional ball milling equipment. The

particles compositions and sizes were analysed using X- ray Fluorescence (XRF), Particle Size Analyzer (PSA) (Malvern, 2000) and Scanning Electron Microscopy (SEM) characterization tools.

Surface Pre-Treatment of the Aluminium Alloy Substrate Prior to Electrodeposition Process

The AA6061 substrates with the dimension 40 mm x 30 mm x 3 mm were ground using silicon carbide paper sand mechanically polished with diamond paste. The substrates were firstly immersed in 10 wt. % of sodium hydroxide (NaOH) solution for 10 seconds and followed by the immersion in 30 vol. % of nitric acid (HNO₃) solution for 20 seconds. Prior to the deposition of Ni-QD composite coatings, a double zincating process was conducted on the substrates for 45 seconds for the first zincating and for 15 seconds for the second zincating. Acidic etching for 15 seconds in HNO₃ solution at room temperature was performed between consecutive zincating operations.

Electrodeposition Process of Nickel- Quarry Dust Composite Coatings

The pre-treated substrates were then attached to a three-electrode system glass cell fitted with a water jacket, through which water from a thermostat bath (Daihan Scientific Co, WB 11) was circulated. A power supply (Keysight) was employed for nickel electrodeposition using a typical two-electrode system consisting of a working electrode (WE) of AA7075 and a counter electrode (CE) of a nickel plate (99.99%). During the electrodeposition process, the nickel electrolyte was stirred continuously with a PTFE-coated magnetic stirrer at 300 rpm using a magnetic stirrer equipment. The current density used for the electrodeposition process was 3 A/ dm² under direct current mode with the total time of electrodeposition set at one hour. The quarry dust particles which were prepared at various ball milling durations were added to the electrolyte. The chemical composition and operating condition for electrodeposition of Ni-QD composite coating are summarized in Table 1 and Table 2. The schematic diagram of electrodeposition set-up is shown in Figure 1.

Table 1. Composition of modified nickel Watt's bath

Chemical compound	Concentration (g/L)
Nickel sulphate hexahydrate	200
Nickel chloride	20
Sodium citrate	15
Quarry dust prepared from 5,10,15 and 20 hours ball milling duration	30

Table 2. Electrodeposition operating condition.

Parameter	Condition
Temperature (°C)	40
Time (min)	60
Current density (A/dm ²)	3
Speed (rpm)	300

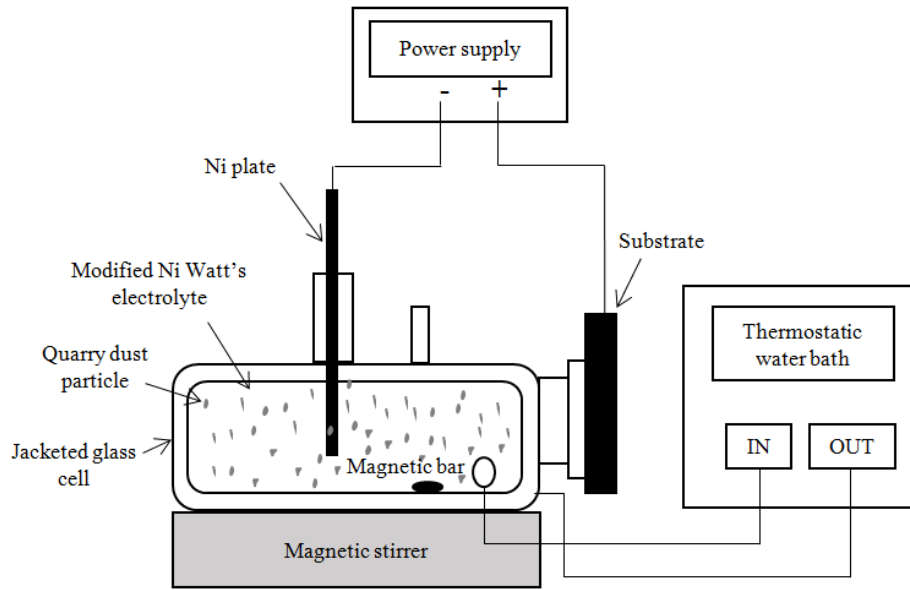


Figure 1. Schematic diagram of electrodeposition process.

Characterization and Testing of Quarry Dust and Composite Coatings Wear Testing

The wear behaviour of the composite coating was analysed by using high frequency reciprocating rig (HFRR) test. The schematic diagram of HFRR wear tester is shown in Figure 2. A stainless-steel ball with 6 mm in diameter and 60 HRC in hardness were used in reciprocate manner by the machine against the sample under constant load of 10 N. The frequency was set constant to 5 Hz. The sliding time was set to 0.5 hours and the stroke length was 2 mm. This testing was carried out at room temperature. The wear behaviour of the coating was noted by analysing the tribology properties of the wear scar.

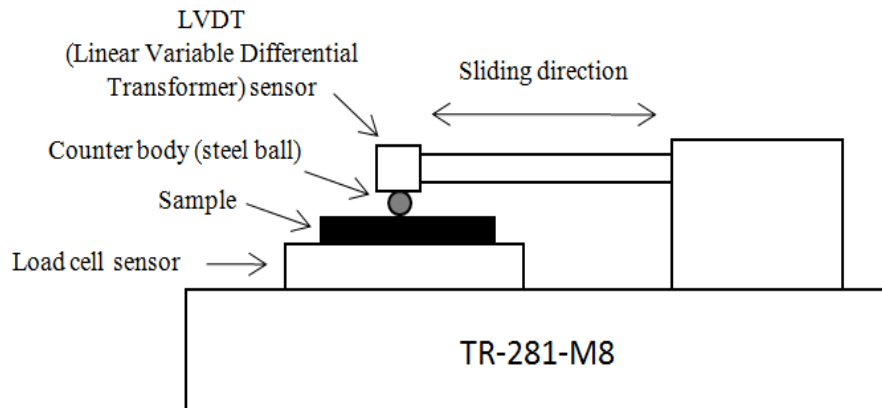


Figure 2. Schematic diagram of HFRR wear tester.

Hardness Testing

Microhardness testing was conducted using Vickers microhardness tester. The indentation time was set to 15 seconds in each indentation. A set of 10 indentations were acquired for each of the testing. The load was fixed to 0.5 Kgf. The hardness value (HV) was calculated automatically by the computer measuring the average distance of the diamond- shape produce after the indentation.

X-Ray Fluorescence Spectroscopy

The elemental compositions of quarry dust were determined by an X-ray fluorescence (XRF) spectrometer prepared by compacting powder quarry dust into disc shape. The powder was sieved below 40 μm before it was turned to disc shape.

X-Ray Diffraction

XRD measurements were carried out by (PAN Analytical X'Pert Pro) using ($\lambda = 1.5^\circ$) in the 2θ range of $10-90^\circ$. The sample of quarry dust was prepared in the powder form and the sample of the coating was by electrodepositing the substrate.

Morphology Studies

A scanning electron microscopy (SEM) (Zeiss Evo 50) was used to evaluate the morphology of quarry dust and wear test. The images were taken at suitable accelerating voltages using the secondary electron imaging.

Particle Size Measurement

Particle size analyzer (PSA) (Malvern, 2000) was used to analyse the size distribution of quarry dust particles. The quarry dust was analysed in the powder form.

RESULTS AND DISCUSSION

Characterization of Quarry Dust Particle

Figure 3 demonstrates the quarry dust particle size from as received to 20 hours after ball milling process. It is apparent in the figure that the particle size has been reduced from $330.28\mu\text{m}$ to $157.12\mu\text{m}$ after ball milling process for 20 hours. The particle size of quarry dust becomes finer after the ball mill process, due to the collision between the balls and quarry dust particles.

Table 3 demonstrates the concentration of quarry dust particles as found by XRF procedure. The elemental composition for quarry dust particles is found by XRF technique and shown in Table 3. The quarry dust particles contain silica (SiO_2), alumina (Al_2O_3), calcium oxide (CaO), press (III) oxide (Fe_2O_3), magnesium oxide (MgO), sodium oxide (Na_2O), potassium oxide (K_2O), sulfur trioxide (SO_3), titanium dioxide (TiO_2) and phosphorus pentoxide (P_2O_5). It can be concluded from this XRF result that the quarry dust particles contain of high content of SiO_2 and Al_2O_3 elements, which these hard oxides are widely used as a reinforcement in composites. This result is consistent with Cohen et al. (2018), who developed a sustainable alternative building material from quarry dust [13].

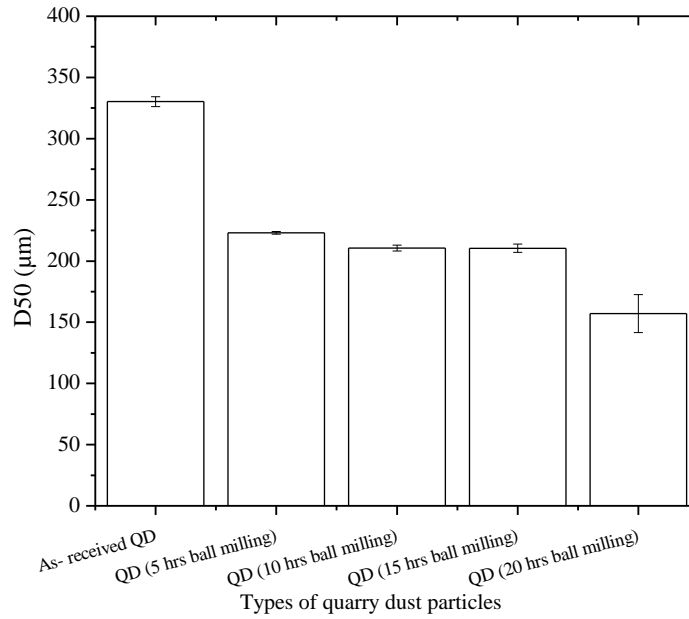


Figure 3. Particle size distribution of various quarry dust particles.

Table 3: The concentration of quarry dust particles as found by XRF technique.

Element	SiO ₂	Al ₂ O ₃	K ₂ O	Na ₂ O	Fe ₂ O ₃	CaO	MgO	TiO ₂	SO ₃	P ₂ O ₅
Concentration (Wt%)	72.6	15.1	4.9	3.0	1.9	1.1	0.8	0.3	0.2	0.1

The morphology of quarry dust particles was studied by SEM. It can be seen clearly from Figure 4 that the quarry dust particles are in irregular and angular shape with various sizes. It is found that, the morphology of quarry dust particles consisted of smaller particles sizes from 190µm to 157µm at increased ball milling duration (Figure 4(b and c)), compared to as received (Figure 4(a)). It is also found that the 20 hours ball milled quarry dust particles become less agglomerated compared to crushed and 10 hours ball milled quarry dust.

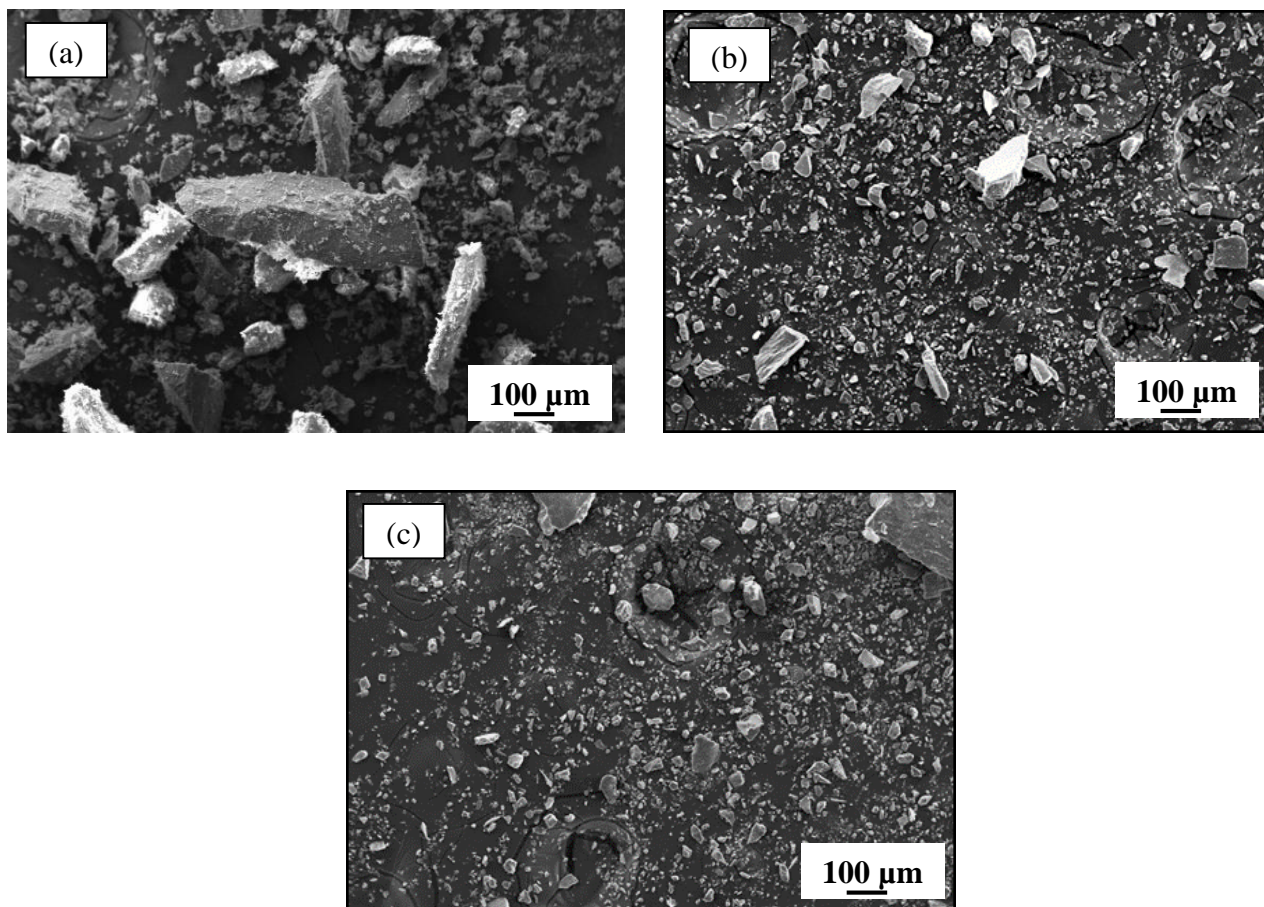


Figure 4. SEM images of quarry dust at various conditions (a) as received, (b) after 10 hours of ball milling and (c) after 20 hours of ball milling process.

Characterization of Nickel-Quarry Dust Composite Coatings

The comparative result of XRD pattern on the pure nickel and nickel-quarry dust composite coatings at various ball milling durations are shown in Figure 5. It demonstrates that the scale on pure nickel coating only consists of nickel, while the scale on nickel composite coatings constitutes SiO_2 . The occurrence of this phase in the nickel composite coatings indicates that quarry dust particles have successfully been incorporated with nickel matrix in the deposits. At position 2θ of 65.10° and 76.49° , SiO_2 exists on each of the nickel-quarry dust composite coatings at various ball milling durations, compared to pure nickel coating. This observation obviously shows that the introduction of quarry dust particles into nickel matrix has a significant effect on the intensity peak of the Ni, SiO_2 , and Al. The SiO_2 's peak at 65.10° increases with increase in the ball milling duration. This is most probably due to the ball milling process which has refined the quarry dust particles. Thus, more quarry dust particles floated in the electrolyte and was trapped in the nickel matrix during the electrodeposition process.

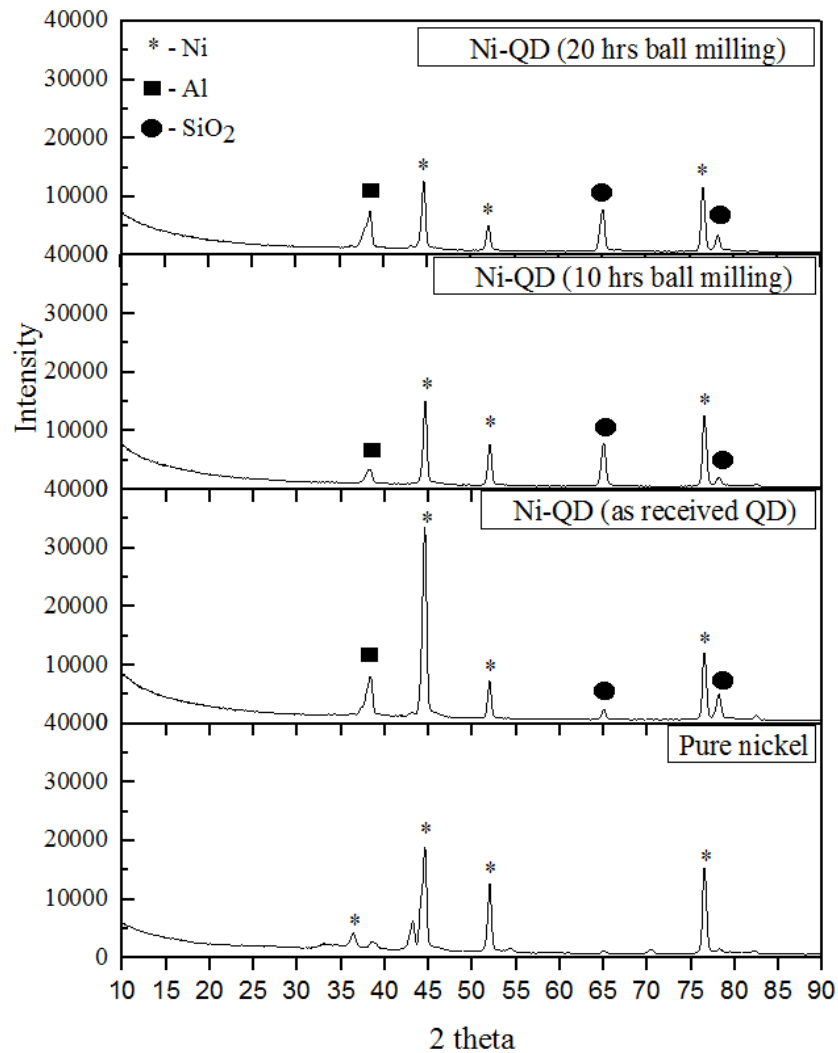


Figure 5. XRD pattern of nickel composite coating at various ball milling durations.

Hardness and Wear Testing of Nickel- Quarry Dust Composite Coatings

There is a clear trend of increase in the microhardness values as the ball milling duration increases, as shown in Figure 6. It is apparent in the figure that the microhardness of the nickel- quarry dust composite coatings produced from ball milled quarry dust was higher than the as- received quarry dust. The hardness value gradually increases from ball milled duration of 5 to 20 hours due to the refining size of quarry dust particles from the ball milling process. The refining in the quarry dust particle size has led to the high density of quarry dust particle in the electrolyte during the electrodeposition process. Thus, nickel ions in the electrolyte entrap more particles and incorporate them into the deposit, as illustrated in Figure 7. Quarry dust particles consist of hard oxide particles, such as SiO₂ and Al₂O₃ particles. This is also consistent with the work of Viet et al. (2013), who found that increase in the fly ash concentration in solution leads to the improvement of microhardness values, due to an increased content of fly ash incorporated to deposit [14].

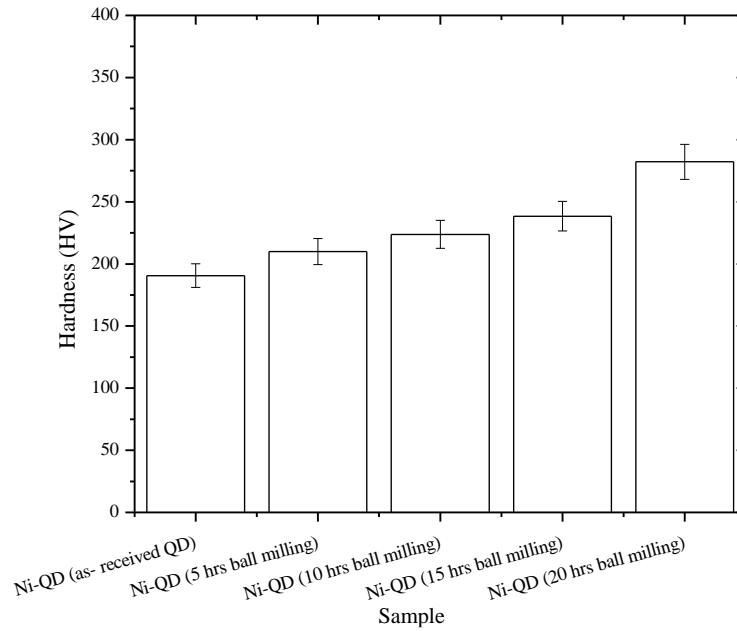


Figure 6. Microhardness value of nickel- quarry dust composite coating at various ball milling durations.

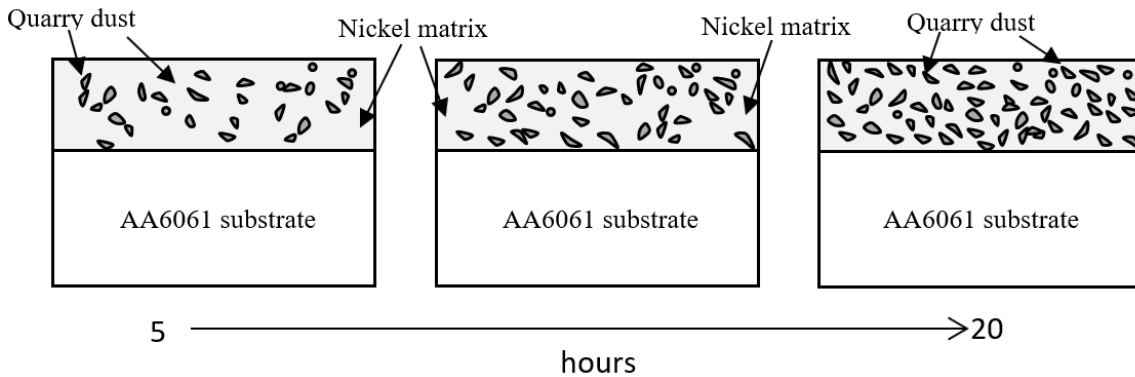


Figure 7. Illustration of cross- section of nickel- quarry dust composite coatings at various ball milling durations.

Figure 8 shows the wear scar analyzed by using SEM to identify the wear mechanism occurred on the nickel-quarry dust composite coatings produced at various ball milling durations. From the SEM images in Figure 8, the width (vertical line) and length (horizontal line) of wear scar were identified. Figure 9 shows the wear scar's width and length of bare AA6061 and nickel-quarry dust composite coatings at various ball milling durations, 10 and 20 hours after friction against steel ball. It is clearly shown that the wear scar's width and length were reduced significantly when nickel matrix was deposited with quarry dust. It is apparent from the figure that without any coating material on the substrate, rigorous damage

has occurred due to the direct contact of the steel ball with the substrate which can increase wear (Figure 8 (a)).

This is most probably due to the low hardness value of the AA6061 substrate. Introduction of quarry dust particles into the nickel matrix has increased the microhardness and reduced the width and the length of the wear scar (Figure 8 (b- d)). The damage of the wear scar was improved, as the ball milling duration increases from 5 to 20 hours.

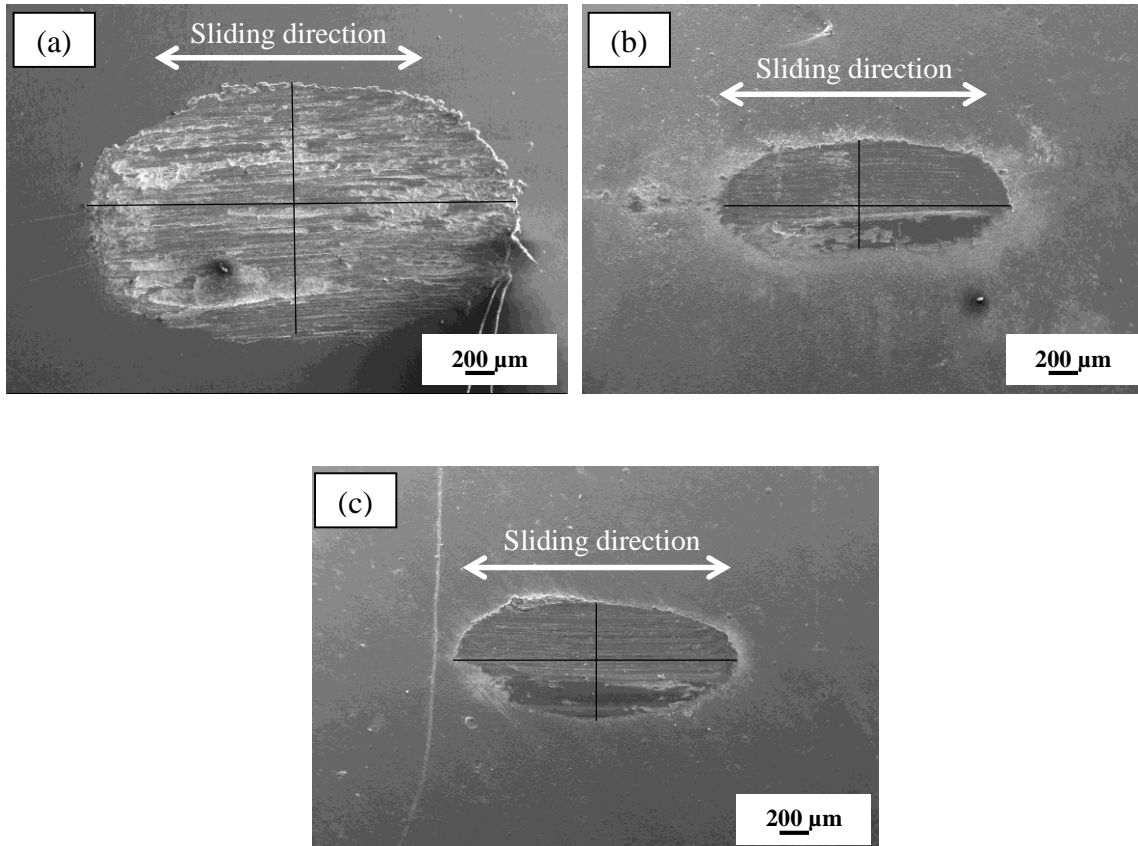


Figure 8. Wear track of (a) bare AA6061 and nickel- quarry dust composite coating at various ball milling durations, (b) 10 hours and (c) 20 hours.

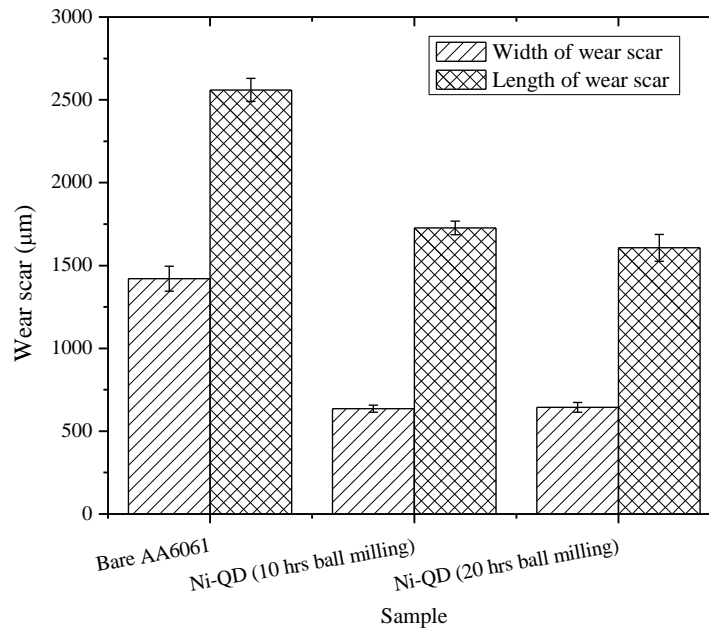


Figure 9. Wear scar width and length of bare AA6061 and nickel- quarry dust composite coating at various ball milling durations 10 and 20 hours.

Figure 10 shows the magnified SEM images of wear scar in Figure 8. Introduction of quarry dust particles into the nickel matrix has reduced the worn- out patches on the wear scar (Figure 10 (b- d)). The worn patches are consisted of longitudinal furrows and small size grooves along the sliding direction of the steel ball [15]. It is obviously seen that the surface material is pushed to the sides of the wear scar, as well as the formation of wear debris expelled to the edge of the scar. The scar indicates a typical aspect of ploughing wear.

Figure 11 shows the friction coefficient value of bare aluminium alloy 6061 and nickel-quarry dust composite coatings at 10 and 20 hours after friction against steel ball. It is obviously seen that the bare aluminum alloy 6061 possesses the highest friction coefficient compared with the nickel-quarry dust composite coatings. This is probably because of the incorporation of quarry dust particles in the nickel matrix, which acts as a solid lubricant during the wear process

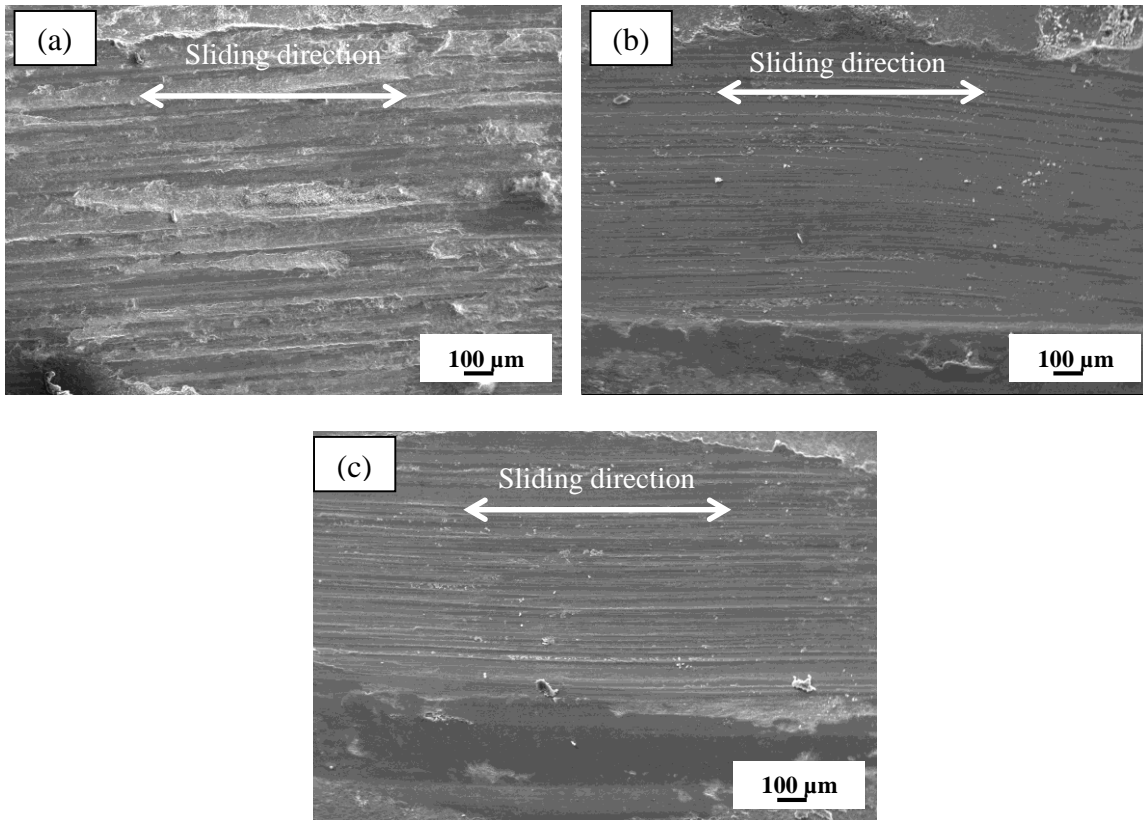


Figure 10. SEM images of magnified wear track of (a) bare AA6061 and nickel- quarry dust composite coating at various ball milling durations (b) 10 hours and (c) 20 hours.

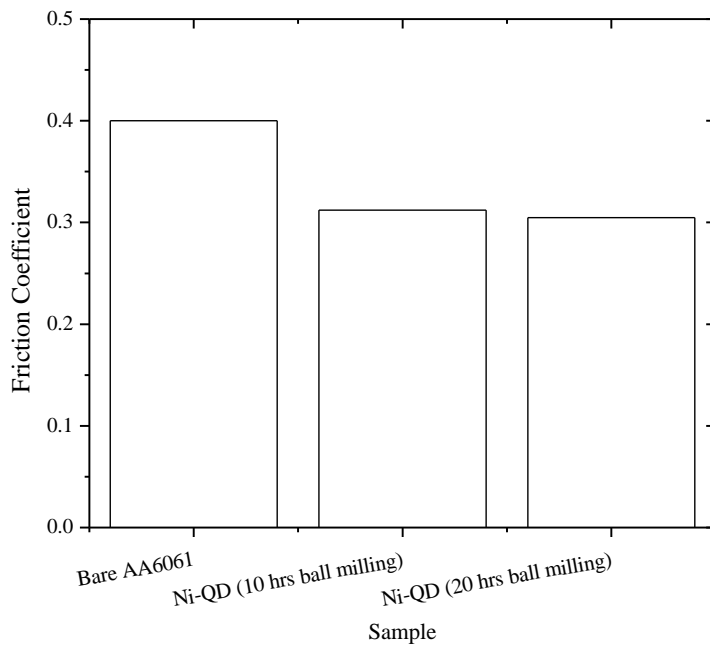


Figure 11. COF value of bare aluminium alloy 6061 and different ball mill durations.

Table 4 shows the surface roughness of bare aluminium alloy 6061 and nickel- quarry dust composite coatings produced at different ball milling durations. It clearly shows that the surface roughness values of bare aluminium alloy 6061 and composite coatings are slightly similar to each other. These embedded quarry dust particles in nickel matrix show no significant difference to the surface roughness of the composite coatings.

Table 4. Surface roughness values of composite coatings.

Sample	Surface roughness (μm)
Bare AA6061	0.66
Ni-QD (10 hours of ball milling duration)	0.65
Ni-QD (20 hours of ball milling duration)	0.65

CONCLUSIONS

From the investigation, the quarry dust particles size decreases as the ball milling process increases from 5 to 20 hours. The surface morphology of the quarry dust particles shows less agglomeration of ball- milled particles, compared to as- received particles. The XRD peak of SiO_2 at 65.10° increases with the increase in the ball milling duration due to the high density of quarry dust in the electrolyte. The high density of quarry dust in the electrolyte results in high incorporation of quarry dust in the nickel matrix, which improves the microhardness value of the composite coating. Microhardness of nickel composite coatings increases as the quarry dust content increases from 190.6 HV to 282.2 HV due to the presence of hard oxide in the particles. The damage of the wear scar is improved, as the ball milling duration increases from 5 to 20 hours.

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