Jurnal Tribologi

Evaluation of corrosion and erosive wear behaviour of Co-Ni-Fe coating deposited by electrodeposition method

Nik Roselina Nik Roseley [1*], Koay Mei Hyie [2], Nik Rozlin Nik Masdek [1], Noor Syahadah Yussoff[1], Muhammad Ridzwan Abu[1]

[1] School of Mechanical Engineering, College of Engineering, Universiti Teknologi MARA, Shah Alam, 40450, Selangor, MALAYSIA.
[2] Centre for Mechanical Engineering Studies, Universiti Teknologi MARA Cawangan Pulau Pinang, Permatang Pauh Campus, 13500, Pulau Pinang, MALAYSIA.
*Corresponding author: roselina_roseley@uitm.edu.my

KEYWORDS	ABSTRACT
Co-Ni-Fe Coating Corrosion Wear	Corrosion and erosive wear are insidious problems in the offshore industry. It could not only contribute to the rise of cost for maintenance and repair but also may lead to hazardous working conditions. The potential reduction may be achieved by the application of appropriate coating to protect the surface. Current study reported fabrication and characterisation of Co-Ni-Fe coatings deposited at various deposition times. The main aims are to investigate the physical and mechanical properties, as well as the corrosion and erosive wear resistance behaviour of the coatings. Co-Ni-Fe coatings were deposited using electrodeposition method with deposition time of 30 to 90 minutes. The as-deposited coatings were characterised using Scanning Electron Microscope (SEM), Vicker Hardness, X-ray Diffraction (XRD) and Alicona Infinite Focus. The corrosive and wear behaviours of coatings were investigated using dip test and slurry test respectively. The findings showed the existence of voids on coatings deposited for 30 minutes. The particle size of Co-Ni-Fe increased as the deposition time was prolonged to 90 minutes with the largest size of 8 to 15 μm. Results showed that the corrosion and wear resistance behaviour of Co-Ni-Fe coatings improve by 32% and 78% with a prolonged deposition time of 90 minutes.

1.0 INTRODUCTION

Corrosion and erosive wear have been recognised as one of the main contributors to energy and material losses in many industries. The oil and gas industry for instance spent millions of its budget to overcome this issue. The major escalating cost in this industry comes from the maintenance of equipment which mainly is due to corrosion and wear. The outcome of the survey conducted by the NACE-IMPACT in 2016 reported that very low percentage of companies has a proper corrosion management practice (Prasad et al., 2020). In 2008, Simon reported that the annual cost of corrosion was estimated to be $1.372 billion for the oil and gas industry (Simon, 2008). Being in open space areas exposed the equipment to violent variables such as humidity and temperature, thus reducing the lifespan of the material and equipment in the oil and gas sector (Marita & Yaacob, 2010). Coating application to base materials has been proven to effectively protect materials in harsh conditions such as offshore.

Electrodeposition is one of the most facile methods for surface modification through coating deposition by electric current. In electrodeposition, the substrate to be coated will be immersed into an electrolyte solution containing the salt of the metal to be deposited. It is attached to the cathode in the electrolytic cell which acts as the negative pole of a power supply. Over the past decade, the electrodeposition method has been used by number of researchers to fabricate various types of coatings. Among the coatings are alloys and composite coatings which involve the integration of more than one type of element in the coating to improve the corrosion and wear performance of the surface (Mahalingam et al., 2009). Recently, huge attention has been given to the deposition of alloy matrix coatings such as Ni-Fe, Ni-Co-P and Co-Ni/WS2 (Torabinejad et al., 2017) (Sarkar et al., 2018) (Liu et al., 2020). Co-Ni-Fe coating is expected to be a good protective layer to improve mechanical, corrosion and erosive wear resistance. However, there are several considerations that need to be taken into account during deposition. One of the most influential parameters for coating deposition is deposition time. In our previous work, a short deposition time between 15 to 45 minutes was used to deposit Co-Ni-Fe coating (Hyie et al., 2016).

In this work, Co-Ni-Fe coatings were deposited on mild steel using deposition time from 30 to 90 minutes. The wear behaviour of the coatings was investigated using the Slurry Erosion Tester. The chemical, physical and mechanical properties of the coatings were characterised using XRD, SEM, Alicona Infinite Focus and Vickers microhardness tester.

2.0 EXPERIMENTAL PROCEDURE

Rectangular mild steel plate substrates with dimensions of 76 mm x 25 mm x 4 mm were prepared. The substrates were cleaned from the oxide layers using 80 to 320 grit sandpaper flat disks and then polished using a polishing disk grinder. Substrates were then cleaned with acetone and ethanol.

The electrodeposition process was conducted in a sulfate bath electrolyte. The electrolyte was prepared by dissolving analytical grades of cobalt sulfate ($CoSO_4$), nickel sulfate ($NiSO_4$), ferum sulfate ($FeSO_4$), boric acid (H_3BO_3) and Saccharin in 1000ml distilled water. The solution was mixed using a magnetic stirrer. Graphite was chosen as a counter electrode. Mild steel substrate was connected to the cathode, while the graphite was connected to the anode. The current and pH was maintained at 2.0A and 3 respectively, while the temperature was kept at 50°C. The duration of the coating deposition was various from 30 to 90 minutes. Figure 1 illustrates the schematic diagram of the experimental setup used in this work. The electrodes were completely immersed in an electrolyte. Prior to deposition, both magnetic stirrer and heater were turned off.
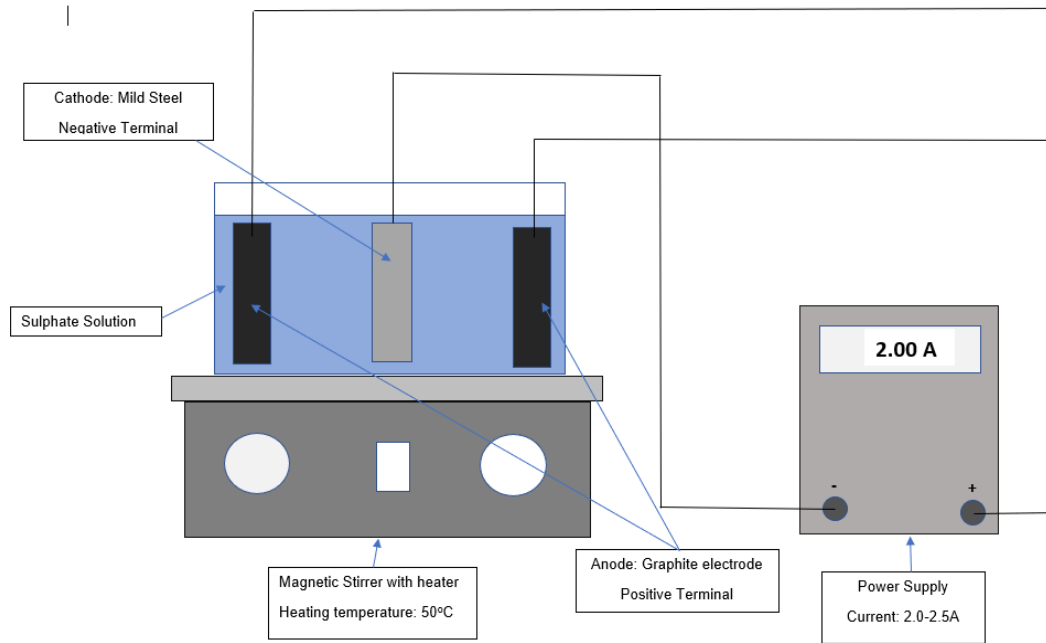
Figure 1: Schematic diagram of the experimental setup for the electrodeposition process.

The morphology of the deposited coatings was observed under Scanning Electron Microscope (SEM) with electron beam energy of 15keV using magnification between 500 to 3000x. The phase structure of coatings was investigated by XRD ULTIMA IV FD 3668N using a Cu target. The scan region (2θ) used was from 30° to 120° at a scan rate of 5° min-1 The microhardness of coatings was measured using Vicker Microhardness tester under indentation load of 1000 gram. An average of five (5) different measurements was recorded in plane as the coating hardness. Alicona Infinite Focus machine was used to obtain the surface roughness of Co-Ni-Fe coating sample.

Wear test was conducted using a Slurry Erosion Test Rig (Model TR-40 Ducom Instruments, USA) using ASTM G40 Standard. The schematic diagram of the slurry pot is shown in Figure 2. The coatings were placed at the specimen holder that was immersed in the slurry mixture. The slurry mixture comprises distilled water and 60% concentration of beach sand with an average particle size range of 125-250 μm at room temperature. Rotation of 1200 rpm was individually run for eight (8) hours with a time interval of two (2) hours for each coating. The operating parameters for the slurry test are summarized in Table 1. During the test, coatings were in contact with the sand particles which cause erosion on the surface. Three (3) samples for each coating type were tested. Samples were cleaned using acetone after each test before being weighed. The weight of the sample before and after the test was recorded.
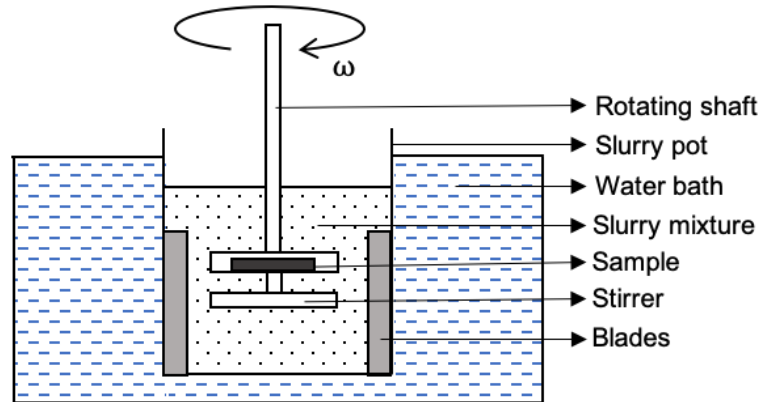
Figure 2: Schematic diagram of slurry pot in slurry erosion test.

Table1: Parameters for Slurry Erosion Test

Parameters	Experimental Condition
Slurry	Distilled water + 60% beach sand size 125-250 μm concentration
Rotation speed	1200 rpm
Rotation time	8 hours at intervals of 2 hours
Temperature	Room temperature

Corrosion test of the coating was performed by corrosion dip method. The corrosion dip test is a test that replicates the real condition of corrosive attack on surface to investigate the corrosion resistance capability of the coatings under a corrosive environment. The salt solution for the corrosion dip test was prepared by adding 5% of coarse sea salt (50g) and 95% of tap water (1000ml) to mimic the seawater at a ratio of 1:50. (Valdez, 2006). The initial temperature and pH of the salt solution were kept at 28°C and 7.6 respectively. All samples were fully immersed into the solution and placed at open-air and room temperature for 11 days (264 hours).

3.0   RESULTS AND DISCUSSION

The morphology of the coating was assessed using Scanning Electron Microscope (SEM). Figure 3(a-c) represent the micrographs of Co-Ni-Fe coatings deposited for 30, 60, and 90 minutes respectively. As can be seen in Figure 3, in general, the size of deposited particles increased as the deposition time increased. The size of particles for 60 and 90-minute depositions (Figure 3(b and c)) were measured to be approximately ~1 μm and ~1.5 μm respectively. Areas of void can be obviously seen in coating deposited for 30 minutes and are marked as the orange dotted box in Figure 3(a). Void may be the strong evidence of inadequate deposition time to entirely coat the surface of the specimen (Bockris & Damjanovic, 1964). Void in the coating vanished gradually as deposition time was increased. However, particles agglomeration can also be seen on some areas in coating deposited for 90 minutes. The particles distribution for coating deposited for 90 minutes are more compact and denser compared to the other coatings. The common observation has been reported by many other works.
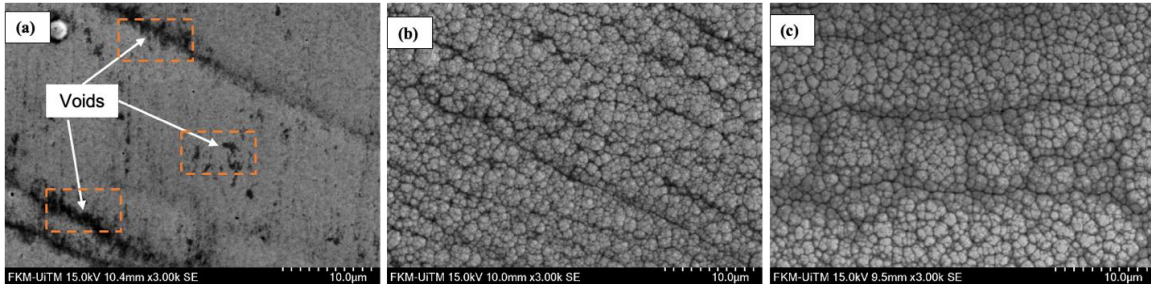
Figure 3: SEM images of Co-Ni-Fe coating deposited for (a) 30 minutes (b) 60 minutes and (c) 90 minutes.

In addition, the surface roughness of coatings was also evaluated. Surface roughness was measured using Alicona Infinite Focus. Figure 4 presents the correlation between surface roughness, Ra and coating deposition time. The surface roughness of coating deposited for 30 minutes was measured to be slightly higher than the bare substrate coating. However, the surface roughness was tremendously reduced as the deposition time increased to 60 and 90 minutes. The highest surface roughness of 3.77 µm was recorded from a 30-minute coating sample. This can be associated with the imperfection of coating deposited for 30 minutes that created areas with voids as in Figure 3(a). The voids create higher surface roughness for the samples (Lee and So, 2000). As the deposition time increases, the number of voids that exists on the coating surface may have been reduced thus reducing the surface roughness. Low surface roughness of coatings deposited for 60 and 90 minutes is also evidence that no severe particles agglomeration happened at higher deposition time. In general, a thin coating will replicate the surface morphology of the substrate. However, as the thickness of the coating increases, the surface roughness is expected to be lower than the substrate (Salvadori et al., 2006). Based on our previous work, the thickness of the coating can be estimated to be approximately 24.6 µm for 30 minutes deposition, and more than 63.8 µm for 90 minutes deposition (Hyie et al., 2016).
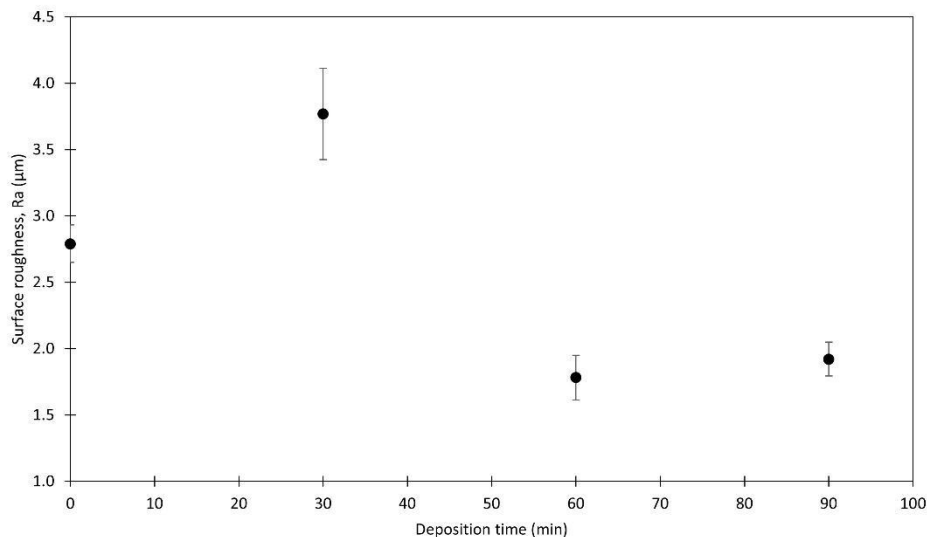

Figure 4: Surface roughness of Co-Ni-Fe coatings deposited for 30 to 90 minutes.

Figure 5 shows the XRD spectra of the Co-Ni-Fe coatings deposited for 30 to 90 minutes. As can be seen from the spectra all coatings show almost similar crystallite structures. The reflection pattern of all Co-Ni-Fe coatings showed the main characteristic peaks of (111), (200), (220) and (311) peaks of FCC structure. These peaks are associated with characteristic peaks of Ni-Co-Fe and are in agreement with many reported works (Dao et al., 2007, Herojit et al., 2016, Zavareh et al., 2014). However, the width of all peaks obtained from sample deposited for 30 minutes is smaller compared to the other two coatings. This indicates higher crystallinity and larger crystallite size for this coating (Cancado et al., 2006). No other element or compound can be detected in any of the coatings. Thus, it can be concluded that no contamination or side reaction occurred during the electrodeposition process.
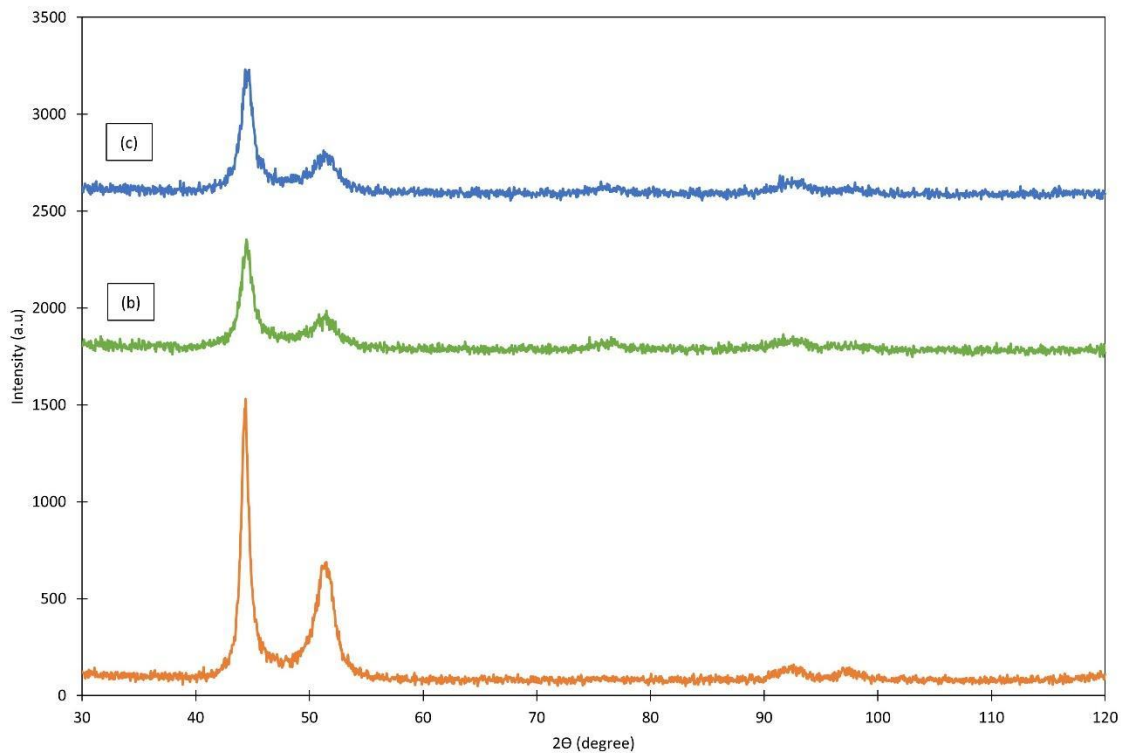

Figure 5: XRD spectra of Co-Ni-Fe coatings deposited for (a) 30 minutes, (b) 60 minutes and (c) 90 minutes.

Microhardness of the samples was measured with an average of five (5) measurements for each coating using Vickers Microhardness. The hardness of Ni-Co-Fe coatings deposited for 30, 60 and 90 minutes are plotted in Figure 6. The hardness of mild steel substrate was also measured and indicated as deposition 0 minute in the figure. The results show that the hardness of coatings gradually increased as the deposition time extended from 30 to 90 minutes. The highest coating hardness of 383.7 HV was recorded from the Co-Ni-Fe coating deposited for 90 minutes. In contrast, the coating deposited for 30 minutes showed the lowest hardness of 224.3 HV, approximately 33% higher than the hardness of mild steel substrate before deposition. This is

evident that the hardness of mild steel was modified to be higher even with only low deposition of Co-Ni-Fe particles. The high hardness value for a 90-minute coating sample may be related to the small crystallite structure and compact arrangement of the particle as has been observed from the coating surface morphology in Figure 3. This has resulted in lesser void formation on the surface thus improving the hardness of the coating (Brenner, 1963). Moreover, it is expected that the number of deposited layers of the particles increases as the deposition time increases, hence raising the hardness of the coating. The conformance of deposition time and coating thickness has been widely discussed in several works of literature (Bakhit and Akbari, 2012, Genova et al., 2019)
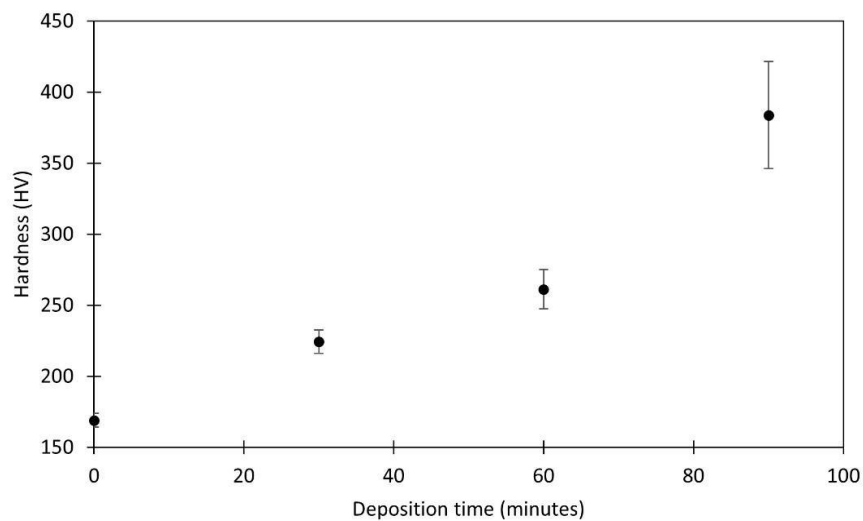

Figure 6: Hardness of Ni-Co-Fe coatings deposited for different 30 to 90 minutes.

The corrosion behaviour of Co-Ni-Fe coatings was tested by using a corrosion dip test. Using this method, the change of weight of the sample after test is measured to assess the damage. Figure 7 summarised the total weight loss of each coating after the dip test. The measured weight loss of samples represents the total loss of materials due to corrosion during the dip test. According to Figure 7, corrosion rate (weight loss) decreases with respect to deposition time of coating. The effect of corrosion on the coating has also been observed from the surface of each sample as shown in Figure 8. From the observation, it can be clearly seen that the bare substrate which is indicated as deposition zero (0) in Figure 8(a) has turned to a completely black colour after the test. Coating deposited for 30 minutes in Figure 8(b) shows some corrosion spots which are believed to have started from the point where no coating was deposited at the top centre of the sample. In this work, all samples have uncoated point at the top centre of the sample which is the point of clipping to hold the sample during the deposition process. As expected, coatings deposited for 60 and 90 minutes show less corrosion reaction compared to others. The corrosion reaction may have started at voids where the substrate was exposed before spreading it to the other area of the sample. The corrosion resistance capability of Co-Ni-Fe coating deposited for 90 minutes increased by 48% compared to the bare substrate and 32% higher than the 30-minute deposited coating.
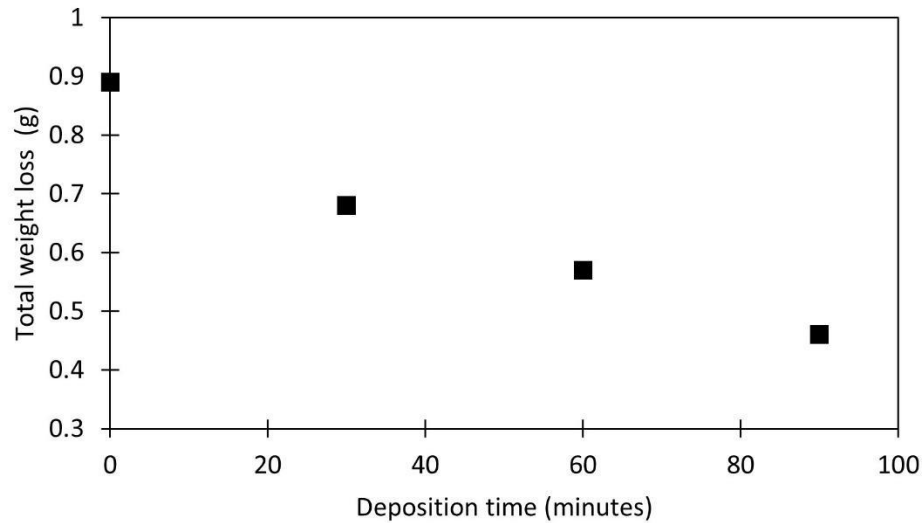
Figure 7: Total weight loss of Co-Ni-Fe coatings after corrosion dip test as a function of coating deposition time.
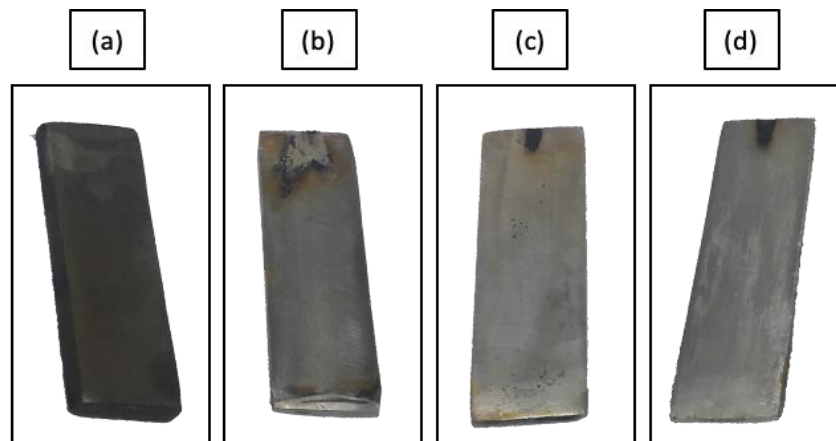

Figure 8: Observation after dip test of samples of Co-Ni-Fe coatings deposited for (a) 0 minutes, (b) 30 minutes, (c) 60 minutes and (d) 90 minutes.

The erosive wear behaviour of the coatings was analysed using a slurry erosion wear tester. The rotation speed was fixed at 1200 rpm and was run for straight and constant eight (8) hours. In this test, beach sands with particle size range of 125- 250 μm were used in the slurry. Figure 9 shows the results of progressive mass loss measured during test time at 120, 240, 360 and 480 minutes of Co-Ni-Fe coatings deposited for 30 to 90 minutes. All coatings showed evidence of erosion wear with the removal of approximately 3 – 15% of coating throughout the eight (8) hour test. At a lower test duration of 120 minutes, coating deposited for 30 minutes had low weight loss, even slightly lower than coating deposited for 60 minutes before it abruptly increased with prolonged test duration. The coating deposited for 90 minutes had the lowest and slowest weight loss percentage rate of all throughout the test. The rate of weight loss for coating deposited for 60

minutes is almost the same as 90 minutes coating but with a higher percentage. On the contrary, a significant weight loss rate was recorded for coating deposited for 30 minutes. This can be associated with the existence of voids in coating deposited for 30 minutes that elevated the erosion process. Weight loss after eight (8) hours for coating deposited 90 minutes showed a significant improvement of wear performance by approximately 78% compared to 30 minutes deposition.
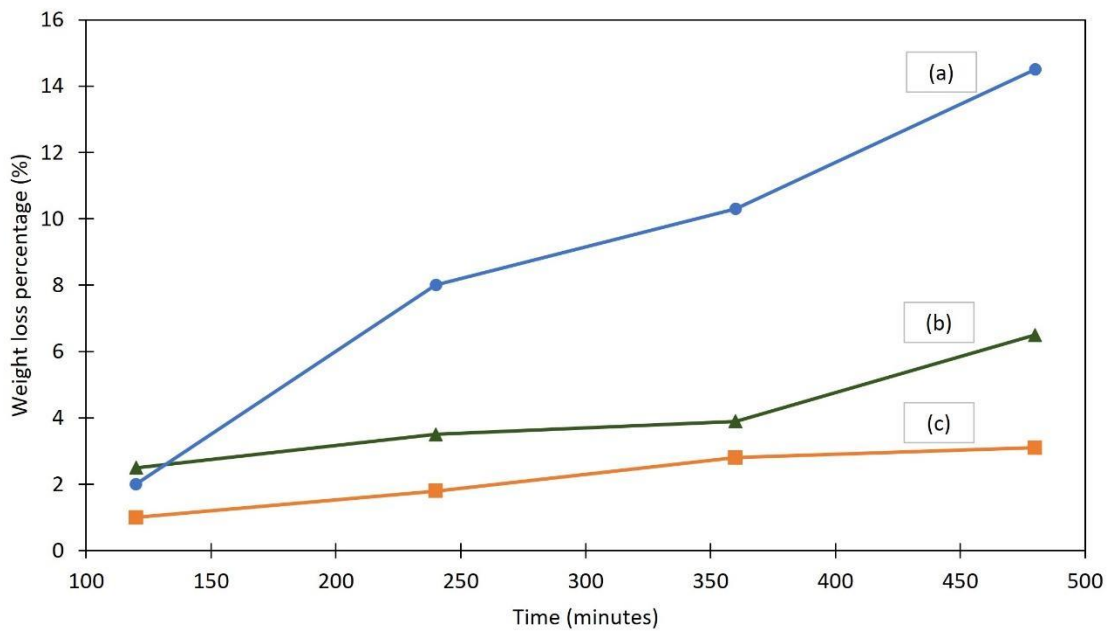

Figure 9: Weight loss percentage of Co-Ni-Fe coatings deposited for (a) 30 minutes, (b) 60 minutes and (c) 90 minutes after slurry erosion test.

The macrographs of the samples and their magnified wear areas after the slurry test are shown in Figure 10. The colour and saturation of the magnified areas have been adjusted to distinctly show the wear areas. From the observation, wear on coatings deposited for 30 and 60 minutes are rather visible. Formation of craters and pits; and flaking and peeling of the coating were apparent and marked as red dashed boxes in Figure 10 (a and b). Nonetheless, as have been measured in Figure 9, more than 80% of the Co-Ni-Fe coatings remained intact on the surface even after the eight (8) hour test.
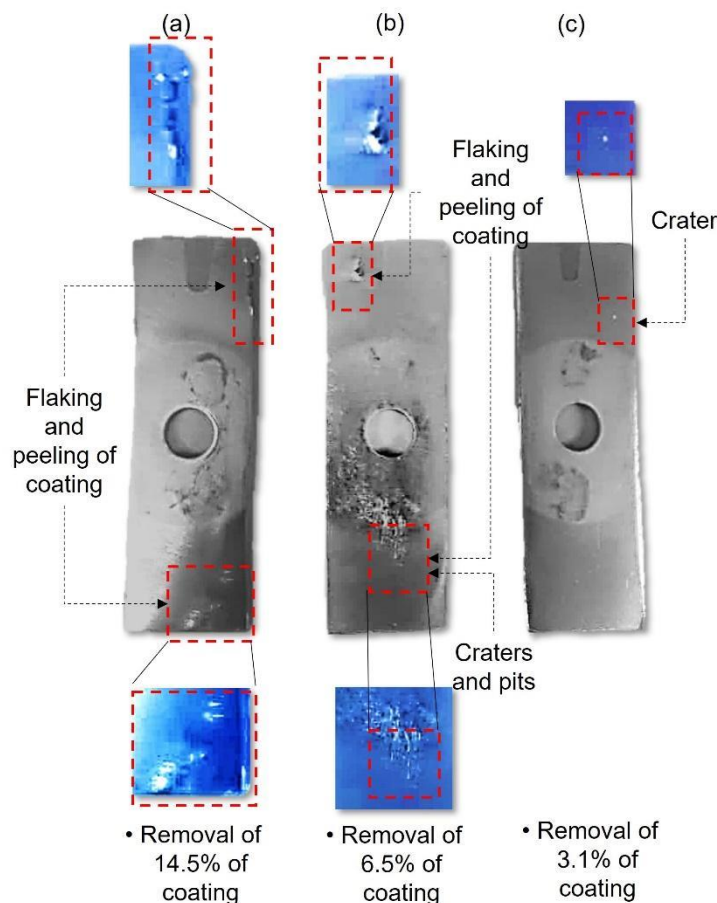
Figure 10: Macrographs of Co-Ni-Fe coatings after slurry erosion test deposited for (a) 30 minutes, (b) 60 minutes and (c) 90 minutes.

The correlation between hardness and surface roughness of coatings after deposition, with total weight loss of all coatings after eight (8) hours of slurry erosion are presented in Figure 11. Coating deposited for 30 minutes recorded the highest weight loss of 0.12 g, followed by coatings deposited for 60 minutes of 0.08 g and the lowest of coating deposited for 90 minutes of 0.04 g. A comparison of bare mild steel with a total weight loss of 0.14 g has been reported earlier by Masdek and group (Masdek et el., 2021). The slurry test was conducted at a relatively lower speed of 700 rpm for eight (8) hours. It can therefore be predicted that the damage may double if the same speed of 1200 rpm is used in this work. Direct correlation between weight loss (erosion wear) and the hardness of coatings can be seen in Figure 11 as dashed line. Numerous works have also reported a strong relationship between high hardness and good wear resistance (Idriss et al., 2021, Mousavi et al., 2020). High hardness of coating has lessened the impact energy of the slurry sand from penetrating into the surface thus improving the erosion wear resistance. While the effect of surface roughness on weight loss due to erosion wear is illustrated as a black line in Figure 11. The weight loss of 60-minute coating was approximately two (2) times than the coating

deposited for 90 minutes with both coatings having almost similar surface roughness of 1.80 - 1.90 μm. The weight loss was further increased to about 0.12 g for coating deposited for 30 minutes that have a profoundly higher surface roughness of ~3.77 μm. These results suggest that surface roughness may have a great influence when the surface roughness is relatively higher than 2.00 μm but may not substantially contribute to the erosion wear when the surface roughness is lower.
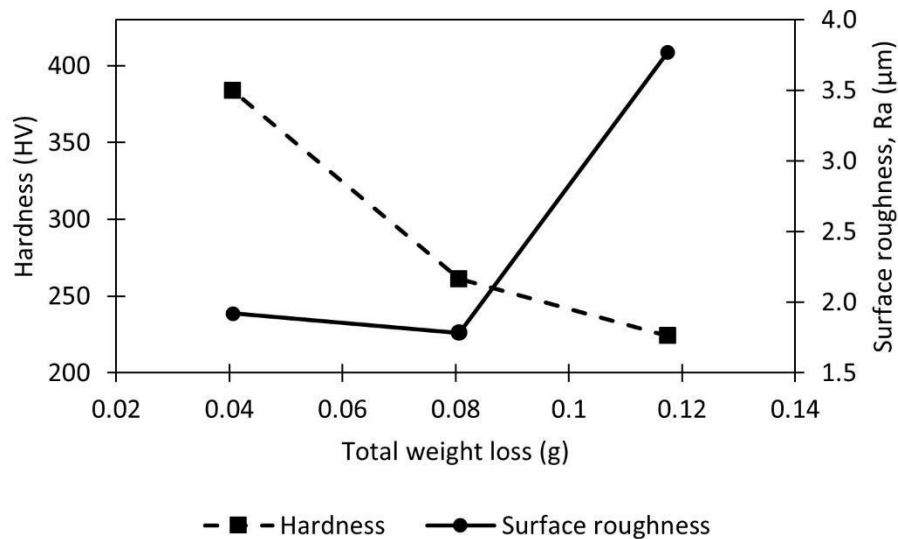

Figure 11. Relationship between hardness and surface roughness with total weight loss after eight (8) hours of slurry erosion.

CONCLUSIONS

Co-Ni-Fe coatings were deposited onto mild steel using the electrodeposition method. The deposition time was varied between 30 to 90 minutes. Deposition time tremendously affect the surface morphology of coating. The density and homogeneity of deposited particles improved as deposition time increased. XRD analysis has proven that the crystallite size of Co-Ni-Fe reduced as the deposition time increased to above 60 minutes. The results also explain the high hardness of coating deposited for 90 minutes at 383 HV, ~71% higher than the hardness of coating deposited for 30 minutes. Coating deposited for 90 minutes also possesses the highest corrosion resistance performance, 48% higher than the bare mild steel substrate. The erosion wear performance of coating at the 8-hour test also improved significantly by 78% when deposition time was increased from 30 minutes to 90 minutes. The high hardness of coating provides better erosive wear resistance behaviour, while surface roughness have lesser influence when the value is lower than 2.0 μm. These results demonstrate an improvement of corrosion and erosive wear behaviour of Co-Ni-Fe coating with deposition time thus may be able to expand the lifespan of surface in extreme conditions like the oil and gas industry.

REFERENCES
Bakhit, B., & Akbari, A. (2012). Effect of particle size and co-deposition technique on hardness and corrosion properties of Ni–Co/SiC composite coatings. Surface and Coatings Technology, 206(23), 4964-4975.
Brenner, A. (1963). Electrodeposition of Alloys. Academic Press New York.
Cançado, L.G., Takai, K., Enoki, T., Endo, M., Kim, Y.A., Mizusaki, H., Jorio, A., Coelho, L.N., Magalhaes-Paniago, R. and Pimenta, M.A. (2006). General equation for the determination of the crystallite size L a of nanographite by Raman spectroscopy. Applied Physics Letters, 88(16), 163106.
Dao, M., Lu, L., Asaro, R. J., De Hosson, J. T. M., & Ma, E. (2007). Toward a quantitative understanding of mechanical behavior of nanocrystalline metals. Acta Materialia, 55(12), 4041-4065.
G. Fortas, S. Sam, Z. Fekih, N. Gabouze. (2009). Electrodeposition of CoNiFe alloys on n-type Silicon. Materials Science Forum, 609, 207-212.
Genova, V., Paglia, L., Marra, F., Bartuli, C., & Pulci, G. (2019). Pure thick nickel coating obtained by electroless plating: surface characterization and wetting properties. Surface and Coatings Technology, 357, 595-603.
Herojit Singh, L., Govindaraj, R., Ravishankar, C., Rajagopalan, S., & Amarendra, G. (2016). Atomic scale understanding of magnetic properties in Ni50Fe35Co15. AIP Advances, 6(2), 025005.
Hyie, K., Zabri, M., Nik Roseley, N., & Nik Mohd Masdek, N. (2016). Effect of deposition time on wear and corrosion performance of Co–Ni–Fe alloy coated mild steel. Journal of Materials Research, 31(13), 1848-1856.
Idriss, A. N. M., Kasolang, S., Maleque, M. A., Nasir, R. M., & Tebal, N. (2021). An overview on the importance of surface modification by TIG and lasers incorporating carbides and their relations to wear behaviours. Jurnal Tribologi, 29, 96-116.
J.O'M. Bockris and A. Damjanovic. (1964). The Mechanism of Electrodeposition of Metals in Metals Aspects of Electrochemistry. London: Butterworths Publishing, 3, 224-346.
Liu, C., Wei, D., Xu, R., Mai, Y., Zhang, L., & Jie, X. (2020). Electroplated Co-Ni/WS2 composite coating with excellent tribological and anticorrosion performance. Tribology Transactions, 63(5), 857-866.
Mahalingam, T., Sundaram, K., Velumani, S., Raja, M., Thanikaikarasan, S., Kim, Y. D., & Asomoza, R. (2009). Effect of pH on Composition, Structure and Magnetic Properties of Electrodeposited Co-Ni Alloys. Advanced Materials Research, 68, p. 52-59.
Masdek, N. R. N., Farhana, N. S., Hyie, K. M., & Baharin, Z. A. K. (2021, March). Surface Characterization of Uncoated and CoNiFe Coated Mild Steel. In Recent Trends in Manufacturing and Materials Towards Industry 4.0: Selected Articles from iM3F 2020, Malaysia (pp. 463-469).
Mousavi, R., Deflorian, F., & Bahrololoom, M. E. (2020). Morphology, hardness, and wear properties of Ni-base composite coating containing Al particle. Coatings, 10(4), 346.

Prasad, A. R., Kunyankandy, A., & Joseph, A. (2020). Corrosion Inhibition in Oil and Gas Industry: Economic Considerations. Corrosion Inhibitors in the Oil and Gas Industry, 135-150.

S. H. Lee and M. G. So, (2000). Effects of deposition temperature and pressure of the surface roughness and the grain size of polycrystalline Si1–xGex films, Journal of Materials science, 35, p. 4789 – 4794.

Salvadori, M.C., D.R. Martins, and M. Cattani. (2006). DLC coating roughness as a function of film thickness. Surface & Coatings Technology, 200(16-17): p. 5119-5122.

Sarkar, S., Baranwal, R. K., Lamichaney, S., De, J., & Majumdar, G. (2018). Optimization of electroless Ni-Co-P coating with hardness as response parameter: a computational approach. Jurnal Tribologi, 18, 81-96.

Simons, M. R. (2008). Report of offshore technology conference (OTC) presentation. NACE International oil and gas production.

Tjong, S. C., & Chen, H. (2004). Nanocrystalline materials and coatings. Materials Science and Engineering: R: Reports, 45(1), 1-88.

Torabinejad, V., Aliofkhazraei, M., Sabour Rouhaghdam, A., & Allahyarzadeh, M. H. (2017). Tribological behavior of electrodeposited Ni-Fe multilayer coating. Tribology Transactions, 60(5), 923-931.

Valdez, B. (2006). Corrosion Tests and Standards: Applications and Interpretation. Corrosion, 62(3), 280.

Y.Marita and I.I Yaacob. (2010). Synthesis and Characterization of Nickel-Iron Silicon Nitride Nanocomposite. Advanced Material Research, 97- 101, 1360-1363.

Zavareh, M. A., Sarhan, A. A. D. M., Razak, B. B. A., & Basirun, W. J. (2014). Plasma thermal spray of ceramic oxide coating on carbon steel with enhanced wear and corrosion resistance for oil and gas applications. Ceramics International, 40(9), 14267-14277