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Optimization of Ni–Mo/Al composite coating parameters using Taguchi method

R Mousavi[1,3], **M Esmailzadeh**[2], **M E Bahrololoom**[3] **and F Deflorian**[4]

[1] Department of Materials Science and Engineering, Faculty of Engineering, Shahid Chamran University of Ahvaz, Ahvaz, Iran
[2] Department of Mechanical Engineering, Persian Gulf University, Bushehr, Iran
[3] Department of Materials Science and Engineering, Shiraz University, Shiraz, Iran
[4] Department of Industrial Engineering, University of Trento, via Sommarive 9, Trento, Italy

E-mail: m.esmaeilzade@pgu.ac.ir

Keywords: Ni–Mo/Al composite coating, Taguchi method, L18 orthogonal array, wear

Abstract

In this article, the parameters of Ni–Mo/Al composite coating electroplating were optimized using Taguchi method and L18 orthogonal array. The impact of annealing temperature, annealing time, electroplating current density, stirring rate of solution and Al content in the bath on the mechanical properties of Ni–Mo/Al coatings were surveyed. It was found that among all parameters, annealing at 600 °C, annealing time of 2 h, 5 g l^{-1} Al content in the bath, current density of 30 mA cm^{-2} and stirring rate of 500 rpm can produce Ni–Mo/Al composite coating with better wear properties. In addition, between different factors, electroplating current density and annealing time are the most and least effective on the wear behavior, respectively. Archard's law was established between wear and hardness for this coating in various parameters. In the final, the wear mechanism of the optimal and non-optimal composite coatings was examined. The adhesive wear was the overcoming mechanism in the optimal composite coating.

Nomenclature

DOE	Design of Experiments
CTAB	Cetyl Trimethylammonium Bromide
SDS	Sodium Dodecyl Sulfate
SAE	Society of Automotive Engineers
XRD	x-ray Diffraction
SEM	Scanning Electron Microscope
SN Ratio	Signal to Noise Ratio
SB	Smaller is Better
HB	Higher is Better
NB	Nominal is Better

1. Introduction

A coating with suitable properties can be obtained during electro-codeposition by handling the chemical and operating parameters. The following are some chemical and operating parameters which can be mentioned: (1) current density, (2) pH, (3) temperature, (4) chemical composition of the bath solution, (5) additives, (6) stirring of the solution, and (7) the amount of particles in the solution. When the effects of more than two parameters are simultaneously investigated on a property using conventional experimental methods in which one factor varies while the others are fixed at constant levels, an interaction may appear as a variable [1]. In most experiments, the presence of interaction effects between parameters is usually ignored. In this situation, the effect of each parameter depends on the level of the other parameters and it will be difficult to predict the effect of each parameter directly without considering the effect of other parameters. Hence, the analyses are inefficient. Therefore, to improve deposition conditions and prepare better coatings that fulfill industrial needs, it is important to develop a more scientific approach to obtain optimum conditions. In order to overcome this issue the design of experiments (DOE) may be used [2].

DOE is a collection of mathematical and statistical techniques useful for modeling, analyzing, improving, developing, and optimizing processes that can be used to assess the relative influence of several independent parameters even in the presence of complex interactions [1–3]. It is an important tool used for the study of the influence of various parameters involved in a process on the output [4–11]. The output is termed as 'response variable' or quality characteristic and the inputs are termed as 'process variables or parameters'.

The strategies of DOE such as parameter optimization and Taguchi analysis have rarely been used for electrodeposited composite coatings [12, 13]. According to the Taguchi method, a large number of parameters can be investigated with a small number of experiments using orthogonal array. In order to obtain the best performance of the process, Taguchi method is used which is a time-saving and cost-effective technique useful in finding the optimum process control parameters. Many researchers have found and used Taguchi method as a useful technique in dealing with responses influenced by several variables [6, 8, 10, 13–15]. Baradeswaran *et al* used the Taguchi method for process optimization and detection of the optimal combination of the parameters for wear behavior of Al–Al_2O_3 composites as a given response [6]. Their aim was to find the minimum wear rate under various applied load and sliding distance conditions. They found out that the wear mass loss at 10 N applied load at 400 m sliding distance is the optimum combination for an optimum result. According to the experimental results and Taguchi analysis, the minimum wear mass loss happened at 6 wt% of Al_2O_3 as compared to other composites and base alloy. Rout *et al* [10] indicated that Taguchi method enable to optimize parameters to minimization of erosion rate. Gadhari *et al* [11] utilized Taguchi technique to obtain optimal parameters of Ni–P–TiO_2 composite coating process. They reported that annealing temperature has the most effect on mechanical properties of composite coating. Jegan *et al* [12] employed Taguchi's L27 orthogonal array to increase hardness of Nickel/nano-Al_2O_3 composite coatings. They concluded that the duty cycle had most affecting, while the current density had the least effect. Kalyan Das *et al* optimized the three coating parameters include bath temperature, concentration of reducing agent and concentration of nickel source together with the annealing temperature for electroless Ni–B coating. They reported that concentration of reducing agent has the most affecting on the wear resistance of electroless Ni–B coating. Aruna *et al* [13] studied the influence of various parameters on the area fraction of Ni-composite coatings containing ceramic particles, microhardness and thickness of coating by Taguchi method and analysis of variance.

Among different coatings, it has been reported that Ni–Mo–Al ternary alloy can be useful in some applications such as high sulfidation condition [16, 17] and high wear rate [18–20]. It was demonstrated that in the presence of a given Mo content, by adding Al element, the sulfidation rate of Ni–Mo–Al ternary alloys is decreased [16]. Although the introduction of Al into superalloys of the Ni–Mo system has a negative effect on the heat resistance of the alloy at high temperatures [21]. On the other hand, the Ni_3Al phase [21–24] and Ni_3Mo can be formed [21, 25, 26] at the temperature of 800 °C and below. As a consequence, coatings in the presence of Al and Mo elements can contain Ni_3Al, Ni_3Mo and other intermetallic compounds which improve their wear resistance [23].

Ni–Mo–Al ternary alloy in all the references mentioned in the last paragraph, was prepared via arc-melting in an argon atmosphere from elemental metals [17] and plasma and flame spraying [20]. In our previous work, for the first time, the Ni–Mo/Al composite coating was prepared via electrodeposition methods [27]. It was demonstrated that a crack-free coating and high efficiency can be obtained at the electroplating temperature of 40 °C. Consequently, in this work, the electroplating temperature was considered to be constant. As reviewed in the previous paragraph, high temperature during the preparation of the Ni–Mo–Al ternary alloys was the key factor in the production of intermetallic compounds. Consequently, in addition to the mentioned electroplating parameters in the initial sections, the effects of annealing time and temperature were considered as two operating parameters on the wear and hardness properties of the coating.

In order to obtain a Ni–Mo/Al composite coating with high wear resistance, orthogonal array of 18 which is called L18 in Taguchi method was used. This orthogonal array is useful for determining the main effect of the parameters. It is clear that the composition of coatings is mainly depend on the other operating parameters like temperature, pH, stirring rate of bath, chemical composition of bath solution, and so on. On the other hand, the chemical composition of coating is altered due to changing the operating parameters and it is not necessary to consider as an independent parameter. It means that the effect of chemical composition was indirectly considered. On the other hand, by using L18 the interaction between the parameters is removed. In the present

Table 1. Operating parameters and their levels.

Parameter	Levels
Current density (mA cm^{-2})	15, 30, 50
Stirring rate (rpm)	350, 500, 650
Al content (g l^{-1})	1, 5, 10
Annealing temperature (°C)	300, 600
Annealing time (hr)	1, 2, 3
Trisodium citrate (M)	0.15
Nickel sulphate (M)	0.5
Sodium molybdate (M)	0.01
CTAB (g l^{-1})	0.3
SDS (g l^{-1})	0.5
pH	4

work, to perform the Taguchi analysis, the statistical analysis software MINITAB 16 was used for the design and analysis of the experiments.

2. Materials and methods

2.1. Preparation of Ni–Mo/Al composite coating and tests

All chemical materials and electroplating conditions are prepared in accordance with previous work [27]. The difference is that electroplating parameters were considered at different levels. The parameters of the experiments are listed in table 1. For all trials, the time of electroplating was considered constant at 2 h.

Electrodeposition of Ni–Mo/Al composite coatings was carried out in a 250 ml glass beaker. Before each coating, the electrolyte was homogenized by magnetic stirrer for 12 h. In all conditions, a carbon steel plate (SAE 1008, 4 cm * 15 cm) was used as substrate and cathode in elctrodeposition bath. In contrast, a pure Ni plate was used as anode.

After coating process, XRD and microhardness tests were performed to ensure the formation of the desired composite coating and the study of the effect of coating parameters. In forward, a pin on disk wear test was done to determine the wear resistance of composite coating specimens. The counter body was been a chromium coated steel pin with 6 mm diameter. The wear tests were applied in two sliding distances include 40 m and 120 m and five pressure loads include 200 gf, 300 gf, 400 gf, 500 gf and 600 gf. After wear test, the worn surfaces were studied to specify mechanism of wear by scanning electron microscope (SEM, Cambridge S360 model) micrograph.

2.2. Steps in the Taguchi Method

Figure 1 shows the flowchart of Taguchi method. According figure 1, the integrated approach consists of several steps whose shows and descriptions are as follows.

(1) Select the response variable. Selecting the response variables is the initial step in the Taguchi method. In this study, wear rate and microhardness were selected as response variables. The effect of parameters on the response variables can be obtained by Taguchi method.

(2) Handpick the parameters and their levels. The effective parameters participating in the synthesis of electrodeposited composite coatings were selected. These operating parameters including annealing time, Al content in the solution, current density, and stirring rate were selected at three different levels. Annealing temperature was also selected as an effective parameter on the mechanical property at the two levels of 300 and 600 °C. The details are specified in table 2.

(3) Select the orthogonal array. Selecting an orthogonal array is the third step in Taguchi method. For this purpose, the Taguchi orthogonal array L18 (see table 2) was proposed to categorize the critical level of parameters. After selecting orthogonal array L18, annealing temperature was assigned to the 1st column and the other parameters were assigned to the 2nd to 5th columns (see table 2).

(4) Perform the experiments. The total numbers of experiments run to complete the test was 18.

(5) Examine the data and decide optimal parameter levels. Optimization and obtaining the highest hardness and wear resistance were the main goals of this study. With the Taguchi method, the criteria of SN ratios were used according to table 3 to determine the optimal value for each operating parameter.

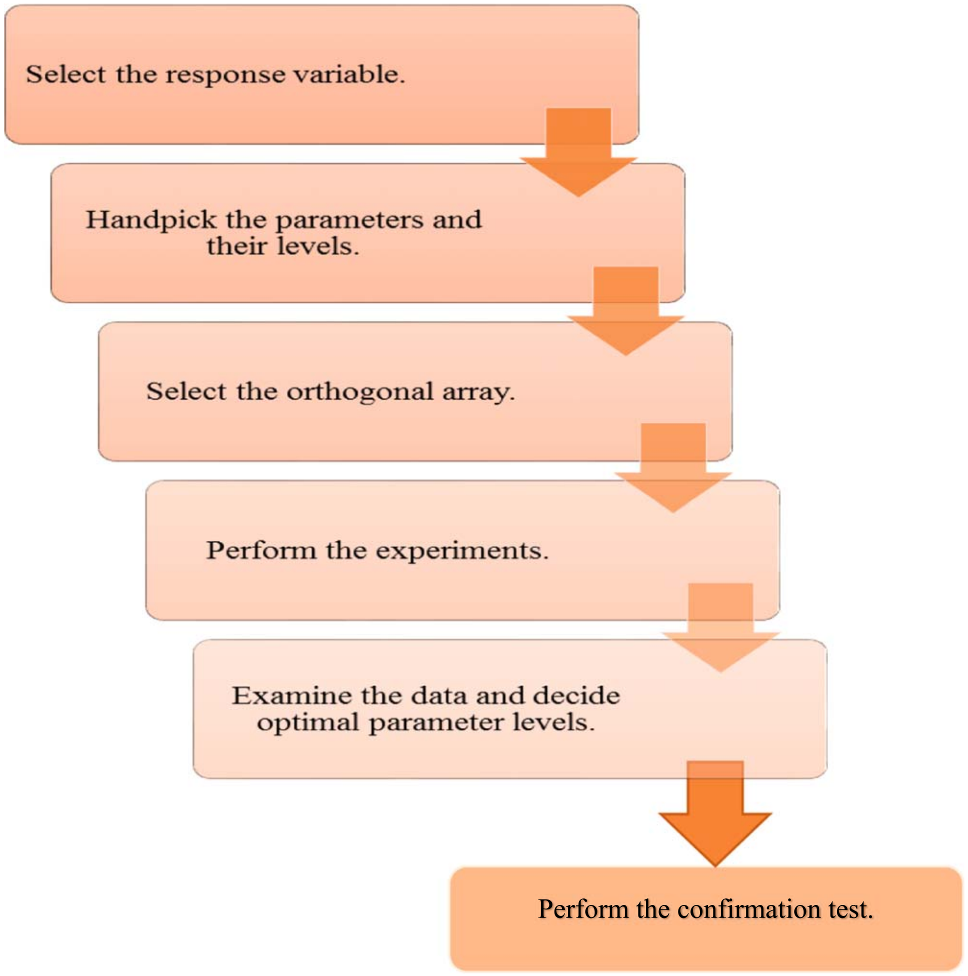

Figure 1. The flowchart of Taguchi method.

Table 2. L18 orthogonal array and the parameters.

Trials	Annealing temperature (°C)	Annealing time (hr)	Al content (g l^{-1})	Current density (mA cm^{-2})	Stirring rate (rpm)
1	300	1	1	15	350
2	300	1	5	30	500
3	300	1	10	50	650
4	300	2	1	15	350
5	300	2	5	30	650
6	300	2	10	50	350
7	300	3	1	30	350
8	300	3	5	50	500
9	300	3	10	15	650
10	600	1	1	50	650
11	600	1	5	15	350
12	600	1	10	30	500
13	600	2	1	30	650
14	600	2	5	50	350
15	600	2	10	15	500
16	600	3	1	50	500
17	600	3	5	15	650
18	600	3	10	30	350

Table 3. The outputs and their SN ratio.

Output	Signal to noise ratio
Wear rate (weight loss)	Smaller is better
Hardness (VHN)	Higher is better

(6) Perform the confirmation test. The validation of the experiment and the results from optimal deposition parameters is the final step in the Taguchi method to attain the target value of the output.

2.3. Signal-to-noise ratio (SN ratio)

The performance of the process response can be measured by signal to noise (SN) ratio proposed by Taguchi. The SN ratio is the ratio of mean to standard deviation and can effectively consider the variations encountered in a set of trials. Depending on the characteristic of the response, SN ratio can be classified according to three criteria: smaller is better (SB), higher is better (HB) and nominal is better (NB). No matter which criteria are utilized for SN ratio, the level with maximum value can be regarded as the optimal level for each parameter. The response variable is the 'output' of the experimental model. The present study tries to minimize the weight loss during wear test and maximize hardness of Ni–Mo/Al composite coating. Hence, these two outputs are taken as the response variables and for the first 'the smaller is better (SB)' is appropriated to the SN ratio and for the second one 'the higher is better (HB)' is appropriated to the SN ratio (table 3). Based on different requirements, Taguchi *et al* suggested three equations for the SN ratio as follows [1]:

For 'the smaller is better':

$$SN_{SB} = -10 \times \log_{10}\left[\frac{1}{n}\sum_{i=1}^{n} y_i^2\right] \tag{2.7}$$

For 'the higher is better':

$$SN_{HB} = -10 \times \log_{10}\left[\frac{1}{n}\sum_{i=1}^{n} \frac{1}{y_i^2}\right] \tag{2.8}$$

For 'the nominal is better':

$$SN_{NB} = -10 \times \log_{10}\left[\frac{\frac{1}{n}(V_m - V_e)}{V_e}\right] \tag{2.9}$$

Where y is the response and n is the number of tests in a level combination, $V_m = \frac{1}{n}(\sum_{i=1}^{n} y_i)^2$, and $V_e = \frac{1}{n-1}\sum_{i=1}^{n}(y_i - \bar{y})^2$.

2.4. Standard orthogonal arrays, input parameters, and their levels

Taguchi recommends that experiments are designed with an emphasis on the main effects as much as possible [1]. Accordingly, the so-called L18 orthogonal array involving eighteen experiments was chosen. Using this design, the influence of up to eight parameters can be analyzed. One parameter must be set at two levels and the other at three levels.

The studied factors and their levels are detailed in table 2. Each row represents one trial. The first row is for column labels and each column represents one variable [2, 28].

3. Results and discussion

3.1. Design of the experiments (DOE)

The trend of chemical composition changes of 18 trials presented in table 4. In addition, figure 2 shows graphically two trends of each 18 trials. In this figure, top curves and low curves are deposited Al and Mo, respectively. It can be seen, the content of deposited Al is truly higher than deposited Mo in all trials. Also, changing the contents of deposited Al and Mo are the opposite in some trials such as trial 18.

The EDS results are related to two coatings prepared for the tests. The EDS analysis was repeated three times for each sample. As seen, the trend is very similar and repeatable for the two coats. The data related to the impacts of the five variable parameters of annealing temperature, annealing time, Al content in the bath solution, current density, and stirring rate on the wear and microhardness are also presented in table 4.

Table 4. The orthogonal array of L18 ($2^1 \times 3^4$) for the experiments.

Trials	Ni (wt%)	Mo (wt%)	Al (wt%)	WR (m^3/N.m)	Micro-hardness (VHN)
1	86	4.8	9.2	0.000 57	551.10
2	75.3	3.3	21.4	0.001 33	357.33
3	63.5	1.6	34.9	0.001 50	337.33
4	86	7.2	6.8	0.000 35	602.67
5	75.8	4.4	19.8	0.000 33	562.00
6	62.1	1.5	36.4	0.018 76	304.00
7	80.4	2.1	17.5	0.000 82	337.67
8	64.8	1.7	33.5	0.000 88	329.00
9	75.5	5.2	19.3	0.000 71	495.67
10	89.9	0.4	9.7	0.000 67	381.67
11	63	3.3	33.7	8E-05	476.67
12	60.9	1.8	37.3	0.000 18	551.67
13	87.1	3.4	9.5	0.000 15	311.00
14	66.1	1.5	32.4	0.000 35	489.67
15	59.5	3.8	36.7	6.24E-05	780.03
16	77.3	3.2	19.5	0.000 52	435.33
17	61.9	6.8	31.3	3.86E-05	773.33
18	60.4	1.9	37.7	0.006 32	538.33

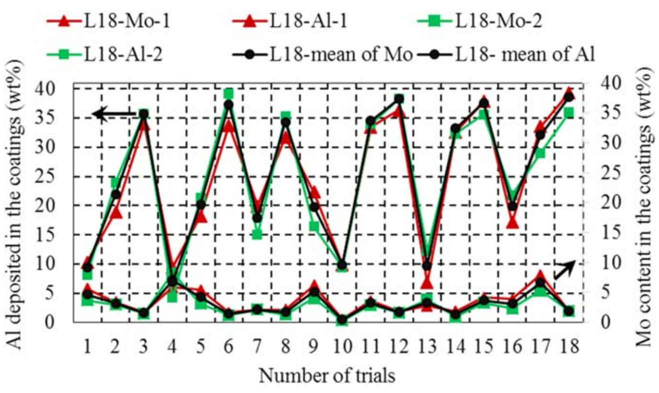

Figure 2. Average chemical composition obtained from Ni–Mo/Al composite coatings for each experimental run (each number on the horizontal axis corresponds to the experiment designation number (first column) and respective experimental conditions listed in tables 4 and 2).

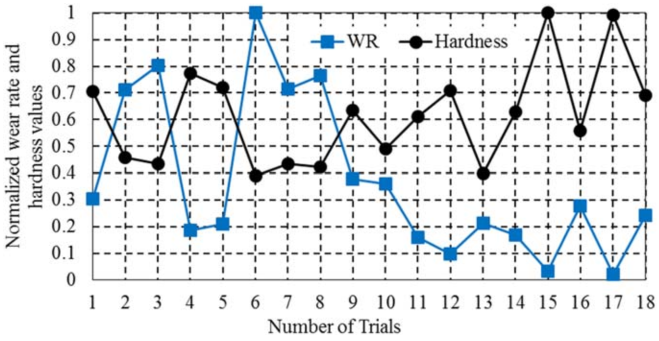

Figure 3. The normalized values of wear rate and microhatrdness obtained from Ni–Mo/Al composite coatings for each experimental run (each number on the horizontal axis corresponds to the experiment designation number (first column) and respective experimental conditions listed in tables 4 and 2).

The trend of wear rate and microhardness changes of 18 trials presented in table 4 is graphically shown in figure 3. It is obvious that they act opposite to each other: when wear rate decreases, microhardness increases. In addition, figure 3 shows that the relationship between wear rate and microhardness obliges the Archad's law for 18 trials. It can be observed that trial 15 and 17 have minimum wear rate and maximum hardness value.

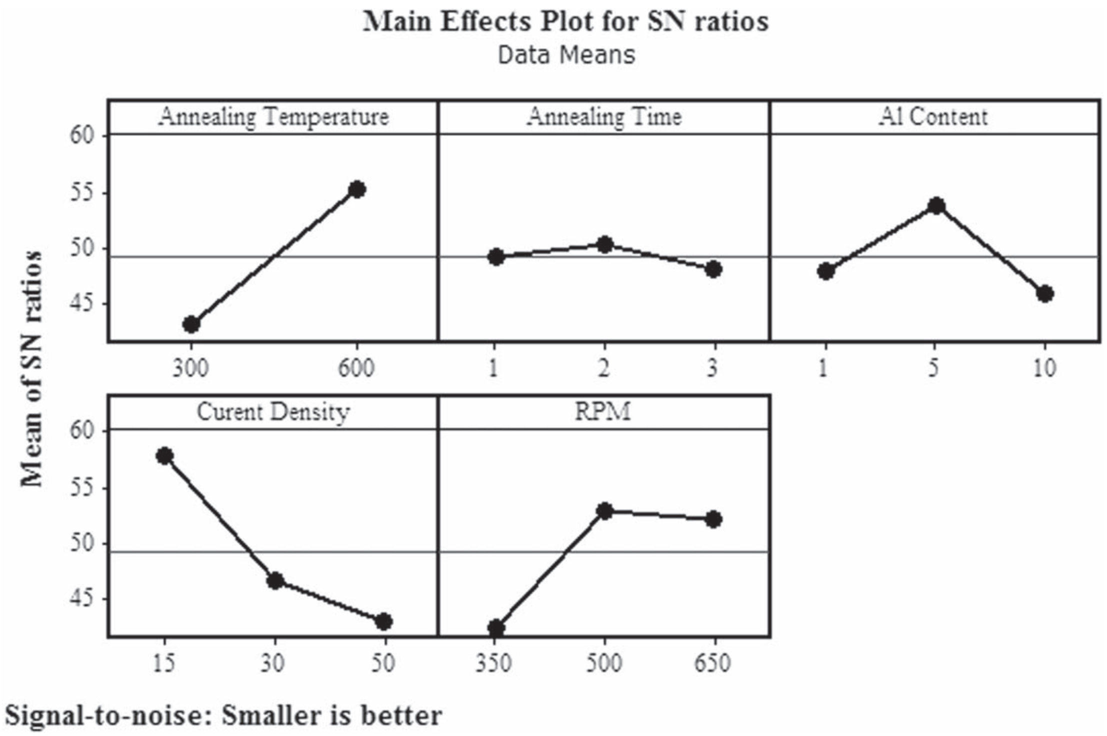

Figure 4. Plots of the main effects of signal to noise ratios for the wear test.

Table 5. Response table for signal to noise ratios for wear test.

Level	Annealing temperature (°C)	Annealing time (hr)	Al content in the bath ($g\ l^{-1}$)	Current density ($mA\ cm^{-2}$)	Stirring rate (rpm)
1	59.78	66.49	66.88	75.21	59.34
2	71.96	67.16	71.26	64.03	68.72
3	—	63.96	59.47	58.37	69.54
Delta	12.18	3.20	11.79	16.84	10.20
Rank	2	5	3	1	4

3.2. Optimization of parameters via Taguchi method

To handpick the optimal parameters from a collection of parameters, orthogonal array of Taguchi is a good candidate [2, 3]. By means of orthogonal array, it is possible to identify the influence of parameters on the response variable and specify the parameters that give the best results for the wear. The results of wear test (WR) presented in table 4, were obtained according to the experimental conditions listed in table 2 for each combination. The results were analyzed for the SN ratio. The SN ratios are different according to the type of output. As the lowest wear rate and/or weight loss of the coatings were required, the 'smaller is better' equation proposed by Taguchi for SN ratio calculation was used [1]. The results of SN ratio effects are shown in figure 4. These plots were used for optimization. Plots for SN ratio were generated using MINITAB 16.

The delta is the highest average SN ratio for each parameter minus the lowest average SN ratio for it. Ranks are allocated based on the delta values: Rank 1 is assigned to the highest delta value, Rank 2 is given to the second highest delta value, and so on.

Among all design parameters, the parameter which has a larger difference in the delta value has a more significant contribution to the response [1]. According to figure 4 and table 5, the maximum–minimum value for the current density has the highest delta value. Hence, the current density is the most significant parameter affecting the wear rate (rank 1) whereas the annealing time has the least effect (rank 5). On the other hand, the parameter for which the line has the largest slope has the most significant effect and vice versa. The rank of other parameters affecting the wear rate can be seen in table 5.

The plot of the main effects gives the optimal combination of coating parameters for minimum friction and wear [15]. Therefore, the level with the highest SN ratio was considered as the optimal value for each parameter. Hence, the optimal deposition parameter levels for obtaining a Ni–Mo/Al composite coating with low weight

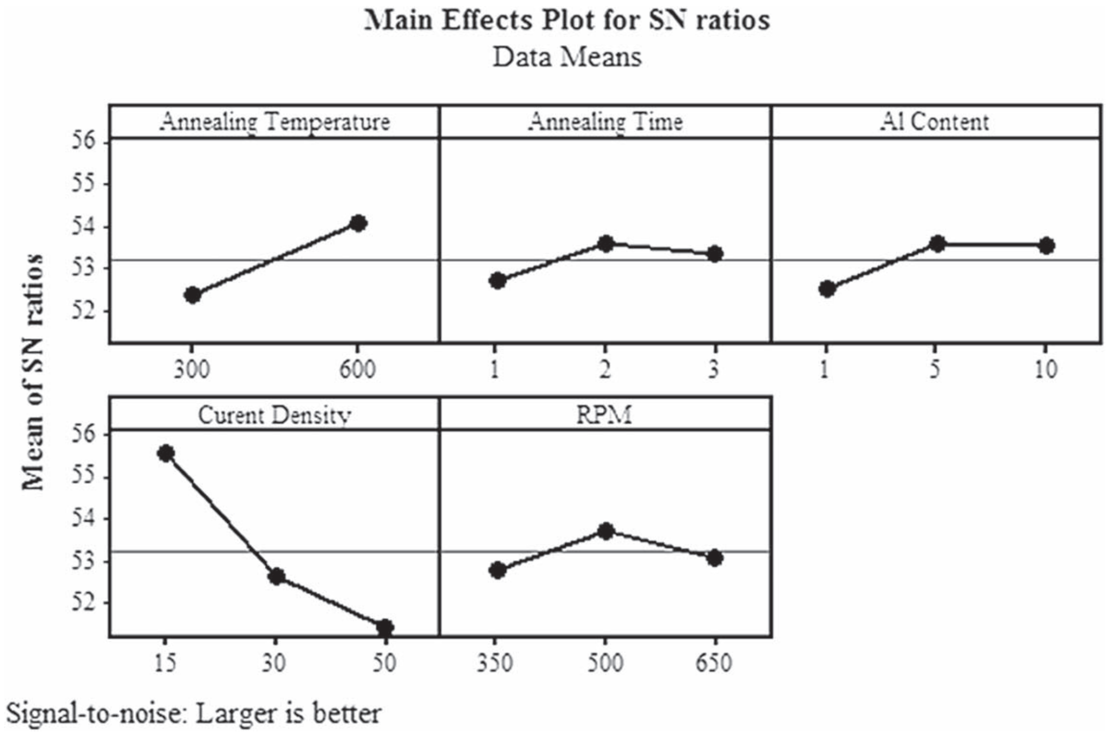

Figure 5. Plots of the main effects of signal to noise ratios for the microhardness test.

Table 6. Response table for signal to noise ratios for microhardness test.

Level	Annealing temperature (°C)	Annealing time (hr)	Al content in the bath ($g\,l^{-1}$)	Current density ($mA\,cm^{-2}$)	Stirring rate (rpm)
1	52.39	52.75	52.54	55.58	52.84
2	54.08	53.62	53.59	52.66	53.74
3	—	53.33	53.57	51.46	53.12
Delta	1.69	0.88	1.04	4.12	0.9
Rank	2	5	3	1	4

loss are annealing temperature of 600 °C (level 2), annealing time of 2 h (level 2), 5 $g\,l^{-1}$ Al content in the bath (level 2), current density of 15 $mA\,cm^{-2}$ (level 1), and stirring rate of 500 rpm (level 2).

According to figure 4, there is little difference between SN ratios of stirring rate at levels 2 and 3. Therefore, the other possibility for optimal condition is the annealing temperature of 600 °C (level 2), annealing time of 2 h (level 2), 5 $g\,l^{-1}$ AlAl content in the bath (level 2), current density of 15 $mA\,cm^{-2}$ (level 1), and stirring rate of 650 rpm (level 3).

The same procedure is equally done for another response, i.e. microhardness for which the results are given below.

As the highest hardness of coatings was required, the 'higher is better' equation proposed by Taguchi for SN ratio calculation was used. The results of the effects on SN ratio are shown in figure 5. These plots were used for optimization.

The delta, rank, and SN ratio values for microhardness are given in table 6. As is seen in figure 5 and table 6, the current density has the highest slope or delta value and is assigned rank 1. Annealing time has the lowest delta value and is assigned rank 5.

As with wear tests, the level with the highest SN ratio was considered as the optimal value for each parameter. Hence, the optimal deposition parameter levels for obtaining a Ni–Mo/Al composite coating with high microhardness are annealing temperature of 600 °C, annealing time of 2 h (level 2), 5 $g\,l^{-1}$ Al content in the bath (level 2), current density of 15 $mA\,cm^{-2}$ (level 1), and stirring rate of 500 rpm (level 2). According to figure 5, there is no difference between SN ratios of Al content in the bath at levels 2 and 3. Therefore, the other possibility for optimal conditions is annealing temperature of 600 °C, annealing time of 2 h (level 2), 10 $g\,l^{-1}$ Al content in the bath (level 3), current density of 15 $mA\,cm^{-2}$ (level 1), and stirring rate of 500 rpm (level 2).

In general, hardness is used as an initial guide to the suitability of coating materials for applications requiring a high degree of wear resistance. According to the reported result in reference [29], the anti-wear performance of the films improved as they hardened. Hence, the hardness of the coating is quite responsible for providing it with the necessary resistance against wear [30].

According to Archard's law [31], which was assigned in equation (3), there is an inverse relationship between hardness and wear loss, and the coating wear decreases as its surface hardness increases, which justifies the outcomes of the reported results in table 4 and figure 4.

$$\Delta V = k.L.S/H(k\text{: wear coefficient}) \tag{3}$$

It is noticeable that there is a group of parameters in which the optimal levels of the parameters for wear and microhardness properties are the same and the optimal conditions in which the highest microhardness and the lowest weight loss are simultaneously obtained are the same. Consequently, and according to Archard's law, the optimal conditions to obtain a coat with the highest wear resistance and hardness are annealing temperature of 600 °C, annealing time of 2 h (level 2), 5 $g\,l^{-1}$ Al content in the bath (level 2), current density of 15 $mA\,cm^{-2}$ (level 1), and stirring rate of 500 rpm (level 2).

The final step is the confirmation experiment in the first iteration of the Taguchi method. The purpose of this experiment is to make a decision about the optimal solution. It was performed by conducting a test with a specific combination of the parameters and levels previously evaluated. According to the Taguchi analysis which is shown in figures 4 and 5, the optimal conditions for each response were obtained and the confirmation test corresponding to the optimal conditions was performed. The weight loss and microhardness values of coatings obtained under optimal and non-optimal conditions are given in table 7.

3.3. Wear mechanism of Ni–Mo/Al composite coating

Figures 6(A), (B) illustrates the friction coefficient variation of the optimal and non-optimal composite coatings as a function of distance. It is very clear that the coating obtained under optimal conditions has a lower friction coefficient than the non-optimal coating.

As is seen, the Ni–Mo/Al composite coating obtained under optimal conditions has more durability than the one obtained under non-optimal conditions. It can be concluded that preparing a coating under optimal conditions produces fewer plowing grooves: this explains the fewer peak fluctuations in the frictional curve of the composite coatings (figure 6(A)—friction coefficient is changed from 0.2 to 0.4). The SEM micrographs of the worn surfaces of the Ni–Mo/Al composite coatings obtained under optimal and non-optimal conditions are presented in figures 7(A)–(E), respectively.

In the SEM images of the wear tracks on the coatings it is discernible that the composite coating obtained under optimal conditions has lower depth and width abrasive grooves compared to the one obtained under non-optimal conditions (figure 7(B) compared with figure 7(E)). As is also seen in figure 7, the width of the worn surface of the composite coating in figure 7(A) is very narrow compared to that in figure 7(D). By comparing the A-E parts of figure 7, it can be observed that the worn scar of the Ni–Mo/Al composite coating obtained under optimal conditions is much smoother than that of the non-optimal Ni–Mo/Al composite coating. Because of lower friction coefficient, this coating show smooth worn surface and lowest weight loss in the wear test. Therefore, the lubricant layer formed on the coating surface during the wear test. One of the SEM micrographs is displayed in figure 7(C) and it can be seen that the load is taken by some of the nodules while the others remain undeformed. The longitudinal grooves along the sliding direction can be clearly observed (see figure 7(B)). Hence, it can be concluded that the abrasive wear is the predominant phenomenon. The presence of cracks indicates that the composite coating obtained under optimal conditions can be harder than the one obtained under non-optimal conditions. The results of the microhardness measurement confirmed that the Ni–Mo/Al composite coating obtained under optimal conditions is harder than the one obtained under non-optimal conditions (table 7).

On the other hand, as shown in figure 6(B), a large variation in friction coefficient (about $0.1 \sim 1$) appeared after 40 m which suggested a change in the wear mechanism and showed a stick-slip behavior. The worn surface of the non-optimal coating indicates the ablation of the surface (by its difference in brightness) suggesting an adhesive wear mechanism (figures 7(D), (E)). According to figure 6(B), it seems that after a distance of about 40 m in the wear test, a severely damaged surface is formed as presented in figure 7(E). The worn surfaces of the non-optimal composite coatings along the sliding direction indicate a greater degree of wear and localized adhesion between the specimen pin surface and the counter body.

From the above results, it can be understood that the optimal coating can bear the wear load without changing the wear mechanism behavior under wear loads up to 400 gf. It was also observed that the friction coefficient could be approximately considered in the range of 0.2–0.4. In order to find the critical wear load in which the wear mechanism was changed and the optimal coating surface was worn, the wear load was increased

Table 7. Confirmation experiments conducted with the results.

		Annealing temperature (A), Annealing time (B), Al content in the bath (C), Current density (D), and Stirring rate (E)		
Response variable		Coating preparation conditions	Values	
Wear rate (m^3/N.m)	Optimized	(A): 600 °C, (B): 2 h, (C): 5 g l^{-1}, (D): 15 mA cm^{-2}, and (E): 500 rpm	Wear load (200 g)	~0
			Wear load (300 g)	~0
			Wear load (400 g)	~0
			Wear load (500 g)	~0
			Wear load (600 g)	0.0009
	Non optimized	(A): 600 °C, (B): 2 h, (C): 5 g l^{-1}, (D): 30 mA cm^{-2}, and (E): 500 rpm	Wear load (200 g)	~0
			Wear load (300 g)	0.0011 (g)
			Wear load (400 g)	0.0012 (g)
Hardness (VHN)	Optimized	(A): 600 °C, (B): 2 h, (C): 5 g l^{-1}, (D): 15 mA cm^{-2}, and (E): 500 rpm	676.7	
	Non optimized	(A): 600 °C, (B): 2 h, (C): 5 g l^{-1}, (D): 30 mA cm^{-2}, and (E): 500 rpm	621.9	

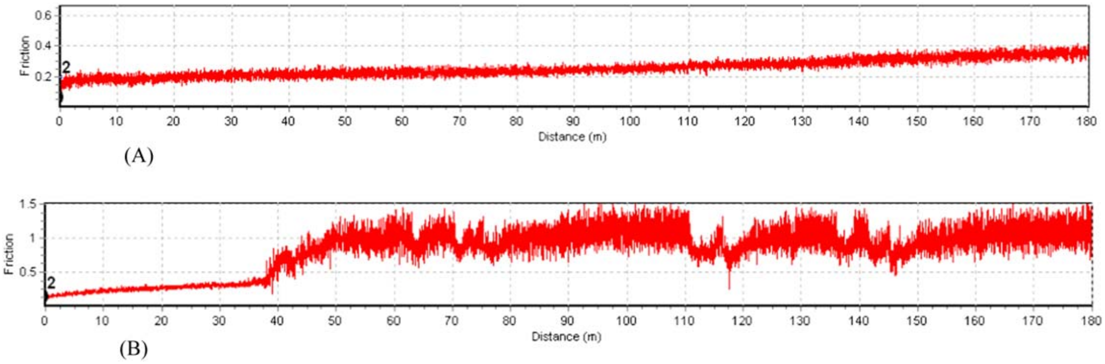

Figure 6. Friction coefficient of coatings deposited under: (A) optimal conditions, and (B) non-optimal conditions (Vertical force: 400 gf and velocity: 0.03 m s^{-1}).

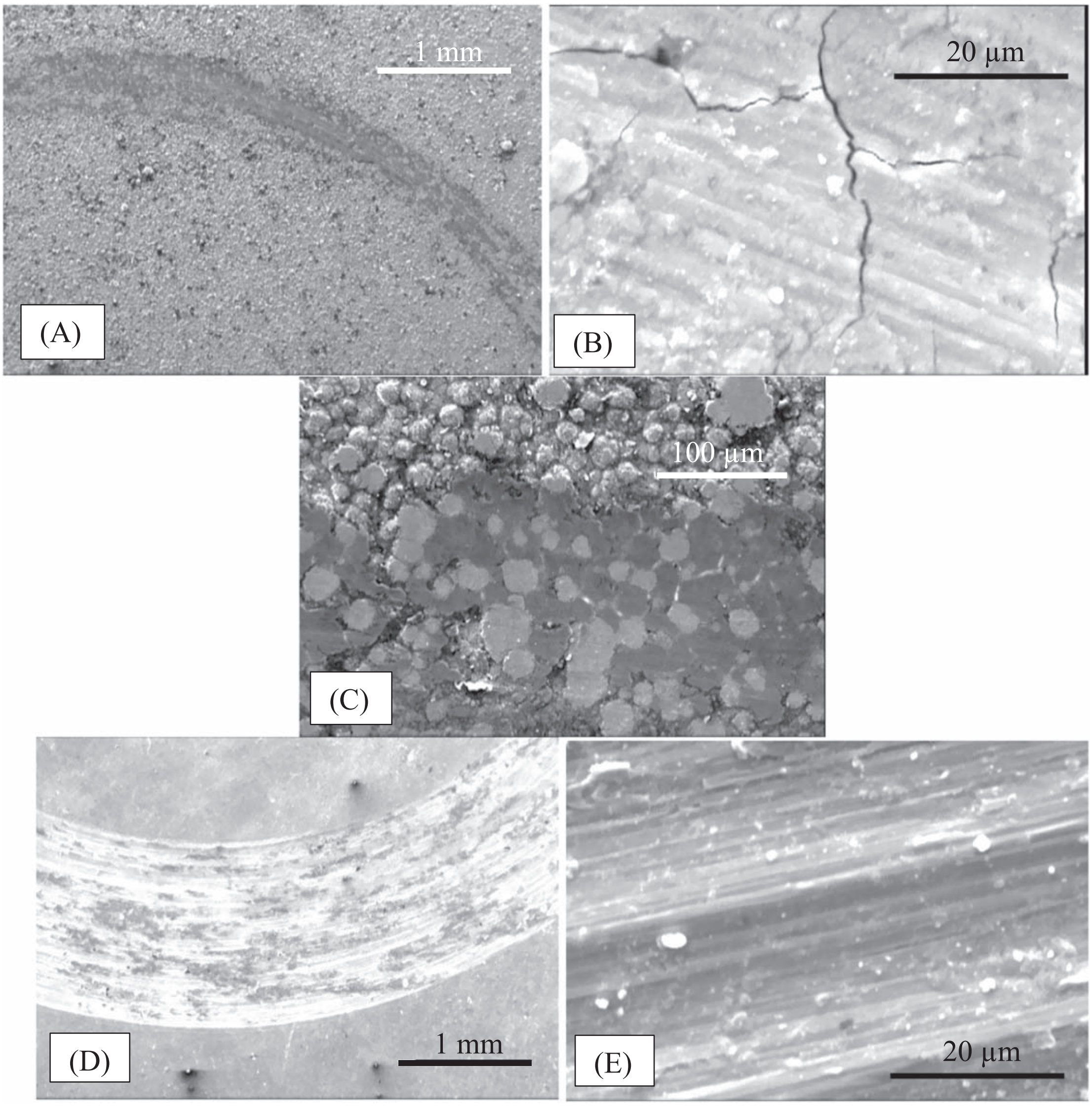

Figure 7. The SEM micrographs of the worn surfaces of the Ni–Mo/Al composite coatings obtained under optimal (A)–(C) and non-optimal conditions (D)–(E) (wear condition was the same for both types of coatings: Vertical force: 400 gf and velocity: 0.03 m s^{-1}).

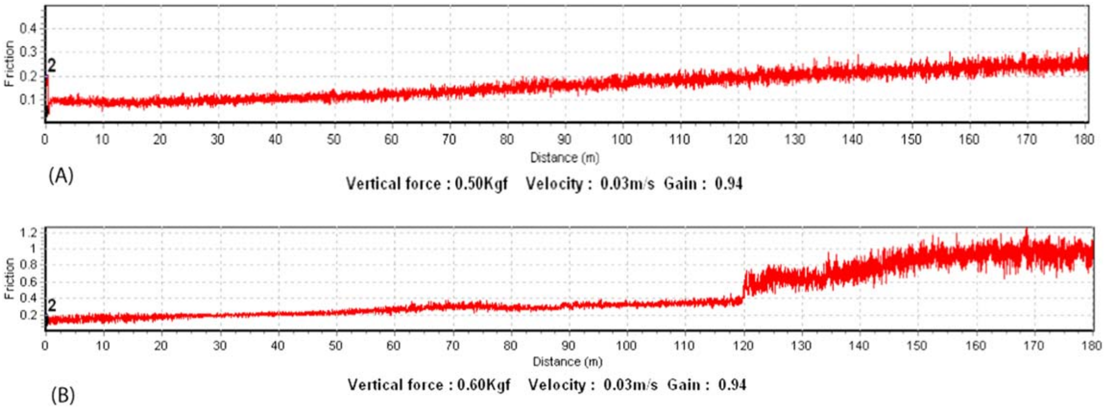

Figure 8. Optimal coating under wear load: (A) 500 gf, and (B) 600 gf.

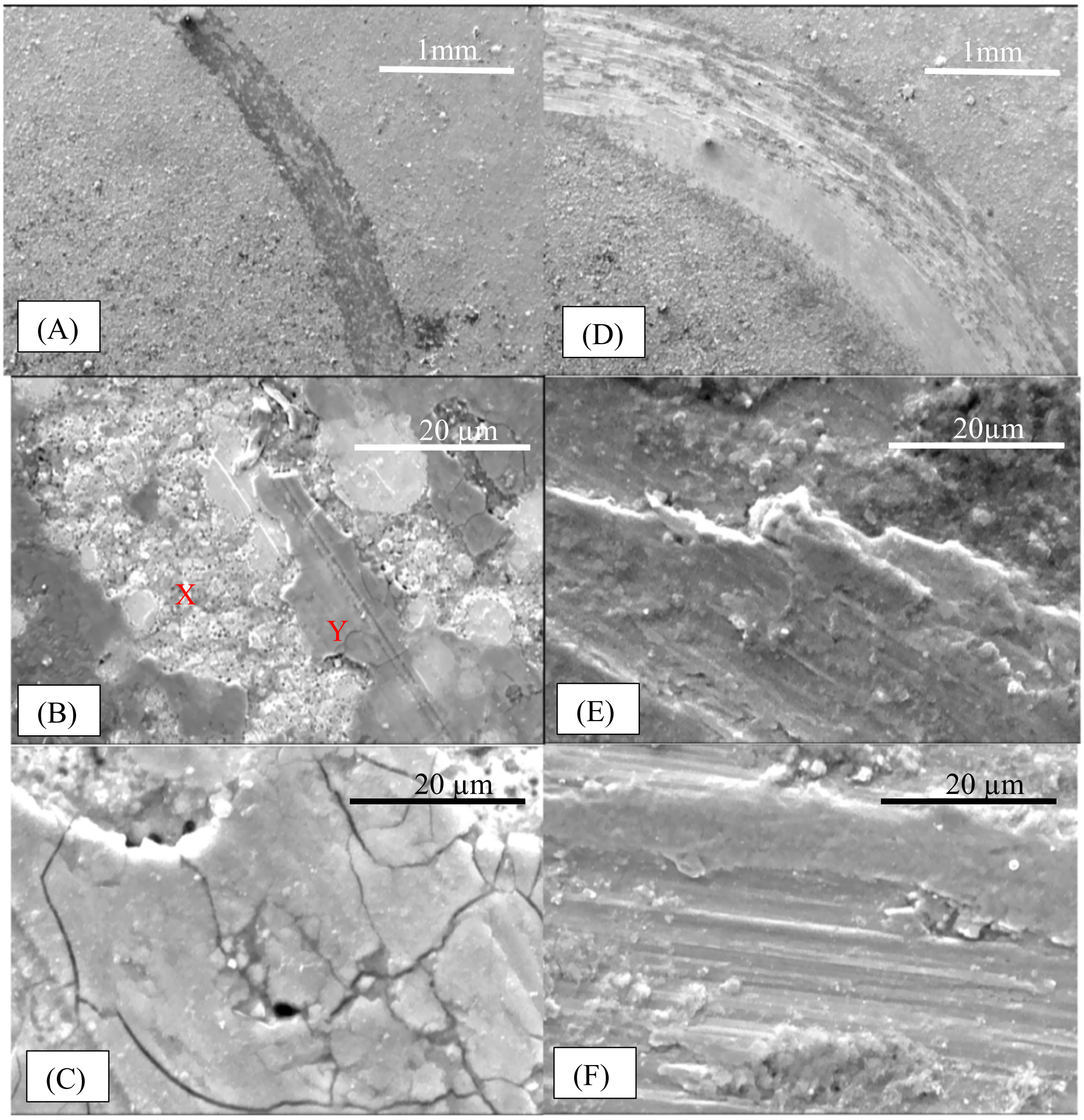

Figure 9. The SEM micrographs of the worn surfaces of the optimal Ni–Mo/Al composite coatings under wear conditions: (A)–(C) Vertical force: 500 gf; velocity: 0.03 m s^{-1}, and (E), (F) Vertical force: 600 gf; velocity: 0.03 m s^{-1}.

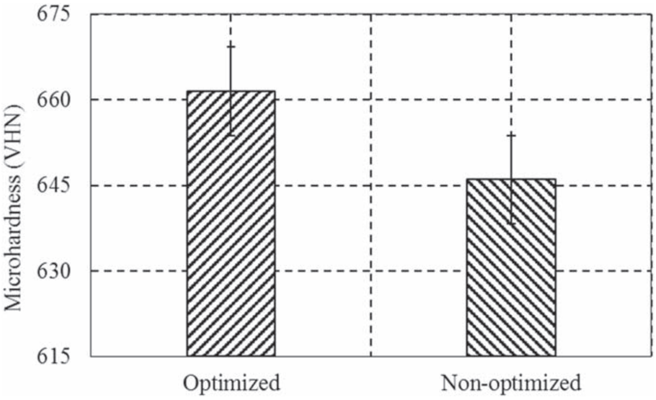

Figure 10. Microhardness of the coatings obtained under optimized and non-optimized conditions.

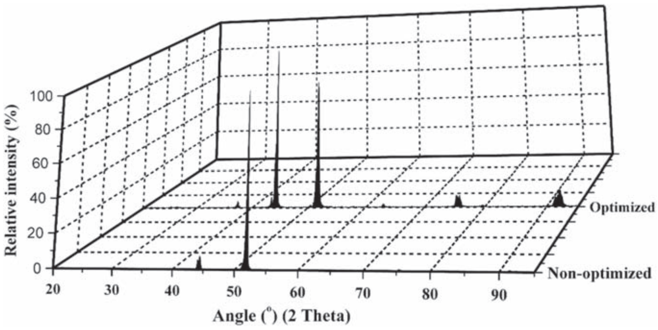

Figure 11. XRD patterns of the coatings obtained under optimized and non-optimized conditions.

up to 600 gf. The results for 500 gf and 600 gf are shown in figures 8(A) and (B), respectively. In spite of increasing wear load to 500 gf, the friction coefficient was not noticeably changed and varied from 0.1 to 0.3.

Figures 9(A)–(F) shows the SEM micrographs of the worn surfaces of the optimal Ni–Mo/Al composite coatings under the wear loads of 500 and 600 gf, respectively.

In the SEM images of the wear tracks on the coatings, it is visible that the composite coating under 500 gf load presents lower depth and width abrasive grooves compared to the composite coating under 600 gf (compare figures 9(A)–(C) with (D)–(F)). The wear mechanism under the load of 500 gf has not changed yet and is similar to that under a load of 400 gf, i.e. the wear mechanism can be considered as an abrasive mechanism. During wear test under a load of 400 gf, it was observed that the load is taken by some of the nodules while the other parts of the coating remain almost undeformed. By increasing the load to 500 gf, the face of the pin could not touch some places on the coating like X point in figure 9(B) and some other places like Y in figure 9(B) which were severely deformed. This shows that the total wear load was taken by these sites. It is clear that the amount of deformation under the load of 500 gf was greater than the deformation of nodules under the load of 400 gf. As is seen in figure 9(C) because of increasing wear load, the number of cracks in the coating increased. The variation in coefficient of friction as a function of sliding distance for optimal coating under the load of 600 gf is also presented in figure 8(B). On the other hand, as was shown in figure 8(B), a large variation in friction coefficient appeared after 120 m which suggested a change in the wear mechanism and showed a stick-slip behavior. The worn surface of the coating under a load of 600 gf indicates the ablation of the surface and suggests an adhesive wear mechanism (figures 9(D)–(F)). According to figure 8(B), it seems that after a distance of about 40 m in the wear test, a severely damaged surface is formed as was presented in figures 9(E), (F). The worn surfaces of the composite coatings along the sliding direction indicate a greater degree of wear and localized adhesion between the specimen pin surface and the counter body.

The results of microhardness test are also shown in table 7 and figure 10. As is seen, the coating obtained under optimal conditions is harder than that obtained under non-optimal conditions which are confirmed by Archard’s law.

The XRD patterns of the coatings obtained under optimal and non-optimal conditions are shown in figure 11. As is seen, the predominant texture for non-optimal coating is (200), whereas the mixture of (111) and

(200) planes exist on the surface of the optimal coating and the intensity of (111) planes is higher than that of (200) planes. It is known that (111) planes have a high planar atomic density than a (200) plane. Based on this property, (111) planes have low roughness and wear rate. As a consequence, the high wear resistance and hardness of the optimal coating compared to the non-optimal coating can be linked to the presence of these planes.

4. Conclusion

Ni–Mo/Al composite coatings were prepared via a citrate bath containing Al particles in different conditions. Taguchi method was used to achieve optimum conditions and the effect of different parameters. Accordingly, the following results were obtained:

1. Taguchi method could be a suitable DOE for Ni–Mo/Al composite coatings.
2. Taguchi method showed that flow density had the highest impact on the hardness and wear resistance of Ni–Mo/Al composite coatings.
3. Optimal parameters for the formation of Ni–Mo/Al composite coatings included annealing at 600 °C, annealing time of 2 h, 5 $g\,l^{-1}$ Al content in the bath, current density of 30 $mA\,cm^{-2}$ and stirring rate of 500 rpm.
4. The wear force and slip distance changed the wear mechanisms in Ni–Mo/Al composite coatings, especially in non-optimal coating.
5. The adhesive wear was predominant mechanism in Ni–Mo/Al composite coatings that produced in optimal conditions.

For the future research, the relationships between different parameters can be further explored and the comprehensive models with specific equations are extracted. In addition, the parameters of wear such as environmental effect can be investigated by Taguchi technique.

ORCID iDs

M Esmailzadeh https://orcid.org/0000-0002-3241-0667

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