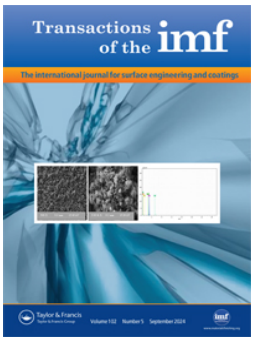

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Production of superhydrophobic surfaces on hydrophilic AA 6063 aluminium alloy and optimisation using a Taguchi design approach

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ABSTRACT

In this study, superhydrophobic surfaces were fabricated on aluminium alloy plates with two different experimental procedures using the dip coating method. Each procedure was optimised using the Taguchi design to evaluate the significance of factors such as SiC abrasive paper number, etching time, and chemical modification time. By determining three levels for these factors, the L9 orthogonal array was formed. Water contact angle (WCA) measurements determined the wettability of the surfaces against the water. Scanning Electron Microscopy (SEM) was used to assess the general appearance and roughness of the surfaces. The coating thickness of the surfaces was also measured. For both experimental procedures, the most effective parameter in providing superhydrophobic coatings on the surfaces was determined as the etching time. SEM analyses demonstrated the roughness created by the clustered and protruding structures formed on the surface after the coating. The WCAs above 150° indicate that superhydrophobic surfaces were prepared.

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KEYWORDS
Dip coating; experimental design; hydrophobic; Taguchi method

1. Introduction

In 1805, Thomas Young first described the angle at the contact surface due to the interaction between a liquid droplet and a solid surface.[1,2] This definition, which has remained an important feature for the characterisation of solids ever since, is called the *contact angle* and expresses the behaviour of the solid surface towards the water.[3,4] Wettability describes the contact state of the liquid at the contact surface with the solid surface and is an indicator of the hydrophobicity of the material.[5] A hydrophobic surface is a surface that has the ability to repel water, while hydrophilicity is a property of materials that have an affinity for water.[6,7] For a drop, when the contact angle with the surface is less than 90°, greater than 90° and greater than 150°, the surface is called hydrophilic, hydrophobic and superhydrophobic, respectively.[8]

A superhydrophobic surface is reported to have water-repellent and self-cleaning properties.[9] Self-cleaning is the ability of the surface to wash away dirt particles with drops of water flowing on it.[10] In addition to the leaves of the lotus plant (Nelumbo nucifera), which are the best-known examples of self-cleaning hydrophobic surfaces, butterflywings and the leaves of cabbage and Indian cress are examples of many highly hydrophobic surfaces in nature.[11]

It is also possible to obtain synthetic superhydrophobic surfaces inspired by natural materials. One of the preferred methods to obtain a synthetic superhydrophobic surface is coating applications. Surface coating is an engineering process that aims to change the properties of the material, such as performance and appearance, and to extend the life of the material.[12] Superhydrophobic coatings have applications and practicality.[13] They can be obtained using sol–gel, chemical etching, wet chemical, template deposition, electrochemical deposition, laser electrodeposition, solution immersion and spray coating methods.[14,15] Superhydrophobic coatings can be applied to many different materials, of which metals are among the most used. Metals preferred to obtain synthetically superhydrophobic surfaces include aluminium, copper, magnesium and steel.[16] Among these materials, aluminium and its alloys are important materials because they are abundant in nature, easy to process and have high technological value.[17] Since aluminium has a soft structure, it is often used in alloys with other metals.[18] Aluminium and its alloys have many uses such as construction, aerospace, automotive, transportation, marine, medical and civil engineering industries.[19,20]

Experimental design is a method that allows us to better understand and evaluate the factors affecting a system with statistical approaches and evaluates theoretical knowledge and working knowledge about the relevant factors together.[21] Traditionally, many experiments are carried out to determine the effect of a factor, keeping other factors constant, and this is quite time-consuming and requires huge resources.[22] Experimental design ensures that experimental conditions are optimised to maximise the amount of useful information obtained through a minimum number of experiments.[23] The Taguchi method is among the most commonly used experimental design approaches.[24] It is a method that requires a certain number of experiments depending on the factors of a process to determine optimum conditions and analyses the experimental results statistically and reveals the factors affecting the process.[25] It introduces an orthogonal array, performs variance analysis, and uses the Signal to Noise (S/N) ratio to minimise sensitivity to variations.[26] Among optimisation techniques, it targets the minimum quality loss and is the easiest option for process optimisation through experimental design.[27]

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Table 1. Chemical composition (wt %) of AA 6063 aluminium alloy.

Al	Mg	Si	Fe	Mn	Zn	Cu
98.7448	0.4804	0.4492	0.1709	0.0488	0.0293	0.0171

The current study has focused on obtaining superhydrophobic coatings on aluminium alloy surfaces. Aluminium alloy plates were used as target surfaces in the study. The dip coating technique was used to obtain the coatings. Two experimental procedures were used to form superhydrophobic coatings on aluminium alloy surfaces, in which the same processes were carried out with different chemicals and times in the etching and modification stages. The coated surfaces were characterised by Scanning Electron Microscopy (SEM-EDX), contact angle and coating thickness measurement. The obtained water contact angles (WCAs) were optimised with the Taguchi experimental design technique. For this purpose, the Minitab 18 programme was used and the experimental design was obtained in the L9 orthogonal array. Additionally, the effect of uncontrollable parameters was reduced through S/N ratios, and the effects of controllable parameters on the result were determined by analysis of variance (ANOVA).

2. Materials and methods

2.1. Materials

AA 6063 aluminium alloy plates were sized as 7 cm x 2 cm x 1.5 mm. The plates were supplied by Arslan Alüminyum Incorporated Company, Bilecik. The chemical composition of AA 6063 aluminium alloy plates, obtained from the supplier, is given in Table 1.#400, #600 and #800 SiC papers were used in the pretreatment abrasion processes. Hydrochloric acid (HCl – 37%, Merck, Rahway, New Jersey, United States), sodium hydroxide (NaOH – 97%, Merck, Rahway, New Jersey, United States) and acetic acid (CH_3COOH – 99.8%, Sigma-Aldrich, St. Louis, Missouri, United States) were used in the etching processes. Dodecyltriethoxysilane ($C_{18}H_{40}O_3Si$ – 99.8%, Sigma-Aldrich, St. Louis, Missouri, United States) and palmitic acid ($C_{16}H_{32}O_2$ - 99%, Sigma-Aldrich, St. Louis, Missouri, United States) were used as modification chemicals. Ethyl alcohol (99.5%, Sigma-Aldrich, St. Louis, Missouri, United States) was used as the solvent of the modification chemicals.

2.2. Experimental design

This research used experimental design to develop superhydrophobic coatings on the surfaces of aluminium alloy plates. Three factors were determined in the experimental design: coarseness number of SiC papers, etching time and chemical modification time. Three levels of the factors were assigned and their effect on the process and interactions with each other were studied. For this purpose, two different experimental studies were conducted. The significance of the determined factors and their effect on the study and the optimisation of the experimental study were examined with the Taguchi approach. WCA was used as an experimental response according to factors and levels in the design. In the experimental design applied for both of the experimental studies, 9 experiments were performed related to L9 orthogonal array using the Minitab 18 programme.

In the 1st experimental study, aluminium alloy plates were abraded using #400, #600 and #800 SiC papers. The abraded plates were etched by immersing them in HCl solution for different periods (2, 4 and 6 minutes). In the modification process; the etched plates were modified using dodecyltriethoxysilane solution for different periods (8, 16 and 24 hours). Table 2 presents the factors and the levels and Table 3 shows the factor-level combination for the 1st experimental study.

Table 2. Factors and levels for the 1st experimental study.

Factor	Allocated letter	Levels	
SiC paper number (#)	A	A1	400
		A2	600
		A3	800
Etching time (min)	B	B1	2
		B2	4
		B3	6
Chemical modification time (h)	C	C1	8
		C2	16
		C3	24

Table 3. L_9 orthogonal array for the 1st experimental study.

Sample	Factor and level combination		
	A	B	C
1	A1	B1	C1
2	A1	B2	C2
3	A1	B3	C3
4	A2	B1	C2
5	A2	B2	C3
6	A2	B3	C1
7	A3	B1	C3
8	A3	B2	C1
9	A3	B3	C2

In the 2nd experimental study, the plates were abraded using three different SiC papers (#400, #600 and #800). The abraded plates were kept in NaOH solution, and then immersed in a solution of water (H_2O): HCl: CH_3COOH for different periods (3, 5 and 10 minutes). Then, the etched plates were modified by immersing them in a 2% palmitic acid solution for different periods (15, 30 and 60 minutes). Table 4 and Table 5 show the levels of the factors and the factor-level combination for the 2nd experimental study, respectively.

Table 4. Factors and levels for the 2nd experimental study.

Factor	Allocated letter	Levels	
SiC paper number (#)	D	D1	400
		D2	600
		D3	800
Etching time (min)	E	E1	3
		E2	5
		E3	10
Chemical modification time (min)	F	F1	15
		F2	30
		F3	60

2.3. Sample preparation

The combinations given in Tables 3 and 5 were used for the 1st and 2nd experimental studies, respectively. For both experimental studies, firstly the aluminium alloy plates were abraded with SiC papers #400, #600 and #800. To remove residues and impurities, the surfaces were cleaned with deionised water in an ultrasonic bath for 2 minutes and then

Table 5. L_9 orthogonal array for the 2nd experimental study.

Sample	Factor and level combination		
	D	E	F
1	D1	E1	F1
2	D1	E2	F2
3	D1	E3	F3
4	D2	E1	F2
5	D2	E2	F3
6	D2	E3	F1
7	D3	E1	F3
8	D3	E2	F1
9	D3	E3	F2

dried at room temperature for 1 hour. The dried plates were stored to be used in the experiments. After this process, two different experimental studies were carried out for the plates.

The procedure applied in the 1st experimental study is given below:

The etching process was conducted similarly to that used by Li *et al.*[28] and Liu *et al.*[29] A 4 M HCl solution was prepared using distilled water to etch the surfaces. By the orthogonal array obtained in the Taguchi design, the plates were dipped in 4 M HCl solution for 2, 4 and 6 minutes. The etched plates were cleaned in an ultrasonic water bath for approximately 10 minutes and dried at room temperature for 1 hour. The method presented by Fu and He was used for modification.[30] An ethanol solution of dodecyltriethoxysilane was prepared at a concentration of 10 mM (0.01 M). According to the orthogonal array obtained in the Taguchi design, the etched plates were modified by immersing them in this solution for 8, 16 and 24 hours. The samples were washed with ethanol and then cured at 150 °C for 5 minutes.

The procedure in the 2nd experimental study was carried out as explained below:

For the etching process, the published method by Esmaeilirad *et al.* was used.[31] A two-stage etching process was applied. In the first stage, all the plates were immersed in 1 M NaOH solution for 10 minutes. Then, the plates were washed with deionised water in an ultrasonic bath for approximately 2 minutes and dried at 110 °C for 10 minutes. In the 2nd stage, an etching solution containing chemicals in the ratio of 20 mL H_2O: 8 mL HCl: 1 mL CH_3COOH was prepared. By the orthogonal array obtained in the Taguchi design, base-treated plates were immersed in this acidic solution for 3, 5 and 10 minutes. The plates, whose etching process was completed, were washed with deionised water in an ultrasonic bath for approximately 2 minutes and dried at 110 °C for 10 minutes. A combination of previously presented studies was used for the modification process.[19,32,33] An ethanol solution of palmitic acid (2 wt%) was prepared. The solution temperature was set at 70 °C. According to the orthogonal array obtained in the Taguchi design, the etched plates were immersed in this solution for 15, 30 and 60 minutes. After the modification process, the samples were washed with ethanol and dried at 80°C for 30 minutes in an oven.

2.4. Characterisation techniques

The WCAs were determined using a digital optical microscope (Biolin Scientific Attension-Theta LiteContact Angle Meter, Stockholm, Stockholms Lan, Sweden). The WCA values were measured with 4 µL Merck water at three random points on each sample at room temperature and these values were averaged. The surface characteristics of the samples were investigated by SEM (Zeiss Supra 40 VP, Oberkochen, Baden-Württemberg, Germany) analysis at 15.0 kV. The thickness of the coatings on the samples was determined using a Fischer MP0 (Hampton, New Hampshire, United States) device. The device determines the coating thickness by magnetic induction or eddy current.[34] The coating thicknesses were measured in 'µm' and the average of 3 measurements was calculated.

3. Results and discussion

3.1. Taguchi design of 1st experimental study

In the Taguchi optimisation approach, WCA values of the prepared samples were assigned as the response of the design. The Taguchi experimental design evaluates the effect and significance of a factor for each experiment by the S/N ratio.[35] There are nominal-the-best, the smaller-the-better, and the larger-the-better alternatives for the S/N ratio depending on the study.[36] Since it was aimed to obtain the highest WCA value in the L9 orthogonal array, the 'larger is better' approach was used. S/N ratio in decibel (dB) unit is defined by Equation (1)[37]:

$$S/N(dB) = -10log_{10}\left(\sum_{i=1}^{n} y_i^2\right) \tag{1}$$

In Equation (1), n symbolises the variable number, and y_i symbolises the value of each variable.[37] WCA, S/N ratio and standard deviation (STD) of each sample for the 1st experimental procedure are listed in Table 6. The experimental results listed in Table 6 show that the 9 prepared samples have hydrophobic (WCA > 90°) surfaces. In addition, it is seen that the WCA values for some of the coated samples are even higher (WCA > 150°), meaning that the surfaces are superhydrophobic. Accordingly, as seen in Table 6, the samples 3 (161°), 6 (157°) and 9 (162°) exhibited superhydrophobic properties. Thus, it was determined that the surface that showed the strongest superhydrophobic feature belonged to sample 9 with the highest WCA value as 162°. The STD values of Samples 1–9 ranged between 0.577-2.516 and revealed the homogeneity of the coated surfaces.[38] In the Taguchi approach, the result should be highly dependent on the signal and have minimal variability with noise.[39] According to the S/N ratios in Table 6, the design considers the experimental study used in the preparation of Sample 9 as suitable for this explanation.

Table 6. Taguchi L_9 array response of the 1st experimental study.

Sample	Factor and level combination			Response		
	A	B	C	WCA (°)	S/N (dB)	STD (°)
1	A1	B1	C1	122	41.72	1.732
2	A1	B2	C2	133	42.47	1.000
3	A1	B3	C3	161	44.13	0.577
4	A2	B1	C2	121	41.65	1.527
5	A2	B2	C3	144	43.16	1.154
6	A2	B3	C1	157	43.91	0.577
7	A3	B1	C3	128	42.07	0.577
8	A3	B2	C1	138	42.86	2.516
9	A3	B3	C2	162	44.19	1.732

Table 7. E_f and factor ranking for the 1st experimental study.

Level	Factor		
	A	B	C
1	138.7	123.7	139.0
2	140.7	138.3	138.7
3	142.7	160.0	144.3
E_f	4.0	36.3	5.6
Rank	3	1	2

Table 8. ANOVA for the 1st experimental study.

Source of variance	Degree of freedom	Sum of square	Mean square	Factor effect (%)	F-ratio	*P*-value
A	2	24.00	12.00	1.14	1.29	0.438
B	2	2004.67	1002.33	95.10	107.39	0.009
C	2	60.67	30.33	2.88	3.25	0.235
Residual error	2	18.67	9.33	0.89		
Total	8	2108.00		100.00		

Factor main effect (E_f) analysis is a method used to determine the effect of factors on the performance of the process. To calculate E_f, the average of the responses of the factors at the specified levels is taken and then the maximum average is subtracted from the minimum one.[40] The E_f analysis of the 1st experimental study is listed in Table 7 which shows the factor B, etching time, is the most effective factor with rank 1. A second significant factor is C, modification time. And SiC abrasive paper coarseness number has less effect on the experimental study and is ranked as 3.

To assign the optimum level for each factor, the factors main effect plot also can be used. The plot is presented in Figure 1. According to Figure 1, the best performance for wetting characteristics of the coated surfaces can be obtained at #800 SiC paper (A3), 6 minutes etching time (B3) and 24 hours chemical modification time (C3).

ANOVA for the 1st experimental study was performed using the S/N ratios obtained based on the experimental results for a 95% level of confidence. The effects of the factors on the WCA values were determined and the results are given in Table 8. The impact and statistical evaluation of the factors are given in Table 8. ANOVA created an F-value that indicates the effect of the quantitative change of each factor.[41] Based on the Taguchi approach, ANOVA accounts for the statistical significance of the factor by creating a *P*-value corresponding to each S/N ratio.[42,43] A *P*-value less than 0.05 indicates the significance of the factor, while a *P*-value greater than 0.1 expresses that the factor is not significant.[44,45] A high F-value and a *P*-value less than 0.05 indicate the effective parameter.[46] Thus, while factor B, i.e. etching time, was assigned as the most effective factor, it was determined that the effect of abrading and modification time were insignificant. In this way, the factor effect percentages in Table 8 were supported. The ANOVA results were found to be consistent with E_f analysis.

For the 1st experimental study, the effective factor was determined to be etching time. Additionally, among the three various etching times, the highest contact angle value was obtained for the longest time. In this case, it can be said with the support of other studies in the literature that increasing the etching time increases the surface roughness and therefore leads to higher WCA values.[47,48] Thus, the sensitivity of contact angle to surface roughness was also supported.[49]

Figure 1. Factors main effect plot for the 1st experimental study.

The model summary for the 1st experimental study is given in Table 9. A high regression coefficient value (0.9911) was obtained for the model for the 1st experimental study. The high adjusted R^2 value (0.9646) clarifies that the model is adequate to predict WCA values for the superhydrophobic coating of aluminium alloy plate surfaces.[50]

Another important issue in Taguchi experimental design is the estimation of experimental WCA values. It is aimed to achieve the closest value to a value obtained in experimental studies, with the lowest difference and error, and this way it is confirmed. Confirmation studies were carried out with a known WCA result, and the results are presented in Table 10. The difference between the predicted WCA (160°) and the experimental WCA (162°) of the sample was negligible. The low error of 1.23% demonstrated the agreement between the experimental result and the prediction, and therefore the consistency and repeatability of the study.

3.2. Taguchi design of 2nd experimental study

In the 2nd experiment, unlike the 1st experiment, during the etching process, the samples were first kept in a basic solution and then in an acidic solution for different periods. In addition, the chemical modification agent and the chemical modification times also are different. As in the 1st study, it was aimed to obtain the highest WCA value in the L9 orthogonal array and the 'larger is better' approach was used for the 2nd study. WCA, S/N ratio and STD of each sample for the 2nd experimental study are given in Table 11. According to the WCA values given in Table 11, the surfaces of all coated samples exhibited hydrophobic properties. In addition, Samples 3, 6 and 9 have WCA values of 168°, 162° and 158°, respectively, and have gained superhydrophobic properties.

Table 9. Model summary for the 1st experimental study.

S	R-sq	R-sq(adj)	PRESS	R-sq(pred)
3.05505	0.9911	0.9646	378	0.8207

(Regression equation; result = 93.00 + 0.01000A + 9.083B + 0.333C)

Table 10. Validation experiment results for the 1st experimental study.

A3B3C2	Taguchi prediction	Validation experiment	Absolute difference	% Absolute error
S/N (dB)	44.07	44.19	0.12	0.27
WCA (°)	160	162	2	1.23

Table 11. Taguchi L_9 orthogonal array response of the 2nd experimental study.

	Factor and level combination			Response		
Sample	D	E	F	WCA (°)	S/N (dB)	STD (°)
1	D1	E1	F1	109	40.74	0.577
2	D1	E2	F2	105	40.58	1.527
3	D1	E3	F3	168	44.50	1.000
4	D2	E1	F2	119	41.51	1.527
5	D2	E2	F3	124	41.86	1.527
6	D2	E3	F1	162	44.19	2.886
7	D3	E1	F3	106	40.50	1.527
8	D3	E2	F1	125	41.86	0.577
9	D3	E3	F2	158	43.97	2.000

Thus, Sample 3 has a surface that demonstrates the superhydrophobic feature effectively with a WCA of 168°. The STD values of the coated samples were 0.577–2.886 and again indicated the homogeneity of the surfaces.[38] According to the S/N ratio value of Sample 3, the Taguchi approach considers this experimental study as optimum.

Table 12 represents the E_f analysis of the 2nd experimental study. As given in Table 12, the most effective factor is the factor E, etching time, ranked as 1. SiC paper coarseness number was assigned as the second significant factor with rank 2. And the less effective factor was determined as modification time.

Additionally, the main effect factors plot is presented in Figure 2. According to Figure 2, the best superhydrophobic properties of the coated surfaces can be obtained at #600 SiC paper (D2), 10 minutes etching time (E3) and 60 minutes chemical modification time (F3).

ANOVA for the 2nd experiment is given in Table 13. This analysis was performed at a 95% confidence level. According to the values in Table 13 created by ANOVA, the effects and significance levels of the factors were evaluated for the 2nd experimental study. Consistent with the E_f analysis, it was shown by ANOVA that the most influential factor was E, i.e. etching time. Additionally, it was determined that the SiC paper number and modification time were not significant factors.

Table 12. E_f and factor ranking for the 2nd experimental study.

Level	Factor		
	D	E	F
1	127.3	111.3	132.0
2	135.0	118.0	127.3
3	129.7	162.7	132.7
E_f	7.7	51.3	5.4
Rank	2	1	3

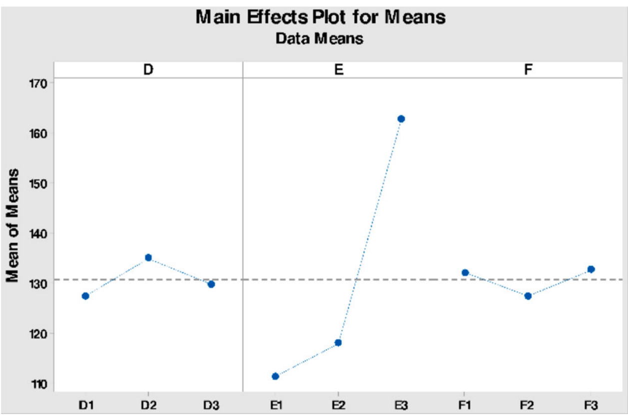


Figure 2. Factors main effect plot for the 2nd experimental study.

Table 13. ANOVA results of the 2nd experimental study.

Source of variance	Degree of freedom	Sum of squares	Mean squares	Factor Effect (%)	F-value	*P*-value
D	2	92.67	46.33	1.83	0.36	0.733
E	2	4674.67	2337.33	92.17	18.40	0.052
	2	50.67	25.33	1.00	0.20	0.834
F	2	254.00	127	5.01		
Residual error	8	5072.00		100.00		

Similar to the 1st experimental study, the effective factor in the 2nd study was determined as etching time and the highest WCA value was obtained at the highest etching time. Here again, it can be said that increasing roughness with increasing etching time also increases WCA.

The model summary of the 2nd experimental study is given in Table 14. The study yields a high regression coefficient value (0.9499) and an adjusted R^2 value as 0.7997.

For the estimation of the WCA value for the 2nd experimental study, confirmation studies were performed. The results of these are presented in Table 15. The predicted WCA (161°) and experimental WCA (168°) of the sample were found to be close to each other and the error (4.16%) confirmed the consistency and repeatability of the study.

3.3. Contact angle measurements

The water-repellent properties of the aluminium alloy plate surfaces were determined by WCA measurements. WCA measurements were carried out on the surfaces of untreated, abraded and coated plate samples. Measurements were repeated three times on random points of the surfaces and averaged. Table 16 includes the WCA images of untreated aluminium alloy plates. According to the measurements, the average WCA value was determined as 60° similar to previous studies.[51]

The WCAs of the aluminium alloy plates measured after the abrading processes are also given in Table 16. According to the images seen in Table 16, the average WCA values of the abraded surfaces were determined as 94°, 91° and 75° using #400, #600 and #800 SiC paper, respectively. It has been determined that as the number of abrasive SiC paper increases, the measured WCA value decreases.

WCA analysis of the 9 plates prepared in the 1st experimental study according to Taguchi design was performed. WCAs were measured for three random measurements on the surfaces of each sample. The average WCA values of the plates prepared as Samples 1–9 were determined as 122°, 133°, 161°, 121°, 144°, 157°, 128°, 138°, and 162°, respectively. According to these results; it can be seen that the WCA value

Table 14. Model summary for the 2nd experimental study.

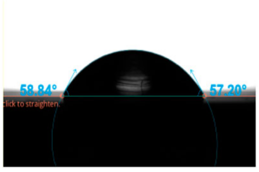
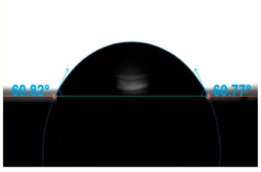
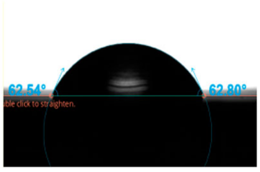
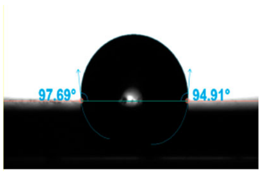
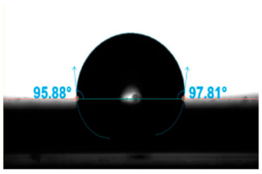
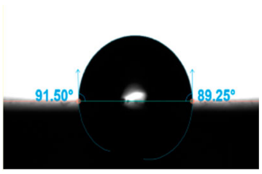
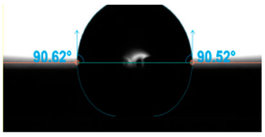
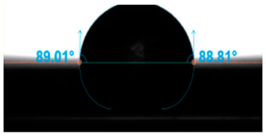
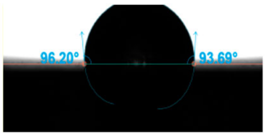
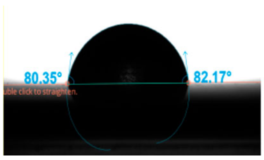
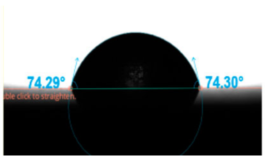
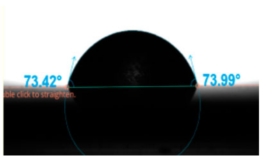
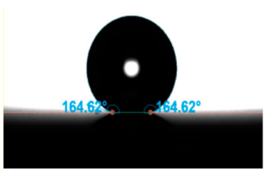
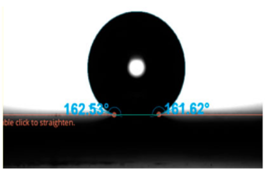
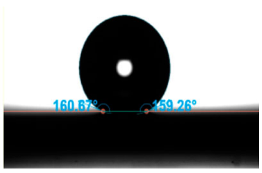
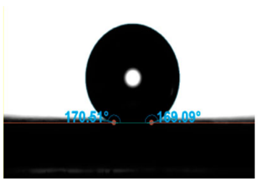
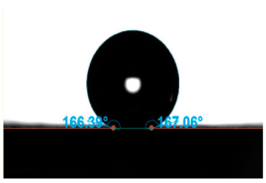
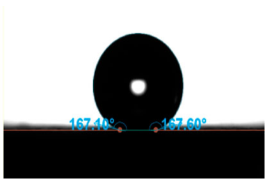
S	R-sq	R-sq(adj)	PRESS	R-sq(pred)
11.2694	0.9499	0.7997	5143.5	0.0000

(Regression equation; result = 79.0 + 0.0058A + 7.78B + 0.027C)

Table 15. Validation experiment results for the 2nd experimental study.

D1E3F3	Taguchi prediction	Validation experiment	Absolute difference	% Absolute error
S/N (dB)	44.03	44.50	0.43	0.96
WCA (°)	161	168	7	4.16

Table 16. Contact angle measurements.

Sample	Measurement 1 (°)	Measurement 2 (°)	Measurement 3 (°)	average (°)
Aluminium alloy plate				60
Aluminium alloy plate abraded with #400 SiC paper				94
Aluminium alloy plate abraded with #600 SiC paper				91
Aluminium alloy plate abraded with #800 SiC paper				75
Superhydrophobic surface obtained in the 1st study 1st experimental study.				162
Superhydrophobic surface obtained in the 2nd study				168

of 150° and above, which is necessary to define the superhydrophobic character, was obtained for Samples 3, 6 and 9. It is clear that the coating process provides hydrophobic and superhydrophobic properties to the hydrophilic aluminium alloy plate surfaces. In the examination of the factors for Samples 3, 6 and 9, it is seen that different SiC papers and modification times are used, but the etching time is the same in the preparation of the coatings. This supports the conclusion that etching time is the most effective factor. In addition, the highest WCA value among the surfaces that have superhydrophobic properties was obtained for Sample 9 as 162°. WCA images and values of Sample 9 are presented in Table 16.

WCA values and images of the plates fabricated in the 2nd experimental study according to the Taguchi L9 orthogonal array were taken. The average WCA values of Sample 1–9 were determined as 109°, 105°, 168°, 119°, 124°, 162°, 106°, 125°, 158°, respectively. It is also seen here that the applied experimental study leads to hydrophobic and superhydrophobic coatings on aluminium alloy surfaces. According to the WCAs, the superhydrophobicity was obtained for Samples 3, 6 and 9 and Sample 3 had the highest WCA. WCA analysis of Sample 3 are given in Table 16. Samples 3, 6 and 9 were abraded using different SiC papers and modified various times, but etched at the same time. Similar to the 1st experimental study, etching time stood out as the most effective factor.

3.4. SEM-EDX analysis

SEM-EDX analysis was used to investigate the structure and morphology of the surfaces of the aluminium alloy plate, abraded plate surfaces with three different SiC papers and superhydrophobic surfaces obtained in the 1st and 2nd experimental studies. Figure 3 shows the aluminium alloy plate surface before any treatment. As seen in Figure 3, the plate exhibited an almost smooth surface with some scratches, similar to the presented image by Zhu *et al.*[52] Additionally, high Al content is seen in the EDX image of the plate surface.

SEM-EDX analyses of the aluminium alloy plates abraded with #400, #600 and #800 SiC papers are given in Figure 4 (a), (b) and (c), respectively. When the images in Figure 4 are examined, it is seen that the abrading process causes the formation of irregular structures on the surfaces, the depths of which may differ due to human handicraft,

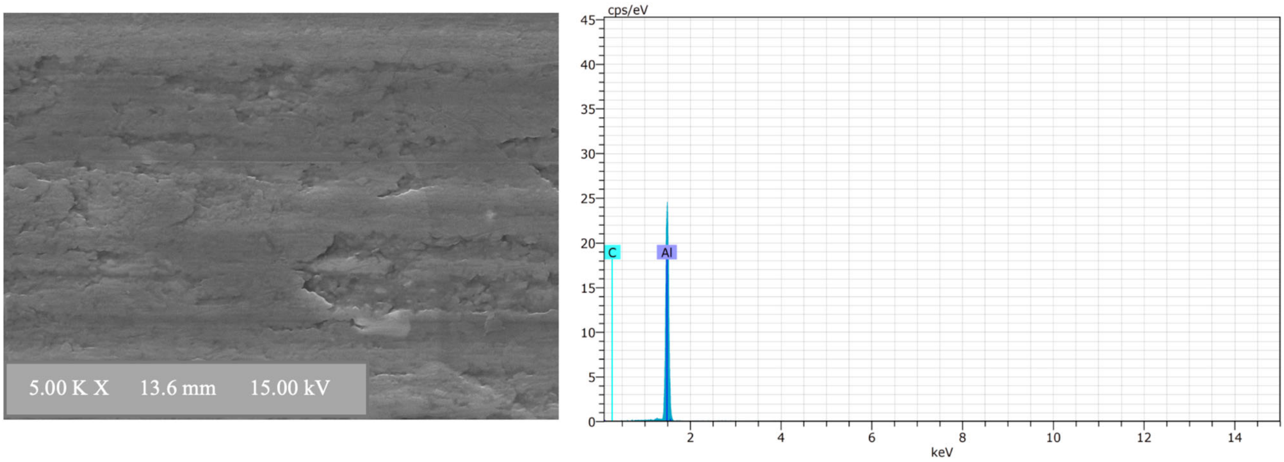


Figure 3. SEM-EDX analysis of aluminium alloy plate surface.

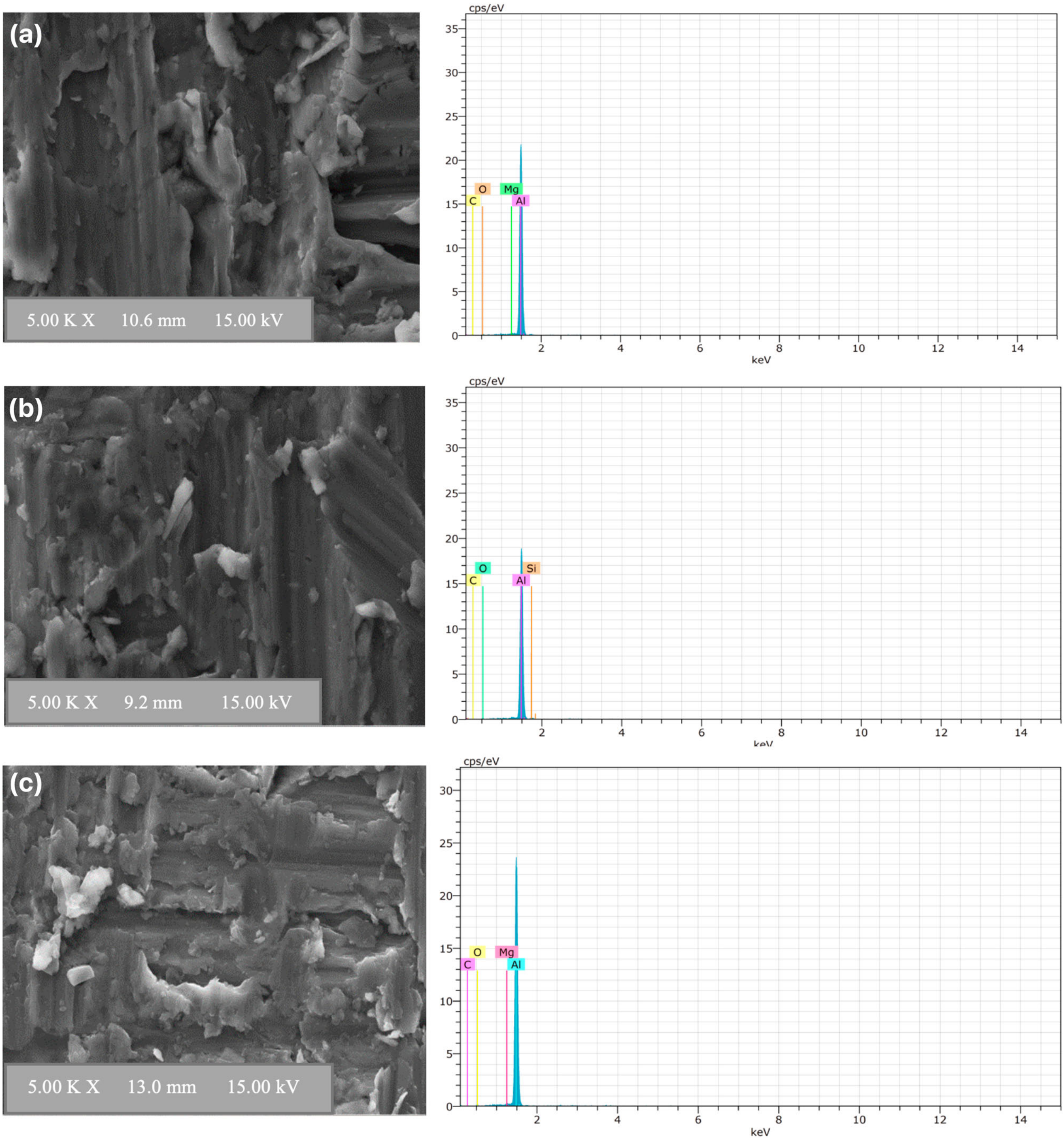


Figure 4. SEM-EDX images of aluminium alloy plates abraded with **(a)** #400, **(b)** # 600 and **(c)** #800 SiC paper.

compared to Figure 3. Accordingly, it was determined that the abrading process created coarse roughing on the surface of the plates. Similar images were presented by Saifaldeen *et al.*[53] Additionally, as shown in Figure 4 (a)-(c), it is seen that the protruding and granular structures on the aluminium alloy plate decrease in size from abrasive paper #400 to #800. In the EDX image, in addition to the high Al content, other compositions such as Si and Mg contained in the alloy are also seen.

The SEM-EDX image of the superhydrophobic surface (WCA 162◦) prepared in the 1st experimental study in different magnifications is given in Figure 5. It is known that when aluminium, an active metal, interacts with HCl, it causes cavities and plateaus on the aluminium surface due to its high surface energy regions (crystal boundary, line defect etc.) and presence of halide ions.[29] Supporting this explanation, the coated aluminium alloy surfaces in the 1st experimental study presented surface structures that can be called roughness, as shown in the figure. As shown in the low-magnification SEM image there are various sized clusters on the coated aluminium alloy surface. Additionally, in the high-magnification image, these clusters appear to contain protruding structures. Many regular protrusions are seen on the surface and these protrusions are approximately 2–4 µm in size. As seen in the figure, the alloy composition is dominant in the EDX image of the coated surface.

Figure 6 belongs to the SEM-EDX image of the superhydrophobic surface (WCA 168◦) prepared in the 2nd experimental study. As given in the previous study, in the 2nd experimental study, as a result of the reactions that occur during the contact of aluminium alloy surfaces with NaOH, the roughness of the surface was formed, and the roughness was further improved by using an acidic solution for this surface.[31] In the low magnification SEM image, the surface of the coated aluminium alloy plate with irregular roughnesses is visible. A closer view of these roughnesses is included in the high-magnification image. It can be seen that there were many micro/nano-sized spheres and protrusion-shaped structures on the coated surface. These structures appear to be between 0.5 nm and 2 µm in size. In the EDX image of the coated surface, peaks belonging to the composition of the alloy and the coating material are seen.

3.5. Coating thickness measurements

The thickness of the coatings on the aluminium alloy plate surfaces, which gained superhydrophobic properties by coating them according to the procedure obtained in the 1st and 2nd experimental studies, were measured. Thickness measurements were also made for untreated and abraded plates to compare them with coated plates and to reveal the change between each other. For each plate, measurements were taken from three random areas and the average of these measurements was calculated and presented in Table 17. According to Table 17, the average coating thickness of the aluminium alloy plate before any processing was measured as 1.66 µm. This can be explained by uncontrollable formations on the surface during the production of aluminium alloy plates. Additionally, the reason for this thickness may be the native oxide layer formed on aluminium alloy.[54–56] It can be seen that abraded surfaces have a lower thickness than the unprocessed aluminium alloy plate. The reason for this may be undesirable conditions that create thickness in the

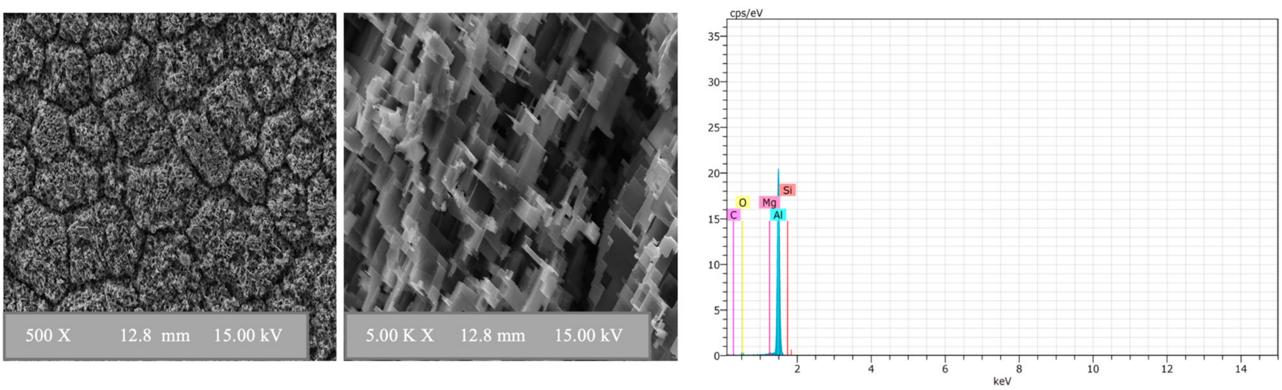


Figure 5. SEM-EDX images of superhydrophobic surface prepared in the 1st experimental study (Sample 9).

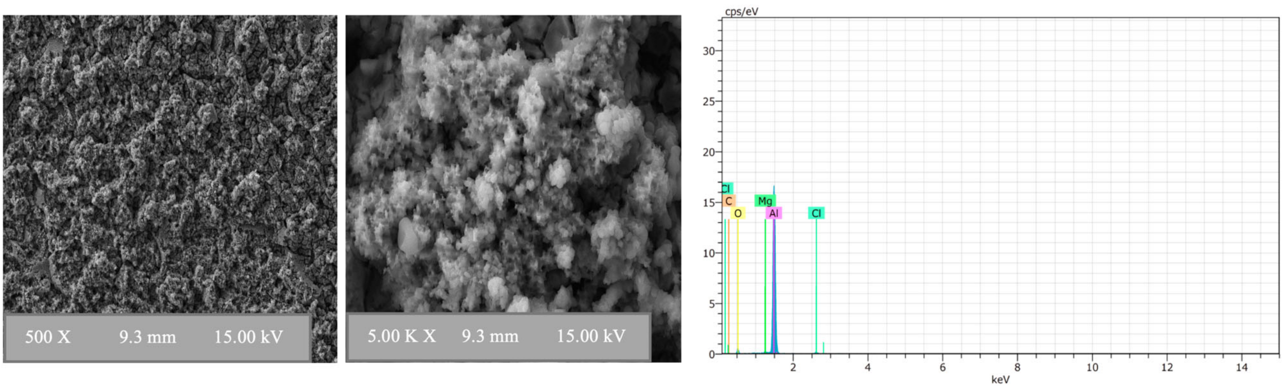


Figure 6. SEM-EDX images of superhydrophobic surface prepared in the 2nd experimental study (Sample 3).

Table 17. Coating thickness measurements.

Sample	Measurement 1 (μm)	Measurement 2 (μm)	Measurement 3 (μm)	average (μm)
Aluminium alloy plate	1.70	1.50	1.80	1.66
Aluminium alloy plate abraded with #400 SiC paper	1.30	1.60	1.80	1.56
Aluminium alloy plate abraded with #600 SiC paper	0.72	0.84	1.20	0.92
Aluminium alloy plate abraded with #800 SiC paper	0.84	0.96	0.80	0.86
Superhydrophobic surface (WCA 162◦) obtained in the 1st experimental study 1st experimental study.	13.40	13.70	13.30	13.46
Superhydrophobic surface (WCA 168◦) obtained in the 2nd experimental study	12.90	13.70	11.80	12.80

aluminium alloy plate are removed by the abrading process. Moreover, similar to Fernández-Hernán *et al*., the thickness decreased as the number of SiC paper increased.[57] In the thickness measurements for the surfaces with the highest WCA value, which were coated according to the procedure in the 1st and 2nd experimental studies, thickness values were obtained as 13.46 and 12.80 μm, respectively. Although the coating thickness obtained for the 1st experimental study was slightly larger than that of the 2nd experimental study, it was determined that they were close to each other.

4. Conclusions

This study aimed to prepare superhydrophobic surfaces on aluminium alloy plates *via* a simple and cost-effective dip coating method. For this purpose, two different experimental procedures were used in which the same processes (abrading, etching and modification) were performed with different chemicals and times. Abrasive SiC paper number, etching time and modification time factors in these experimental studies were optimised with the Taguchi approach and the significance of the factors was researched. In the experimental design, three levels were selected for each factor and the L9 orthogonal array was created. WCA was determined as the output of the experimental studies. Taguchi design revealed that the most effective factor in providing superhydrophobic coatings on aluminium alloy plate surfaces was determined to be the etching time for both experimental procedures. WCA measurements were used to specify the behaviour of the surfaces against water. In both studies, it was determined that the coated surfaces exhibited hydrophobicity with WCA values >90°. It was also confirmed that some surfaces had superhydrophobic properties when the coating was applied, with the WCA value obtained in contact angle measurements being >150°. SEM analysis showed the uncoated, abraded and coated aluminium alloy surfaces and defined the formations on the surfaces that introduced roughness. According to the coating thickness measurements of the surfaces, similar thicknesses were determined in both experimental studies. As a result of this research, it was determined that the surfaces of aluminium alloy plates were coated using two different experimental procedures to provide water repellency, and it was shown that superhydrophobic surfaces were achieved with the roughnesses and WCAs >150°.

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Disclosure statement

No potential conflict of interest was reported by the author(s).

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