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To cite this article: N A Mohamad Basir *et al* 2020 *IOP Conf. Ser.: Mater. Sci. Eng.* **991** 012096

ICCEIB 2020 IOP Publishing
IOP Conf. Series: Materials Science and Engineering **991** (2020) 012096 doi:10.1088/1757-899X/991/1/012096

Removal of ferum ions from acid mine drainage wastewater using jar test technique: Factorial design analysis

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Abstract. A factorial experimental design technique was used to investigate the removal of “Fe^{2+}” from wastewater Acid Mine Drainage (AMD) through jar adsorption test technique. Jar testing is expected to contribute to cost savings in chemicals from which ferum ion can also be extracted. The best condition for slag adsorption from AMD wastewater was obtained via a complete 16 factorial design experiment. Factorial design for screening is chosen to test four effects factors Basic Oxygen Furnace (BOF) slag dosage (0.2 and 2.0g), stirration speed (150 and 250 rpm), interval time (5 and 100 minute) and concentration (200 and 1000 ppm) at two levels. Metal removal capability was evaluated using adsorption result. Using statistical methods, the main effects and interaction effects of the four variables were analysed. A regression model was suggested, and the experimental data was found to match very well. The findings were statistically analyzed using the Student’s t-test, variance analysis, F-test and lack of fit to identify three most significant process variables that influence “Fe^{2+}” removal percentage. Therefore concentration was found to be the most important variable in this analysis.

1. Introduction

The potential presence of acidic environment when these rocks contain metal sulphides, especially iron disulphide (FeS_2) which is pyrite, and or marcasite, they become oxidized by air and water producing Fe^{2+}, H^+ and SO_4^{2-} ions. When these ions get into the solution, sulphuric acid is produced (AMD) [1–3]. Generally acid mine drainage (AMD) has a lower pH value that range between 2 and 3 or in certain cases lower than 5 [4]. AMD is produced in mining areas, minerals and waste materials containing sulphide minerals including pyrite. Pyrite (FeS_2) is exposed to water in coal or hard rock, and oxygen is responsible for initiating the production of acids and dissolving metals [4]. Equation 1 indicates that pyrite is oxidized when exposed to oxygen and water, resulting in release of hydrogen ions (acidity source), sulphate ions, and soluble ferrous iron cations. This oxidation reaction occurs in undisturbed rock at a slow rate, and the water will buffer the acid produced during the reaction. The following chemical reactions from (1) to (4) represent water and oxygen weathering pyrite AMD formation.

$$FeS_2 + 7/2\ O_2 + H_2O \rightarrow Fe^{2+} + 2SO_4^{2-} + 2H^+ \quad (1)$$
$$Fe^{2+} + ¼\ O_2 + H^+ \rightarrow Fe^{3+} + ½\ H_2O \quad (2)$$
$$Fe^{3+} + 3H_2O \rightarrow Fe\ (OH)^{3}\ ^{+}\ 3H^+ \quad (3)$$
$$FeS_2 + 14Fe^{3+} + 8H_2O \rightarrow 15Fe^{2+} + 2SO_4^{2-} + 16H^+ \quad (4)$$

The process of equations 2 and 3 is perpetuated when ferrous iron is formed from equation 4 and enough dissolved oxygen is present [5]. Equation 4 will continue to complete without dissolved oxygen and water will indicate elevated levels of ferrous iron [5]. High concentration of Fe^{2+} ion is released into the surroundings once acid drainage is formed. For analyze the removal of ferum ion from Acid Mine Drainage (AMD) wastewater by jar test technique, a factorial experimental design approach was used. Jar testing could lead to cost savings in chemicals at the mine, as wastes are planned so it is costless and in the lime plant waste disposal[6–8]. From the other hand, adsorption is considered an ideal method because of its advantage, ease of operation, low operating costs and simplicity of design. The literature suggests the application of various natural and synthetic adsorbents to remove ferum ion from wastewater. For this analysis, the AMD treatment uses Basic Oxygen Furnace (BOF) slag which is a waste from the steel industry. Slag is a powdery substance with a limited hydraulic reactivity, high in alkali and thus capable of neutralizing the acidic state due to the majority of calcium and magnesium oxides [9]. To build an improved method of adsorption using BOF slag, screening factors and fit model are used to achieve the best operating conditions resulting in either maximum or minimum response using Response Surface Methodology (RSM) consisting of 2-Level Factor Modeling (FFD) by Design Expert 7. Four treatment variables (factors) were analyzed and applied for 2-level factorial design. The research variable was concentration, slag dosage, pH value and interval time. The response is for Fe^{2+} final concentration. This response value will be added in equation 5 which is an adsorption capacity for equilibrium.

$$q_e = (C_o - C_e)V/m \tag{5}$$

q_{e} = (mg/g) is the equilibrium adsorption capacity
C_{o}=initial concentration
C_{e}=final concentration
V=(L) volume of aq solution containing respond ions
m= (g) weight of BOF slag

The Langmuir adsorption isotherm is a very simple Type I model. The different reactions were reported in these experiments since certain amounts of BOF slag can be extracted in accordance with Standard A and Standard B [10]. Concentration value until 1000ppm because BOF slag dosage using in this experiment is only small amount, which is maximum amount is 2g [7]. One of the aims of this study is to apply a two-level factor design to evaluate the impact of different parameters and their interactions on the removal efficiency of ferum and then to assess the value of BOF slag as an adsorbent for removing ferum ion from AMD wastewater. Additionally, studies of regeneration were carried out to estimate the potential of this cycle in industrial applications.

2. Methodology

Standard jar test was carried out using adsorption process as shown in Figure 1. In the experiment a conventional jar test tool (JLT6 Jar Test, VELP Scientifica) was used to treat AMD water with the adsorbent. It was conducted as a batch study, accommodating a set of six beakers along with six paddles in spindle steel. There were four processing variables (factors) that were studied and applied for 2-level factorial design. The variable investigate were concentration, slag dosage, pH value and interval time.

Table 1 shows the selected factors and levels to be employed for the experiment. A total sixteen experiment runs were conducted. Factor levels were coded as -1 (low level) and +1 (high level) where low level indicates the lowest range of the factors and high level indicates the highest range of the factors. The samples was mix homogeneously before fractionated into the beakers containing 100 ml per test and 150 rpm per run of sample each [7]. The beakers were agitated at equal speeds of 150 and 250 rpm after the desire quantity of adsorbents was applied to the AMD water. A sample may use a pipette from the top inch of supernatant to test with a drawer, which represents the final concentration.

For Design Expert 7 statistical software specifically Two Level Factorial Design (FFD) the performance of the experimental design was evaluated for assessing the effects as well as the statistical parameters. The answer was evaluated using ANOVA based on the p-value to be appropriate with a confidence level of more than 90 percent. Figure 2 show graphical abstract removal of ferum from acidic solution for this experiment.

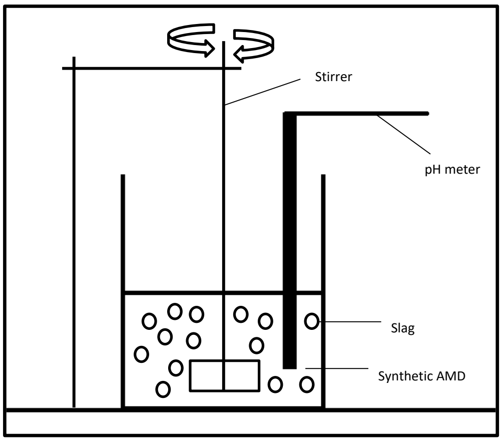


Figure 1. Schematic diagram for standard jar test.

Table 1. The design arrangement and levels of factors.

Run Order	Factor 1	Factor 2	Factor 3	Factor 4
	Slag Dosage	Stirration speed	Interval Time	Concentration
1	0.20	150	5.00	200
2	2.00	150	5.00	200
3	0.20	250	5.00	200
4	2.00	250	5.00	200
5	0.20	150	100.00	200
6	2.00	150	100.00	200
7	0.20	250	100.00	200
8	2.00	250	100.00	200
9	0.20	150	5.00	1000
10	2.00	150	5.00	1000
11	0.20	250	5.00	1000
12	2.00	250	5.00	1000
13	0.20	150	100.00	1000
14	2.00	150	100.00	1000
15	0.20	250	100.00	1000
16	2.00	250	100.00	1000
Factor	Level			
	-1	+1		
Slag dosage	0.5	5.00		
Stirration speed (rpm)	150	250		
Interval time (minute)	5	100		
Concentration of Ferum (II) (ppm)	200	1000		

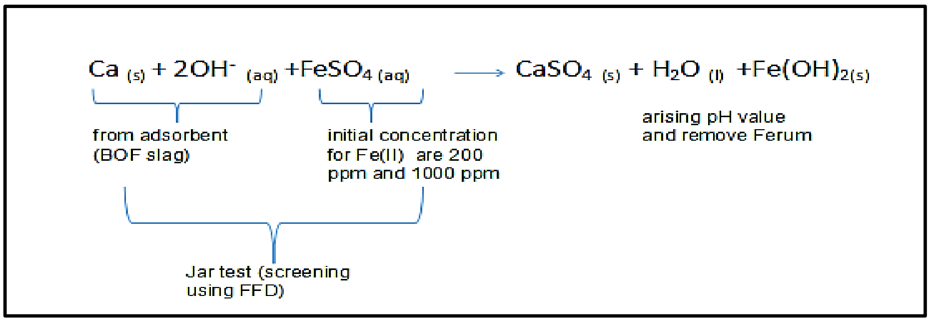


Figure 2. Removal of ferum from acidic solution for this experiment.

3. Results and discussion

3.1. Analysis of variance

FFD's results were evaluated using the windows design expert 7. The key effects were calculated, as well as interaction between variables. The effect of a factor is the response change, here the percentage of ferum ion removal generated by a factor level change, slag dosage, stirration speed, interval time and initial concentration of "Fe^{2+}", from lower to higher. The codified pattern used for 16 factorial designs was

$$q_{e=} 31.21 + 15.8A - 0.57B - 5.33C - 12.76D + 6.35AB + 5.27AC - 18.38AD + 3.91BC - 13.93 -8.00CD + 10.26BCD \qquad (6)$$

where the slag dosage represents A, the speed of stirrings is B, the interval time is C and the concentration is D. Table 2 shows the effect, the regression coefficient, standard errors and P. The key effects for each of them reflect variations of the mean between high and low levels.

If a factor's effect is positive, the percentage removal rate increases as the factor moves from low to high. Additionally, if the results are negative, a drop in percentage elimination occurs for the same factor's high point. Replacing the regression coefficients in equation (7) we obtain model equation relating parameter level and metal removal:

$$q_e = - 104.02977 + 13.58708A + 0.50509B + 0.97186C + 0.29909D + 0.1415AB + 0.12335AC - 0.051057A - 4.83566E - 003BC - 1.2636BD - 0.00258CD + 0.00.1.08020E - 005 BCD \qquad (7)$$

Analysis of variance for the adsorption regression model are shown in Table 3. The single factor's most important value is shown from p value that is less than 0.05 [11]. For both SD and R^2, it should be noted that from an engineering perspective this is a suitable layout. The F-value function of 33.52 suggests the function shown in Table 2 was important. The goal of factorial design on 2-level was to minimize the number of factor. The design gives the final results for the factors affecting the response. In this case, important modeling concepts were A, C, D, AB, AD, and CD. Values above 0.1000 suggest that the terms of the model weren't significant.

Table 2. Experimental data obtained from using 2-level factorial design.

Run Order	Factor 1	Factor 2	Factor 3	Factor 4	Response
	Slag Dosage	Stirration speed	Interval Time	Concentration	q_e (mg/g)
1	0.20	150	5.00	200	2.84
2	2.00	150	5.00	200	4.04
3	0.20	250	5.00	200	22.69
4	2.00	250	5.00	200	9.83
5	0.20	150	100.00	200	5.34
6	2.00	150	100.00	200	7.39
7	0.20	250	100.00	200	7.94
8	2.00	250	100.00	200	9.94
9	0.20	150	5.00	1000	384.88
10	2.00	150	5.00	1000	21.96
11	0.20	250	5.00	1000	5.93
12	2.00	250	5.00	1000	2.52
13	0.20	150	100.00	1000	19.051
14	2.00	150	100.00	1000	3.54
15	0.20	250	100.00	1000	8.81
16	2.00	250	100.00	1000	3.90

Factor	Level	
	-1	+1
Slag dosage	0.2	2.0
Stirration speed (rpm)	150	250
Interval time (minute)	5	100
Concentration of Ferum (III) (ppm)	200	1000

**Analysis of Variance (ANOVA)*

Table 3. Analysis of variance for regression model for adsorption.

Source	Sum of Squares	df	Mean Square	F Value	p-value Prob > F
Model	19663.38	11	1787.58	33.52	0.002
A-slag dosage	4041.46	1	4041.46	75.78	0.001
B-stiration	5.23	1	5.23	0.098	0.7698
C-interval time	455.08	1	455.08	8.53	0.0432
D-concentration	2606.87	1	2606.87	48.88	0.0022
AD	645.54	1	645.54	12.1	0.0254
BC	444.89	1	444.89	8.34	0.0446
BD	5405.56	1	5405.56	101.36	0.0005
CD	244.38	1	244.38	4.58	0.099
BCD	3105	1	3105	58.22	0.0016
Residual	1024.48	1	1024.48		
Cor Total	1684.9	1	1684.9		
Std dev.	7.30	R-squared	0.9893		
Mean	31.21	Adjusted R^2	0.9598		
C.V. %	23.40	Predicted R^2	0.8283		
PRESS	3413.15	Adeq Precision	16.429		

*PRESS = Predicted residual sum of squares.

3.2. Student's t-value test

The main analysis of factors analyzed in the FFD was obtained from Figure 3 of the Pareto charts. According to the Pareto chart, in which the bar below the limit of Benferroni 5.2472 indicates the factor is not important. In this study, for response which is adsorption percent, striation (B) and interval time (C) is considered not significant. Pareto charts in Figure 3 is shown for adsorption percent response.

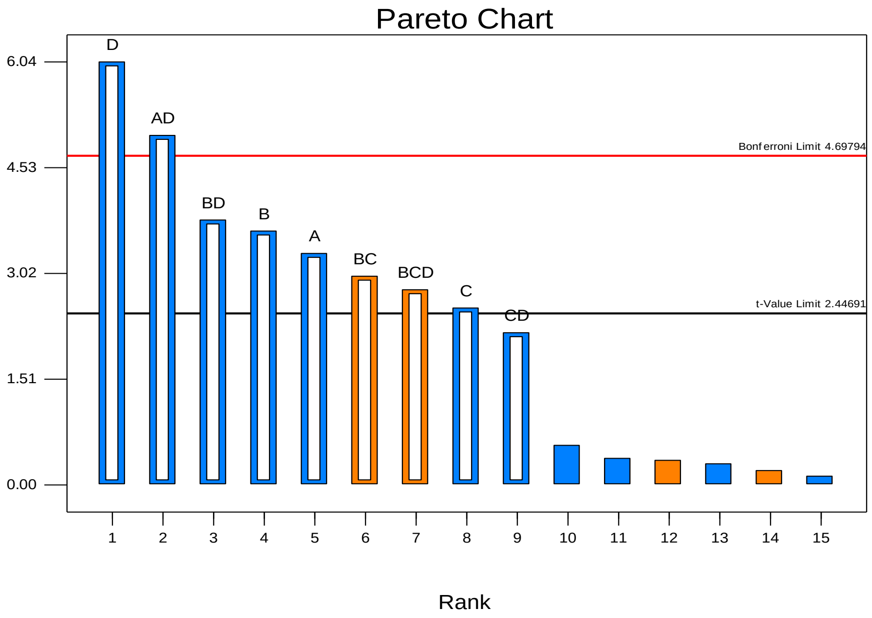


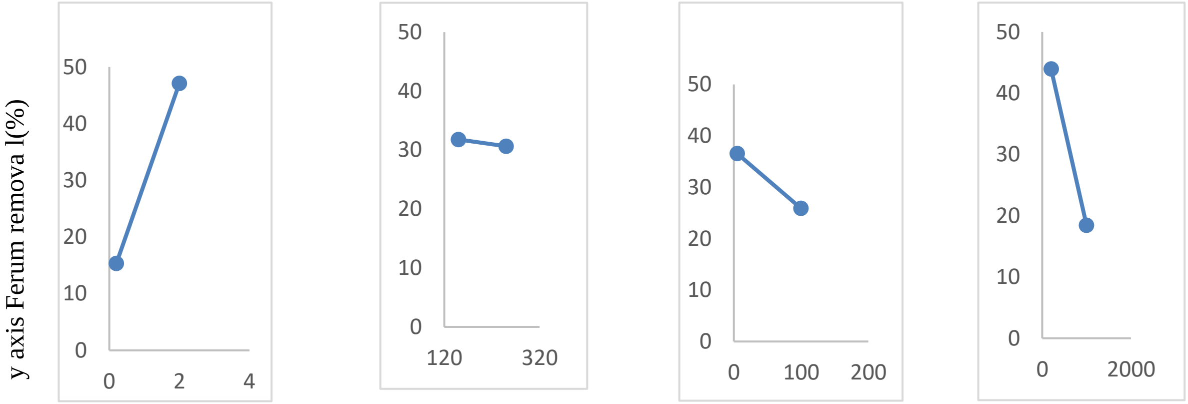
Figure 3. Pareto Chart for adsorption percent

3.3. Affecting on the adsorption value

Slag dosage, concentration, interval time and stirring speed were selected to determine the most effective factors to contribute percentage of adsorption. The initial concentration value for 200 mg/L show the minimum percentage of adsorption is 2.84% and maximum percentage of adsorption is 99.36%. Meanwhile for initial concentration value for 1000 mg/L minimum percentage of adsorption is 1.19% and maximum percentage of adsorption is 76.98%. For 200 mg/L solution, minimum percentage of adsorption is only 2.84% because the slag dosage only 0.20 g, meanwhile maximum percentage of adsorption is 99.36% because of 2.0 g slag dosage. This is similar to result from [12], adsorption is high for slag more than at 10g in 100 mL AMD solution. This is show more volume of slag is needed to remove ferum in AMD.

3.4. Main effects and interaction effects of the factors

The highest percent contribution factors and interaction factors as a model were selected. The analysis of the experimental responses revealed that slag dosage and concentration had significant interactive effects on the adsorption. The plot interaction effects and main effects indicate mean response values for different factors in Figure 4.



slag dosage (g)

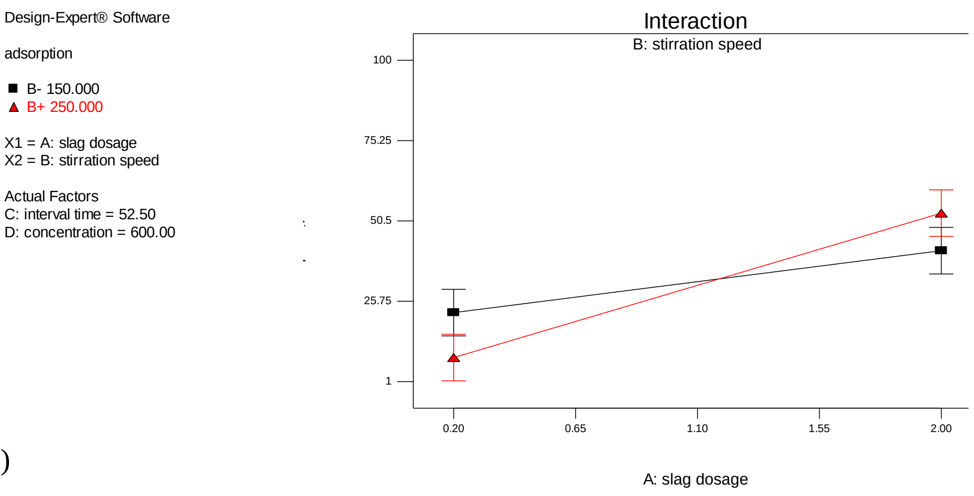
Stirration speed (rpm)

Interval time (min)

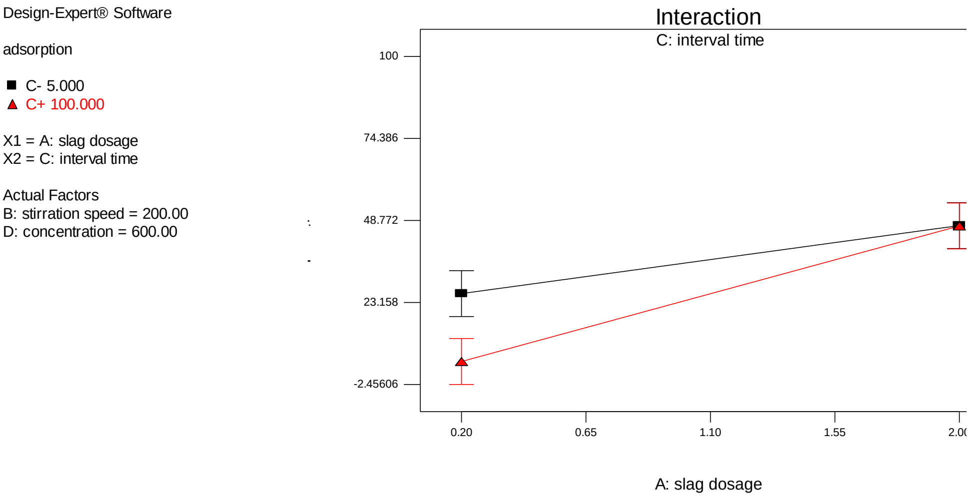
Concentration (ppm)

Figure 4. Main effect plot for ferum removal for y axis vs interaction effect for x-axis.

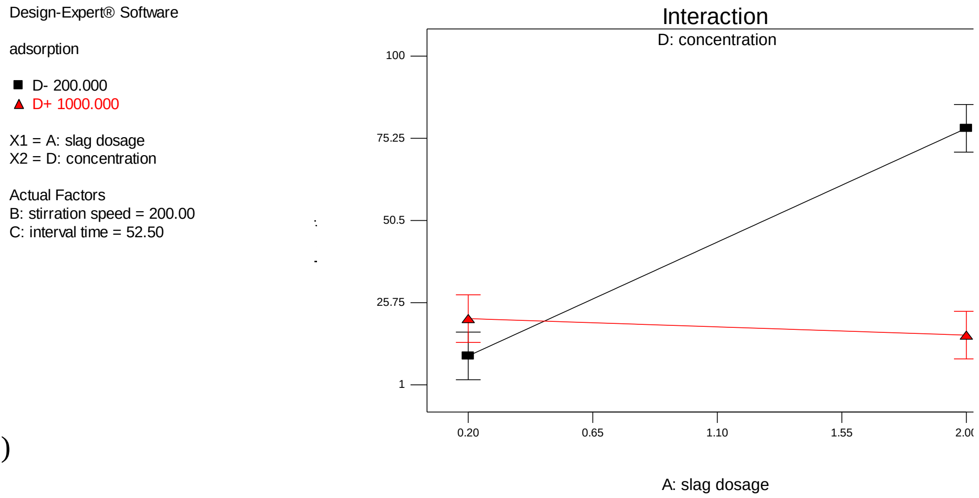
Figure 5 (a) show the interaction between slag dosage and striation speed. It find the adsorption is high when slag dosage is 2.0 g for concentration 200ppm in each experiment due to the more slag use, which resulting a better removal of ferum ion. However, for Figure 5 (c), (e) and (f) for high concentration of ferum adsorption is low when all factor using are high. The Model F-value of 33.52 means the model is relevent. There is only a 0.20% chance that a "Model F-Value" this wide will happen because of noise.


Design-Expert® Software
adsorption
■ B- 150.000
▲ B+ 250.000
X1 = A: slag dosage
X2 = B: stirration speed
Actual Factors
C: interval time = 52.50
D: concentration = 600.00
Interaction
B: stirration speed
100
75.25
50.5
25.75
1
0.20
0.65
1.10
1.55
2.00
A: slag dosage

(a)


Design-Expert® Software
adsorption
■ C- 5.000
▲ C+ 100.000
X1 = A: slag dosage
X2 = C: interval time
Actual Factors
B: stirration speed = 200.00
D: concentration = 600.00
Interaction
C: interval time
100
74.386
48.772
23.158
-2.45606
0.20
0.65
1.10
1.55
2.00
A: slag dosage

(b)


Design-Expert® Software
adsorption
■ D- 200.000
▲ D+ 1000.000
X1 = A: slag dosage
X2 = D: concentration
Actual Factors
B: stirration speed = 200.00
C: interval time = 52.50
Interaction
D: concentration
100
75.25
50.5
25.75
1
0.20
0.65
1.10
1.55
2.00
A: slag dosage

(c)

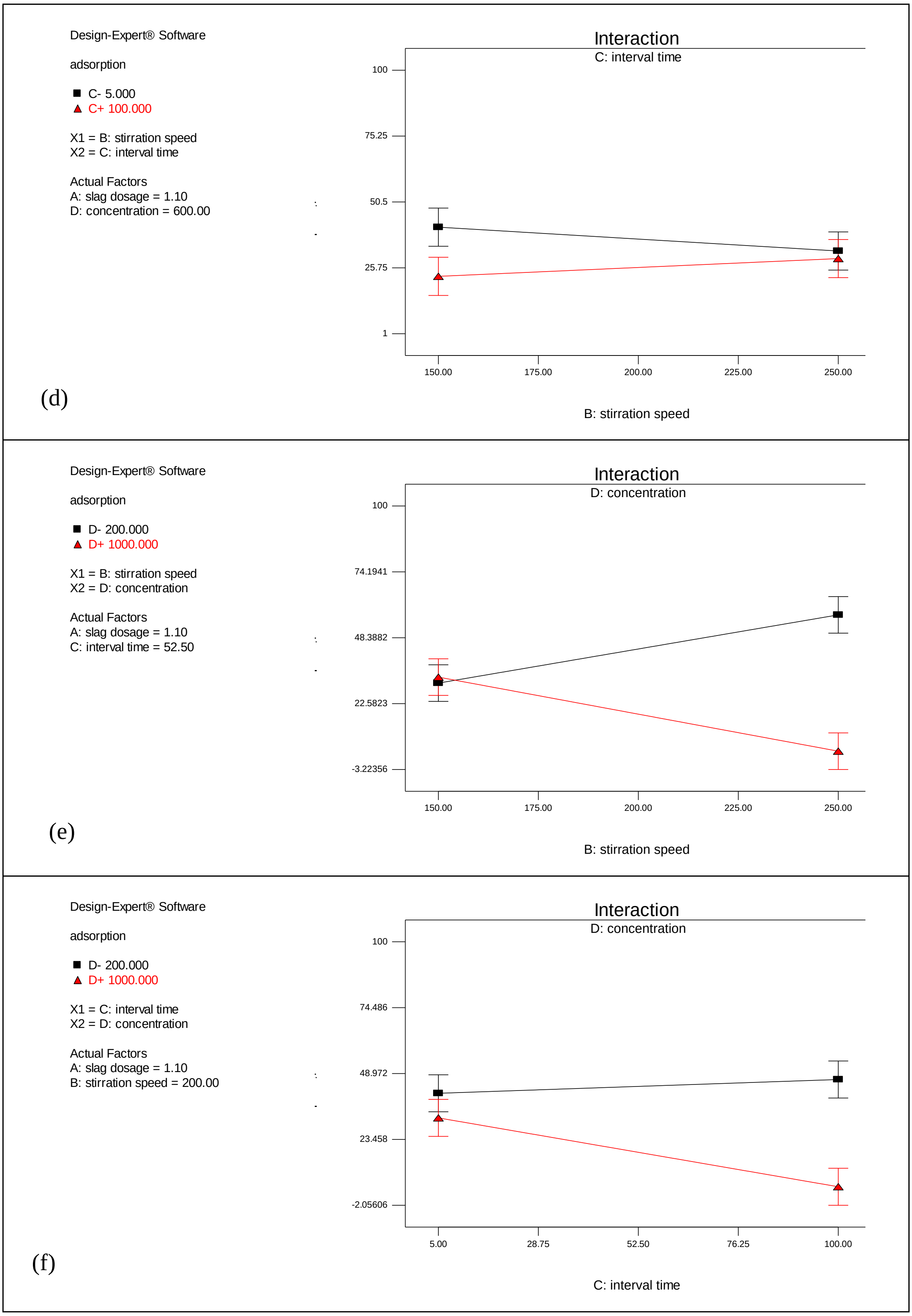


Figure 5. The interaction effect graph between all factors (a) between slag dosage and striation speed, (b) between slag dosage and interval time, (c) between slag dosage and concentration, (d) between interval time and striation speed, (e) between striation speed and concentration, (f) between interval time and concentration.

4. Conclusion

The results of the present study show that jar test adsorption is successful for the early stage of AMD treatment in ferum ion removal. This experiment showed that basic oxygen furnace (BOF) slag can be used as an AMD treatment because of its ability to minimize the amount of ferum ions in AMD water. BOF slag is a byproduct from the steel industry can be used as an alternate source for treating AMD. Jar test method for active treatment is very simple and easy to perform. The weight parameters used in this study are optimal for AMD treatment, but the best selected parameter is 2.0 g BOF slag for treating 200 ppm AMD wastewater concentration.

Acknowledgements

The study was funded through grants PGRS180329 and RDU 1703170 Internal Grant UMP.

5. Reference

[1] Johnson D B and Kevin B H 2005 *Sci. Total Environ*. **338** 3–14.
[2] Erkan S, Nesrin D, Bestamin O and Anna H. K. 2013 *Miner. Eng*. **48** 56–60.
[3] Anita P F and Bernd G L 2015 *Miner. En*g. **82** 107–124.
[4] Bigham J M and Cravotta C A 2016 Acid mine drainage 6-10
[5] Akane I, Tsubasa O, K.S, Kamar S. A, Fei-Y. Y, Tsutomu S 2017 *Malaysia*, 82 89–101
[6] Emma T T, Arja S, Tao H, Jaakko R, Ulla L 2014 *Chemosphere* **117** 419–424.
[7] Anuar O, Azli S, Shamsul K S 2017 *J. Water Process Eng.* **15** 31–36.
[8] Moodleya I, Sheridana C M and Kappelmeyerb U, Akcil A 2017 *Miner. En*g. 1–14.
[9] Nur Athirah M B and Wan Zuhairi W Y 2014 *IP Conference Proceedings* 1614, 808
[10] Environmental Quality Act (EQA), 1974. Laws of Malaysia. Act 127. International Law Book Services
[11] Guohui S, Xin Z, Zixian B, Xuqian L, Zhongzheng Z, Yang L and Chao F 2018 *Carbohydr. Polym*. **189** pp 280–288
[12] Vhahangwele M, Muhammad S O, and Adnan M A M 2017 *Desalination* **424** 45–52