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Response Surface Methodology For Optimization of Sodium Sulphate And Sodium Lauryl Sulphate Removal Using Hydroxyapatite Adsorbent

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Abstract: Sodium sulphate and sodium lauryl sulphate is contaminant that can be found in surface water resulting from agriculture, industrial, or domestic waste discharge. Sulphate can cause acidic condition to soils and form acid rain that damage plants and buildings. It can also cause laxative effect on humans and livestock. This study is conducted to optimize sulphate removal by hydroxyapatite adsorbent through the application of response surface methodology (RSM). The rate of adsorption on sulphate removal was mathematically describe as a function of experimental parameters and was modelled through RSM via historical data design. The results show that that the response of removal was significantly affected by quadratic term of pH and contact time. The statistical analysis was performed by ANOVA which indicated good correlation of experimental parameters with R2 of 0.98. The optimum operation condition for sodium sulphate was obtain at 26.83 min and pH 7.12, while for sodium lauryl sulphate was at 26.41 min and pH 7.18. The predicted removal was found to be relatively close to experimental removal for sodium sulphate were 60.0 % and 59.0 %, while for sodium lauryl sulphate were 70.0 % and 69.0 %, respectively.

Keywords: Sulphate, Hydroyapatite, Response surface methodology, pH, Contact time

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1. Introduction

Sulphate is one of the contaminant that can be toxic to freshwater plants and animals. It can make water unpalatable for safe consumption. Human waste, industrial wastewater, and fertilizer application had contribute to high levels of sulphate in the water sources. In plants systems, it can lead to suppressed growth and development, iron chlorosis, leaf necrosis, suppressed flowering, black and flaccid roots, root decay and even the death of the entire plant. Furthermore, sulphate also effect in our gastroenteritis body such as diarrhoea, nausea and inflammatory bowel disease. High concentration of sulphate had been detected in the stiped bass and amphipod in the water bodies of North America,. Therefore, many research use the alternative way to applied natural waste to produce an efficient and low cost treatment for sulphate.

Adsorbent is one of the alternative method to remove sulphate in water as it easy to produce and cheap. Adsorbents have a highly porous structure, allowing the adsorption of a wide range of target compounds in large quantities. Adsorbent can be classified into natural adsorbent and synthetic adsorbents. Fish waste are waste that can be acquire abundantly from the fish processing industry. It had the potential to be used as adsorbent by synthesized it into hydroxyapatite (HAp) nanopowder. HAp constituted of calcium phosphate that exhibits similar structure and composition to the mineral phase in bones and teeth [1].

Response surface methodology (RSM) is a mathematical and statistical method commonly used for modeling and analyzing process in which the interest response is determined by various variables. This method is intended to optimize the response. Independent variables are the parameters that influence the operation, while response is the dependent variable. The main aims of an RSM is to understand the topography of the response surface, including the local maximum, local minimum and ridge lines. The RSM investigates a suitable approximating relationship between input and output variables and determines the ideal operating conditions for an investigated system or a factor area that meets operational requirements. This study employed RSM via historical data to obtain optimum operating conditions for sodium sulphate (NA_2SO_4) and sodium lauryl sulphate (SLS) removal using HAp adsorbent.

2. Materials and Methods

2.1 Design-Expert tools

Design-Expert is a tool used for product and process improvements. It is used to monitor for critical factors, identify interactions and eventually achieve optimum process settings for the product. It can explore contours on interactive 2D graphs and visualize the response surface from all angles with rotatable 3D plots. Besides that, this tool can find the best model, value of regression (R), and maximize desirability for all responses simultaneously. Design-Expert also provide a powerful tool to lay out an ideal experiment on process, mixture or combination of factors and components. The tool makes easy to see the statistical significant and how to model the results most precisely with wide selection of graphs that help to identify standout effects and visualize results [5].

2.2 Response surface methodology (RSM)

Response surface methodology (RSM) is a technique to determine design factor settings to improve or optimize the performance of a process or product using Design-Expert software from State Ease Inc. RSM help with the design and interpretation of multi-factor experiments and for optimizing the prediction of model.

2.3 Historical data design

Historical data design is used for importing data that already exists. For using historical data, the total of numeric factors and rows depends on the previous research data.

2.4 ANOVA

Analysis of variance (ANOVA) emphasize on active factors that show as a collection statistic on models. It also show the summary statistics such as adeq precision, code equation, and significant models [5]. The fit statistics is the modeling statistics for a model design. In fit statistics its shows the value on standard deviation, mean, coefficient of variation, adjusted R- squared, Predicted R-squared and adequate precision. The value of R^2 must greater than 0.9 to shows the value is reasonable. The adequate precision is a signal to noise ratio that measures the ratio of the range of variation in predicted response to an estimate of standard error of the predictions. A high value indicated that the variation observe is large in relation to the underlying uncertainty of the fitted model.

2.5 Verification of model adequacy

Verification of model adequacy is important for regression models that are used for prediction or estimation. Model adequacy is the determination if the model will function successfully in its intended operating environment.

2.6 Three-dimension surface plot

The three-dimensional response surface plots were generated to estimate the effect of the combinations of the independent variables on the response [6].

2.7 Process optimization

Process optimization in RSM is typically done to reduce or eliminate time and resource wastage. In this study, the process optimization of the model was to obtain the optimum operational conditions for pH and contact time toward maximum NA_2SO_4 and SLS removal percentage.

3. Results and Discussion

3.1 Historical Data design of RSM

The Response Surface Methodology (RSM) is use to optimize a process. It is used to explore the effect of operating conditions (the factors) on the response variable. For this study, the historical data design is chosen as method that is suitable for importing the historical or previous research data. Table 1 shows the data use in this study [5]. There are two factors that effects the percentage removal of NA_2SO_4 and SLS which are pH and time.

Table 1: Historical data from previous study

	Factor		Percentage Removal	
std	pH	Time	NA_2SO_4 (%)	SLS (%)
1	2	1	30	38
2	3	3	39	41
3	4	5	45	50
4	5	15	50	55
5	6	20	55	60
6	7	23	59	69
7	8	25	49	58
8	9	30	39	41

3.1 Analysis of Variance (ANOVA)

Statistical testing of the model was performed using the Fisher's statistical test for analysis of variance (ANOVA). Table 2 shows the ANOVA for NA_2SO_4 removal efficiency by HAp adsorbent. The significance of each model was assessed from the coefficient (R^2) which is 0.9864. The high coefficient of determination between the predicted and experimental adsorption activities confirmed that the linear model was adequate to explain the actual relationship between the response and the significant variables. The F statistic value of 96.56 implies that the model is significant and there is only a 0.03 % chance that the model could occur due to noise. The significance of each term in this model is also evaluated by P value (Prob > F) that was less than 0.05. This was supported by a P-value which confirmed that 2FI model was statistically significant and sufficient to explain the actual relationship between the response and the significant variables.

Table 3 show the ANOVA for SLS removal efficiency by HAp adsorbent. The. Significance of each model was assessed from the coefficient (R^2) which is found to be 0.9880. The high coefficient of determination between the predicted and experimental adsorption activities confirmed that the 2 FI model was adequate to explain the actual relationship between the response and the significant variables. The F statistic value of 109.98 implies that the model is significant and there is only a 0.03 % chance that the model could occur due to noise. The significance of each term in this model is also evaluated by P value (Prob > F) that was less than 0.05. This was supported by a P-value which confirmed that the quadratic model was statistically significant and sufficient to explain the actual relationship between the response and the significant variables.

Table 2: ANOVA for NA_2SO_4 removal efficiency by HAp adsorbent

Source	Sum Squares	Degrees Freedom	Mean Square	F Value	Prob > F (P-value)	
Model	464.48	3	154.83	96.56	0.0003	Significant
A : pH	89.25	1	89.25	55.66	0.0017	
B : Time	100.73	1	100.73	62.82	0.0014	
AB	337.75	1	337.75	210.64	0.0001	
Residual	6.41	4	1.60			
Corrected Total	470.89	7				

R^2 = 0.9864, Coefficient of variation = 2.76 , Adjusted R^2 = 0.9762 , Predicted R^2 = 0.9465 , Adeq precision = 22.4277

Table 3: ANOVA for SLS removal efficiency by HAp adsorbent

Source	Sum Squares	Degrees Freedom	Mean Square	F Value	Prob > F (P-value)	
Model	1261.31	3	420.44	109.98	0.0003	Significant
A : pH	114.14	1	114.14	29.86	0.0055	
B : Time	146.55	1	146.55	38.34	0.0035	
AB	690.33	1	690.33	180.58	0.0002	
Residual	15.29	4	3.82			
Corrected Total	1276.60	7				

R^2 = 0.9880, Coefficient of variation = 3.51 , Adjusted R^2 = 0.9790 , Predicted R^2 = 0.9412 , Adeq precision = 26.0240

The best fitting model was established by a regression analysis. The following Equation 4.1 and 4.2 are regression model with the experimental results in terms of coded factors:

$$Y = 66.98 – 57.69A + 59.26B – 28.66AB \ (NA_2SO_4) \qquad \text{Eq. 1}$$

$$Y = 83.97 – 65.24A + 71.47B – 40.98AB \ (SLS) \qquad \text{Eq. 2}$$

Where Y is the percentage of sulphate removal (%), A is the effect of pH, and B is the effect on contact time (min)

3.2 Verification of model adequacy

Figure 1 and 2 shows the predicted vs actual values for the removal of NA_2SO_4 and SLS. The predicted versus actual values for NA_2SO_4 and SLS removal show straight linear relationship. The correlation coefficient for both plot is 0.9865, which indicates that the model is able to give a good estimate of response. The predicted values were distributed consistently near to the actual responses and this has proven the generated regression models can well explain the relationship between the independent variables and the response. The data are distributed normally in a straight line thus the error is insignificant within the range of operating parameters.

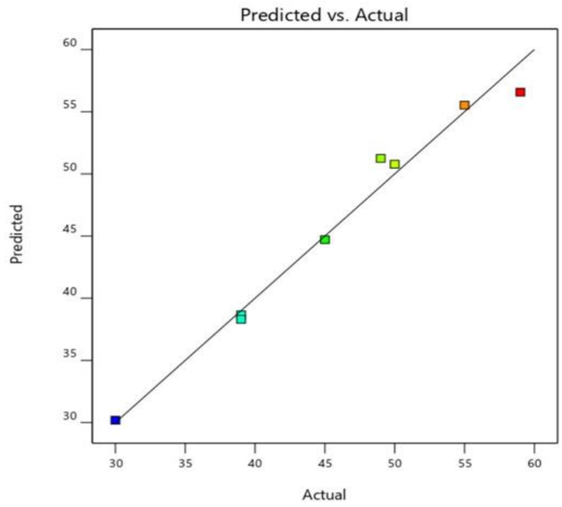

Figure 1: Predicted vs Actual values on NA_2SO_4 removal

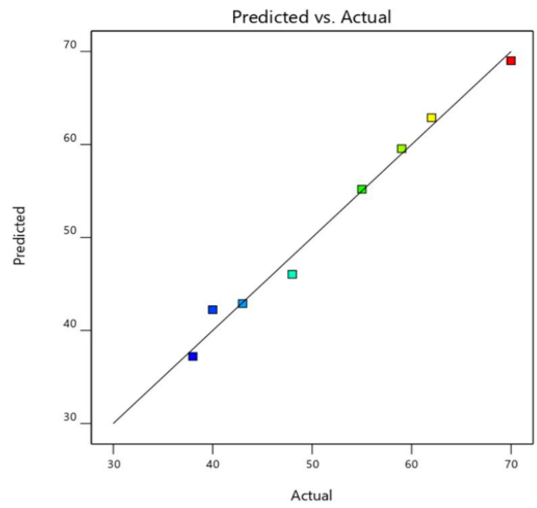

Figure 2: Predicted vs Actual values on SLS removal

3.4 Effect of pH and time NA_2SO_4 and SLS removal

Figure 3 shows the three-dimensional plot for the effects of pH and time on NA_2SO_4 removal. The increase in contact time and pH had increase the NA_2SO_4 removal. The removal of NA_2SO_4 started to increase from pH 2 and achieved the maximum removal of 59.0 % at pH 7 in 23 min. After that, the removal drop to 39.0 % at pH 9 in 30 min.

Figure 4 shows the three-dimensional plot for the effects of pH and time on SLS removal. The decrease in contact time and increase of pH had increase the SLS removal. The removal SLS started to increase from pH 2 and achieved the maximum removal of 69.0 % at pH 7 in 1 min. After that, the removal drop to the lowest of 41.0 % at pH 9 in 30 min.

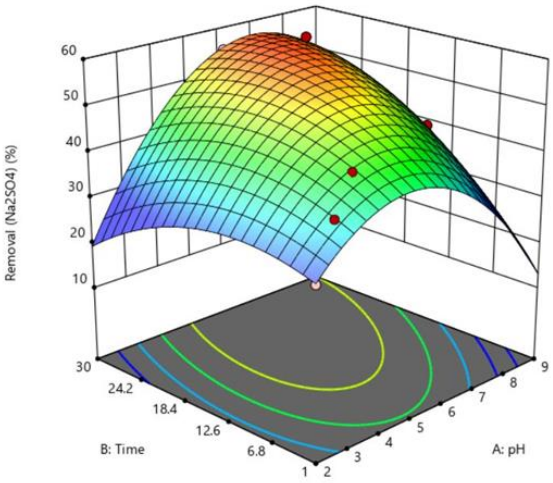

Figure 3: Effects of pH and time on the removal of NA_2SO_4

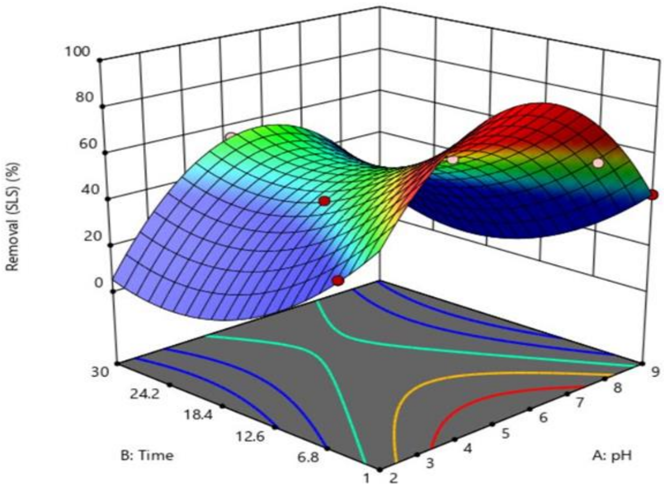

Figure 4: Effects of pH and time on the removal of SLS

3.5 Process optimization

The RSM model capable to predict the maximum adsorption for the removal of sulphate. The optimum condition for NA_2SO_4 and SLS removal is shown in Table 4. The optimum operating condition for NA_2SO_4 removal is at 26.83 min and pH 7.12. The experimental and predicted removal efficiency at the optimum condition was 59.0 % and 60.0 %, respectively. The optimum operating condition for SLS removal is at 26.41 min and pH 7.18. The experimental and predicted removal efficiency at the optimum condition was 69.0 % and 70.0 %, respectively. The predicted removal was found to be relatively close to the experimental removal, confirming the eligibility of the linear model suggested by the software. Therefore, the experimental design using RSM could be effectively applied to optimize the NA_2SO_4 and SLS removal using HAp adsorbent.

Table 4: Optimum conditions for NA_2SO_4 and SLS removal

Optimum conditions	pH	Time (min)	Experimental removal (%)	Predicted removal (%)
NA_2SO_4	7.12	26.83	59	60
SLS	7.18	26.41	69	70

4. Conclusion

The optimum conditions of pH and contact time were found at pH 7.12 and contact time of 26.83 min for NA_2SO_4 removal while for SLS was at pH 7.18 and contact time of 26.41 min. The predicted NA_2SO_4 removal of 60.0 % at the optimum condition was close to the experimental value of 59.0 %. Whereas the predicted SLS removal of 70.0 % was also close to the experimental value of 69.0 %. The findings shows good agreement between the actual and predicted values. This study demonstrate that RSM is a useful and practical tool for optimization of sulphate removal process by HAp adsorbent.

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