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Optimization of Oil Removal by Sugarcane Bagasse using Response Surface Methodology

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Abstract

Oily wastewater is one of the environmental concerns nowadays. The seriousness of oil pollution problem comes in sync with the expansion of oil exploration and production activities, as well as industrial growth around the world. In this study, the ability of sugarcane bagasse in removing oil in synthetic oil wastewater was investigated. Parameters affecting oil removal such as concentrations of synthetic oil wastewater, biosorbent dosage and contact time were optimized using Response Surface Methodology (RSM) via Box Behnken Design. Sugarcane bagasse showed excellent efficiency in removing oil with percentage removal up to 98.73% at 1.3 h contact time with 3.06 g of biosorbent dosage and 16.9% of synthetic oil wastewater concentration. The use of sugarcane bagasse in removing oil in water was successfully prove in this study.

1. Introduction

Oils in wastewater can be found in various forms such as fats, lubricants, heavy hydrocarbons, cutting oils, and light hydrocarbons (Srinivasan and Viraraghavan, 2008). Oil and its derivatives are some of the products with high polluting potentials, because they are very difficult to undergo biodegradation and also being stable to light and heat (Obuekwe, 2009). Adsorption process is effective in condensing and concentrating oil from aqueous phase to the surface of adsorbent, it is a well-established technology that employed the use of synthetic adsorbent that are usually scarce and expensive in wastewater treatment. Hence, there is a need to develop an adsorbent that are readily available at low cost to remove oil contamination in wastewater. Therefore, the use of sugarcane bagasse as an oil adsorbent will be alternative cheap treatment method due to a lot of excessive of sugarcane bagasse in our environment. Some people just burn the sugarcane bagasse and will contribute to air pollution. In this study, the efficiency of sugarcane bagasse for the removal of oil from synthetic oil wastewater was investigated.

2. Materials and Methods

2.1. Materials

Cooking oil was obtained from the local store in Jeli area. All chemicals used were analytical grade. N-Hexane (95% pure, Bendosen) was used as the solvent in this experiment. Sugarcane bagasse were collected from local stall at Jeli, Kelantan.

2.2. Preparation of Untreated Sugarcane Bagasse (USB)

The raw sugarcane bagasse were washed using distilled water and dried in an oven (Model FD115, Binder, Korea) at 70˚C for 7 days. The dried sugarcane bagasse were then cut into small pieces, and grounded by using a blender (Model ELB-A1812G(SS)) to a size of 200-400 μm size fraction and used in the adsorption test.

2.3. Preparation of Treated Sugarcane Bagasse (TSB)

The raw sugarcane bagasse were washed using distilled water and dried in an oven (Model FD115, Binder, Korea) at 70˚C for 7 days. The dried sugarcane bagasse were then cut into small pieces, and ground using a blender (Model ELB-A1812G(SS)) to a size of 200-400 μm size fraction. It was then soaked in 1M of Sulphuric Acid (H2SO4) for 24 hours. After that, the sugarcane bagasse were washed with distilled water once and soaked again with 1M sodium bicarbonate for an overnight. It was dried for 24 hours at 70˚C before starts the experiment.

2.4. Preparation of Synthetic Oil Wastewater

The synthetic oil wastewater was prepared by adding the cooking oil with distilled water. The stock solutions were blended using blender (Model ELB-A1812G(SS)) to mix the solution well. The decided concentration of oil was 50%, 30%, 15.25%, 10%, 5%, 1% and 0.5%.

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2.5. Biosorption Studies

Different biosorption studies were carried out at different contact time, adsorbent dosage and oil concentration to observe on how those parameters influence the biosorption process. All the experiments were conducted at room temperature in Environmental Science Laboratory, UMK. Experiments were conducted in 250 ml of conical flask containing 100 ml of synthetic oil wastewater. Different concentration of synthetic oil wastewater were tested with different quantities of biosorbent for a desired time, in a Shaking Incubator (Model SI-300/300R/600/600R, JIEO TECH,Korea) at speed of 150 rpm. The biosorbents were filtered out from the conical flask using cloth coffee filter after biosorption experiment. The amount of oil before and after the experiment was determined by gravimetric method.

2.5.1. Gravimetric Method

Gravimetric analysis describes a set of methods in analytical chemistry for the quantitative determination of an analyte based on the mass of a solid. In gravimetric method, the remaining synthetic oil wastewater after the filtration was extracted by n-hexane. 4ml of 1:1 hydrochloric acid solution (HCl) was added to adjust the acidity of the sample at pH 2 or less. It was then serially extracted three times with n-hexane in separatory funnel. After the extraction process, the remaining oil obtained was collected in the round bottom flask that has been weighed beforehand and undergo distillation process to remove the n-hexane. The sample was then being placed in the oven at 100˚C for 12 hours to evaporate water out from the sample. The sample (residual oil) was weighed then by using weighing balance (Model EMB 1200-1, Kern, Korea) to get the final reading of the sample. Knowing the weight of empty round bottom flask, the amount of residual oil can be calculated (Yang, 2011).

2.5.2. Screening of Biosorbent

The synthetic oil wastewater with 50% of oil concentration was tested differently with 0.5 g of untreated sugarcane bagasse (USB) and treated sugarcane bagasse (TSB) for 3 hours in a Shaking Incubator (Model SI-300/300R/600/600R, JIEO TECH,Korea) at speed of 150 rpm. At the end of the shaker time, the biosorbents were filtered out from the conical flask using cloth coffee filter. The amount of oil before and after the experiment was determined by gravimetric method.

2.5.3. Determination of oil removal

The percentage (%) removal of oil by the sugarcane bagasse were calculated by the following equation.

$$\text{Percentage removal of oil, \%} = ((Ci-Cf))/Ci \times 100 \quad \text{(Equation 1)}$$

Where C_i is the initial reading of the sample, C_f is the final reading of the sample.

2.5.4. Experimental Design and Optimization Using Response Surface Methodology (RSM)

Three parameters were chosen as the critical variables namely oil concentration, dosage of biosorbents and contact time. The experimental design consisted of three variables with 17 experimental runs. A total of 17 experiments have been employed in this study to estimate the effects of the main variable in this study. According to Box and Behnken (1960), 17 experiments are sufficient to obtain the best response pattern.

The variables and the level selected for this experiments were oil concentrations (0.5%-30%), biosorbents dosage (0.5g-5g) and the contact time (1hour-24 hours). The experiments were randomized conducted by using a software package by Design Expert Version 6.0.8 to fit the second order model. The experimental runs and variables were shown in Table 1.

Table 1: Ranges of the factors investigated using Box-Behnken Experimental Design.

Variables	Symbols	Low	Medium	High
		-1	0	+1
Concentration of oil (%)	A	0.5	15.25	30
Biosorbent dosage (g)	B	0.5	2.75	5
Contact time (hr)	C	1	12.5	24

The quadratic equation model discussed by Marzuki, et al (2015) was used to estimate the optimum value and also to describe the interaction between factors. For RSM only this equation is used to obtain optimal point. The quadratic equation was expressed as Equation 2 (Marzuki et al, 2015):

$$Y = \beta_0 + \sum_{j=1}^{k} \beta_j X_j + \sum_{j=1}^{k} \beta_{jj} X_{j2} + \sum_{i=1}^{j-1} \sum_{j=2}^{k} \beta_{ij} X_i X_j + \varepsilon \quad \text{(Equation 2)}$$

where Y is the response, i and j are the linear and quadratic coefficients, respectively, Xi and Xj are the uncoded independent variables and regression coefficients, k is the number of studied and optimized factors in the experiment, β0 is a constant coefficient, βj, βjj, and βij are the interaction coefficients of linear, quadratic and second order terms, respectively, k is the number of studied factors and ε is the error. The value of correlation coefficient (R2) was used as a tool to express the quality of the fit of the polynomial model. The significance of a model was represented by a p-value < 0.05. Analysis of variance (ANOVA) was also carried out to obtain optimum condition and interaction between two factors. The stated statistical analysis was discussed by Marzuki, et al (2015).

3. Results and Discussion

3.1. Adsorption of Oil Using Treated and Untreated Sugarcane Bagasse

Figure 1 shows the percentage of oil removal by using different types of sugarcane bagasse. The percentage

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removal of oil by using untreated sugarcane bagasse (USB) is 24.91% while using treated sugarcane bagasse (TSB) is 14.09%.

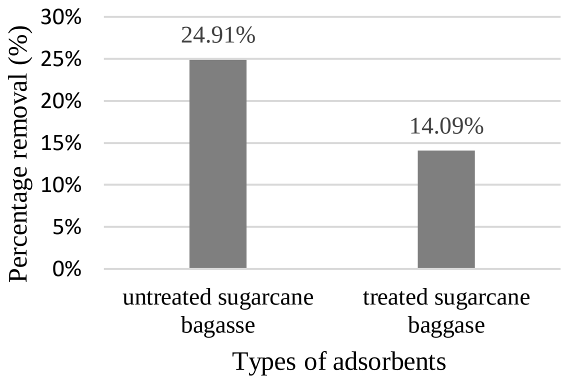

Figure 1: Comparison between the different types of sugarcane bagasse towards the adsorption of oil.

This shows that untreated sugarcane bagasse (USB) gives higher adsorption capability compared to treated sugarcane bagasse (TSB). This is because the untreated sugarcane bagasse (USB) has the texture of origin which is like a sponge and has more pores that allow oil adsorption in it. It is believed the size of pores of untreated sugarcane bagasse smaller compared to the treated one that causes the adsorption process to happen maximally (Wannahari and Nordin, 2011). Therefore, untreated sugarcane bagasse was used for subsequent experiment.

3.2. Response Surface Methodology (RSM)

3.2.1. Experiments and Model Fitting

In this study, the percentage removal of oil in water by using sugarcane bagasse was modelled using the method of RSM, utilizing three reaction parameters: reaction time (h), percentage of oil (%), and the dosage of adsorbent (g). The effect of three experimental factors and their interaction was established and each response surface was plotted as a function of two targeted variables, while other variables were held constant.

The results for the actual values required from the response from the experimental runs and the predicted values obtained from the generated quadratic model (Jaliliannosrati et al., 2013). That corresponded to the percentage removal of oil using sugarcane bagasse are tabulated in Table 2. The actual experimental design matrix which consisted of 17 experiments has been designed by Box-Behnken design as given in Table 2. The percentage removal of oil was found around 31.01% until 100%.

Table 2: Experimental design and results of the response surface design

Std	A: Oil concentration, %	B: Dosage, g	C: Time, hour	Actual percentage removal (%)	Predicted percentage removal (%)
1	0.5	0.50	12.50	60.88	66.97
2	30.00	0.50	12.50	36.65	29.90
3	0.50	5.00	12.50	71.43	78.17
4	30.00	5.00	12.50	40.24	34.14
5	0.50	2.75	1.00	100.00	89.38
6	30.00	2.75	1.00	62.15	64.37
7	0.50	2.75	24.00	100.00	97.78
8	30.00	2.75	24.00	31.08	41.70
9	15.25	0.50	1.00	87.95	92.48
10	15.25	5.00	1.00	97.34	101.22
11	15.25	0.50	24.00	90.24	86.37
12	15.25	5.00	24.00	97.60	93.07
13	15.25	2.75	12.50	84.15	94.90
14	15.25	2.75	12.50	99.60	94.90
15	15.25	2.75	12.50	99.20	94.90
16	15.25	2.75	12.50	98.39	94.90
17	15.25	2.75	12.50	93.17	94.90

3.2.2. Regression Model and Analysis of Variance (ANOVA)

Based on the experiment data and Design Expert Version 6.0.8 a best fitting model was established by a regression analysis (Equation 3). The fitting of the data to various models and their subsequent ANOVA illustrated that the percentage removal of oil was suitably characterized by a quadratic polynomial model. The significant terms and the equation, in terms of coded factors are showed as follows:

$$Y\,(\%) = 94.90 - 20.27A + 3.86B - 3.57C - 31.29A^2 - 11.31B^2 + 9.70C^2 - 1.74AB - 7.77AC - 0.51BC \quad \text{(Equation 3)}$$

where Y is the percentage removal of oil (%), A is the concentration of oil (%), B is the dosage of the adsorbent (g) and C is contact time of the experiments (h).

The statistical significance of ratio of mean square due to regression and mean square residual error was tested using ANOVA. ANOVA is a statistical method that subdivides the total variation in a set of data into component parts linked with specific sources of variation for the purpose of testing hypotheses on the parameter of the model (Rusly et al, 2010). The ANOVA result for percentage removal of oil (%) is shown in Table 3.

The Model F-value of 10.67 implies the model is significant with good R2 of 0.9321. There is only a 0.25%

chance that a "Model F-Value" this large could occur due to noise. Values of "Prob > F" less than 0.050 indicate model terms are significant. In this case A, A2, B2 are significant model terms. Values greater than 0.1000 indicate the model terms are not significant.

The "Lack of Fit F-value" of 3.67 implies the Lack of Fit is not significant relative to the pure error. There is a 12.06% chance that a "Lack of Fit F-value" this large could occur due to noise. Non-significant lack of fit is good.

Table 3: Analysis of variance (ANOVA) and coefficients of the model

Source	Sum of Squares	Degree of Freedom	Mean Square	F-value	Prob > F	
Model	8823.38	9	980.38	10.67	0.0025	Significant
A	3288.32	1	3288.32	35.79	0.0006	
B	119.31	1	119.31	1.30	0.2920	
C	101.75	1	101.75	1.11	0.3276	
A^2	4122.72	1	4122.72	44.87	0.0003	
B^2	538.91	1	538.91	5.87	0.0460	
C^2	395.88	1	395.88	4.31	0.0766	
AB	12.11	1	12.11	0.13	0.7273	
AC	241.41	1	241.41	2.63	0.1491	
BC	1.04	1	1.04	0.011	0.9183	
Residual	634.15	7	91.88			
Lack of Fit	471.77	3	157.26	3.67	0.1206	Not significant
Pure Error	171.37	4	42.84			
Cor Total	9466.53	16				
R^2	0.9321					
Adjusted R^2	0.8447					

3.2.3. Effect of Oil Concentration and Dosage of Adsorbent

Figure 2 shows the effect of oil concentration and adsorbent dosage with constant contact time of 12.50 h. Maximum percentage removal was obtained from all the variables except for experiment 8 based on standard order (Table 2) with only 31.08%.

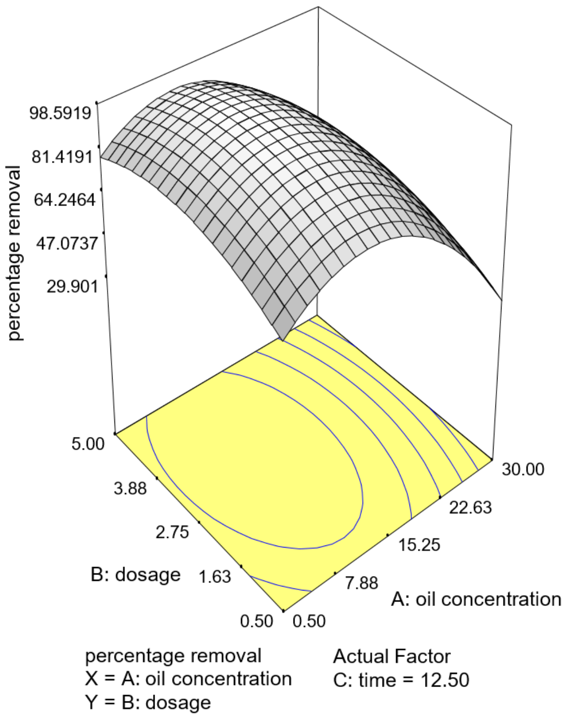

Figure 2: Response surface curve showing the effect of oil concentration (A) and dosage of the adsorbent (B)

The percentage removal decreased with increase in oil concentration. This is due to the increase in availability of active sites surface resulting from the increased dose and accumulation of the adsorbent (Patil et al., 2011). However, at high oil concentration, oil occupies the sorbent surface thus saturation is reached much faster and high amount of unattached oil is left (Huang and Lim, 2006).

3.2.4. Effect of Oil Concentration and Contact Time

As illustrated in Figure 3, the response surface curve is showing the effect of oil concentration and contact time with adsorbent dosage of 2.75 g as the constant factor.

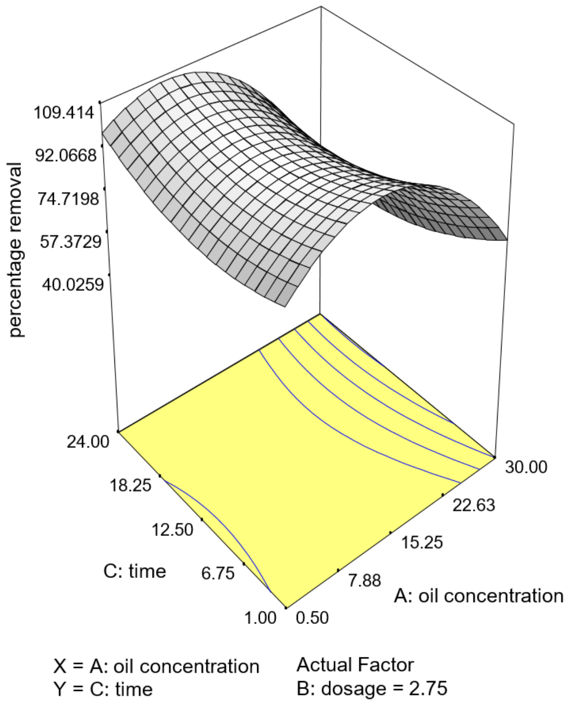

Figure 3: Response surface curve showing the effect of oil concentration (*A*) and the contact time (C)

At 30% oil concentration, the percentage removal of oil after 24 hours contact time gives the lowest result (31.08%) while 62.15% of oil was adsorbed at only 1 hour. This proved that the higher oil concentration need less contact time to allow the adsorption process to occur. For the 0.5% of oil, both 1 hour and 24 hours give the maximum result, which 100% oil removal. This is due to the less concentration of oil, the adsorption can maximumly occur (Alqaragully, 2014).

3.2.5. Effect of Contact Time and Adsorbent Dosage

As displayed in Figure 4, the response surface curve is showing the effect of contact time and adsorbent dosage with oil concentration of 15.25% as its constant variable. Standard 2 and 4 were conducted in 30% of oil concentration after 12.50 hours contact time but different in dosage adsorbent which were 0.5g and 5.0g. The percentage removal of 5.0g adsorbent dosage was higher than 0.5g which were 40.24% and 36.65% respectively.

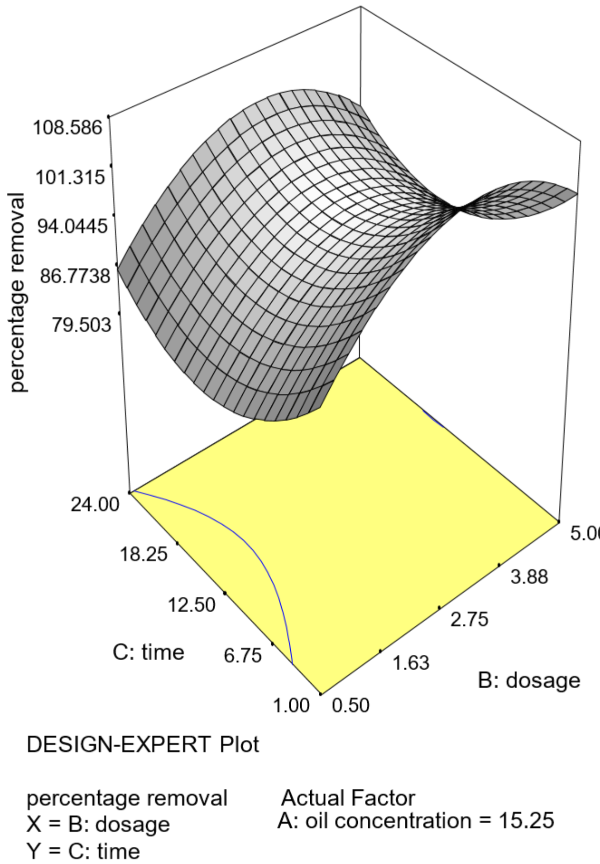

Figure 4: Response surface curve showing the effect of adsorbent dosage (B) and the contact time (C)

Standard 9 and 10 also showed the same result which proved that the higher the dosage of adsorbent gained the higher the percentage removal of oil. With increase the sorbent dosage, oil removal efficiency will increased. The phenomenon is associated with an increase in available binding sites for adsorption in higher sorbent dosage (Ahmad et al., 2005).

3.2.6. Attaining Optimum Conditions and Model Verification

The highest percentage removal from the various run was 100% at 1 h and 24 h of contact time, 2.75 g of biosorbent dosage and 0.50% of synthetic oil wastewater concentration (Table 2). Minimizing the production cost is an important aspect to keep in mind (Nuthalapati et al., 1999) when developing industrial process for the removal of oil from aqueous solution by untreated sugarcane bagasse. Therefore, a statistically assisted optimized process is an appropriate method to solve this problem. In this regard, attaining a high degree of conversion was possible using Design Expert Version 6.0.8 and purely seeking the optimum point on the response surface (Nuthalapati et al., 1999). The determination of optimum point indicates the necessary optimal condition of variables to achieve the highest percentage removal.

To check the adequacy of the model equation, sets of confirmation experiments were carried out within and outside the design space. With the help from the optimization function of the Design Expert Version 6.0.8, two maximum percentage removals were selected based on: (1) the highest percentage removal with all variables in model range and (2) the second highest percentage removal at the shortest contact time. The experiments for both of the proposed optimization condition were conducted and the results are shown in Table 4. The results obtained were found to be relatively close to the predicted percentage removal. The similarities in predicted and actual value confirmed the eligibility of the quadratic model suggested by the software. Thus, Response Surface Methodology (RSM) could be affectively applied with appropriate experimental design to optimize the process parameters in this investigation. The result also showed that the simplistic developed biosorbent revealed good tendency to remove oil from the aqueous solution while giving high oil percentage removal under optimized condition.

Table 4 : The attained optimum condition for untreated sugarcane bagasse in removal of oil in aqueous solution.

Experiment	A: Oil concentration, %	B: Dosage, g	C: Time, hour	Actual percentage removal (%)	Predicted percentage removal (%)	**Deviation (%)**
1	**16.90**	**3.06**	**1.28**	**98.73**	**106.15**	**7.5**
2	**14.19**	**3.10**	**2.98**	**96.45**	**105.75**	**9.6**

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4. Conclusion

The current study on the oil removal from the water using the biosorbent, sugarcane bagasse has been successfully proven. Adsorption reactions were carried out with different oil concentration, contact time and dosage of the adsorbent as the variables. Sugarcane bagasse showed excellent efficiency in removing oil with percentage removal up to 98.73% at 1.3 h contact time with 3.06 g of biosorbent dosage and 16.90% of synthetic oil wastewater concentration. The adsorption data was well described with Response Surface Methodology (RSM). Based on the findings, sugarcane showed a good potential as an oil adsorbing agent.

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