SELECTION OF VARIOUS COAGULANTS FOR SUGAR INDUSTRY WASTEWATER TREATMENT

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ABSTRACT: Sugar industry is one of the industries that produce a high amount of pollutant since its wastewater contains high amount of organic material, Biochemical Oxygen Demand (BOD) and Chemical Oxygen Demand (COD). If this waste is discharged without a proper treatment into the watercourse, it can cause problem to aquatic life and environment. For the primary treatment process, sugar wastewater can be treated by using chemical precipitation method which involves coagulation process. Currently, ferric chloride has been used as the coagulant but it consumes more alkalinity and corrosive. In this study, the suitable coagulant to be used to treat the wastewater from sugar industry and the optimum conditions to achieve high percentage removal of COD was determined. The characteristic of the wastewater was firstly determined. Then, the most suitable coagulant to be used for the treatment was studied by determining their efficiency to reduce COD and TSS in the wastewater at different dosages. Aluminium sulphate (alum), ferric chloride and polyaluminium chloride (PAC) were chosen to be studied for suitable coagulant. The optimum condition of the coagulant (pH, coagulant dosage, fast mixing speed) was determined by using Design Expert software. Results showed that alum can be used to effectively remove 42.9% of COD and 100% of TSS at high dosage (50 mg/l). The optimum condition of alum was at pH 5.2, 10 mg/l of alum and 250 rpm of mixing speed. This shows that at optimum condition, alum can be used to treat wastewater from sugar industry.

KEY WORDS: *Sugar industry wastewater; aluminium sulphate; primary treatment, ferric chloride; polyaluminium chloride*

1. INTRODUCTION

Sugar industry is one of the largest agro-based industry as sugar is one of essential substrate for human dietary consumption and it is an important product for human life. The effluent produced from the sugar industry if it is not properly treated before releasing it into the water sources, it can cause pollution to the environment [1]. The wastewater produced in the sugar manufacturing process has a high content of organic material and subsequently high Biochemical Oxygen Demand (BOD), particularly because of the presence of sugars and organic material in the beet or cane. In sugarcane processing, the typical levels of BOD are 1700–6600 ppm in the untreated effluent, the Chemical Oxygen Demand (COD) is from 2300 to 8000 ppm and the total suspended solids are up to 5000 mg/L, and the ammonium content is high [2].

Numerous systems have been recommended by researchers to treat sugar industry wastewater such as adsorbent [3], electrochemical [4], anaerobic biological treatment [5], biochemical oxidation [6] etc. The wastewater treated by above methods are not meeting the discharge limit; it required modification either in individual treatment or separately.

Chemical precipitation method adopts coagulation and flocculation process and it is proven to be able to remove remarkable amount of pollutants in the wastewater [10]. Coagulation and flocculation is a process of adhesion and contact whereby the dispersed colloid particles form flocs or large cluster and enables them to be removed from water easily by settling, flotation or filtration. In coagulation and flocculation process, there are many types of coagulant that can be used to destabilize the particles and agglomerating the particles into floc form so that it can later be sedimented and separated from the liquid [1].

Alum can achieve high organic removal [7]. This statement can be supported by where the removal efficiency of COD reached up to 48% to 87% in addition to the TSS of the wastewater can be reduced up to 94% [8]. In other study, the percentage of COD and TSS removal was 59.9% to 84.5% and 92.4% to 95.9% respectively [9]. Meanwhile, 62% to 80% of COD removal and 75% to 90% of TSS removal also can be obtained [9].

Ferric chloride, ferric sulphate and ferrous sulphate gave the best performance at too acidic condition [10]. However, by using ferric chloride as coagulant at around neutral pH can gives 44% to 67% of percentage removal of COD and 71 to 76% percentage removal of TSS [9]. Furthermore, by using $FeCl_3$ as coagulant, it can reduce 65.3% to 71.1% of COD and 75.5% to 85% of TSS [10].

Polyaluminium chloride (PAC) allows formation of floc faster compared to other coagulant as it has high positive electrical charge so it can neutralize the charges of the colloidal easily and reduce the repellent between particles thus allows the particles to form larger flocs [1]. By using PAC, percentage of COD that can be removed was 48% to 72% while 78% to 81% of TSS removal [8]. In addition, the percentage removal COD and TSS can be 40% to 56% and 71% to 99% respectively [11].

Hence, in this study, these three coagulants were chosen to treat the wastewater from a sugar processing factory in Shah Alam, Selangor, Malaysia. The most suitable coagulant was determined based on its efficiency to reduce COD and TSS in the wastewater at different dosages. Statistical approach in Design Expert Software was applied to design and optimize the condition of suitable coagulant in terms of its pH, dosage and fast mixing speed.

2. MATERIALS AND METHODS

Materials and methods used are all within certain standards as explained in the following sub-sections.

2.1 Collection and preservation of wastewater from sugar industry

The wastewater was collected from sugar industry which is Central Sugar Refinery (CSR) Sdn Bhd at Shah Alam. Wastewater was collected from influent of the coagulation tank. The sample was then stored in refrigerator at 4 ºC until further use. To prepare for experiment and analysis, the sample was left to warm until 20 ºC. Samples were thoroughly agitated for re-suspension of settled solid before any test was conducted. Initial condition of wastewater (pH, temperature, turbidity, COD, TSS) was taken by performing standard method for examination of wastewater.

2.2 Pre-treatment of the wastewater

The pH of the wastewater was adjusted by using 0.1M HCl or 0.1M NaOH to neutral value (pH 6-7). All tests were performed at the ambient temperature within the range of 20-23 °C.

2.3 Treatment of wastewater using jar test

Chemical precipitation method was simulated by using jar test to coagulate the sample of sugar industry wastewater by using alum, $FeCl_3$ and PAC. It was carried out as a batch test using 6 beakers and 6 spindle steel paddles. Before distributing the wastewater into beakers containing 500 ml of suspension, the sample was mixed homogeneously. The samples of wastewater were distributed in 6 jars and the specific concentration of coagulant (10-50 mg/l) was added so that the total solution in a jar is 500 ml to perform coagulation, flocculation and sedimentation processes. For the first 1 minute, rapid mixing of the solution was done at 200 rpm and it was continued with 15 minutes of rotation at 30 rpm for slow mixing process. The solution was then left for sedimentation for 30 minutes. The treated samples were withdrawn using a pipette from a distance of 2 cm below the liquid level and analyzed for COD and TSS analyses which representing the final concentration.

2.4 Optimization of coagulant condition using Response Surface Methodology (RSM)

Three chosen factors at different ranges which are coagulant pH (5-9), coagulant dosage (10 -50 mg/l) and fast mixing speed (150-250 rpm) were optimized using Face Centered Central Composite Design (FCCCD) under RSM in Design Expert v6.0.8 software. The response was percentage of COD removal. A total of 11 experiments were conducted based on the design of experiment (Table 1).

Table 1: Design of experiment for the chosen factors based on FCCCD

Run	1: pH	2: Dosage of coagulant (mg/l)	3: Fast mixing speed (rpm)
1	7	30	200
2	5	10	250
3	9	50	250
4	5	10	250
5	9	10	150
6	9	10	150
7	9	50	250
8	7	30	200
9	5	50	150
10	7	30	200
11	5	50	150

3. RESULTS AND DISCUSSION

3.1 Characterization of sugar industry wastewater

The characteristics of raw wastewater collected from Central Sugar Refinery (CSR) Sdn Bhd are summarized in Table 2.

Table 2: Characteristics of raw wastewater from a sugar processing factory in Shah Alam, Selangor, Malaysia

Parameter	Value
pH	7.38
Temperature	20.83°C
COD	588.33 mg/l
TSS	132.5 mg/l
Turbidity	186.69 NTU

3.2 Effect of different coagulants on pH, temperature, turbidity, COD, TSS of the wastewater

Based on the results as shown in Figure 1, the pH and temperature of the treated wastewater using $FeCl_3$, alum and PAC as coagulant at all studied dosages were in the range of targeted value thus it can be concluded that either by using $FeCl_3$, alum and PAC as coagulant has no significant effect on pH and temperature of the wastewater.

By using $FeCl_3$, alum and PAC as coagulant, the percentage of turbidity removal was reduced up to 45-76% for $FeCl_3$, 57-80% for alum and 80-90% for PAC [11]. From the graph above, it can be shown that $FeCl_3$ and alum has high percentage of turbidity removal at high dosage and achieved the reduction target. In contrast, PAC shows decreasing of percentage of turbidity removal at high dosage and did not achieve the target**.** This reduction may be due to charge reversal and re-stabilization of colloidal particles by reason of overdosing [12]. Since the percentage of turbidity reduction using $FeCl_3$ and alum were higher compared to percentage of turbidity reduction using PAC, it can be concluded that either by using $FeCl_3$ or alum is suitable for turbidity removal of the wastewater.

In addition, the percentage reduction of COD increased up to 42.9% as the dosage of alum increasing. According to studies done by [13], [10] and [14], percentage of COD removal by using alum can be in the range of 48% to 87%. The percentage of removal might be higher if the dosage of the coagulant is increased. For coagulation with ferric chloride, by using ferric chloride as coagulant at around neutral pH can gives 44% to 67% [8] and COD reduction can be around 65.3% to 71.1% [15]. However, in this study, only 17% of COD can be removed from the wastewater. One of the reason is probably due to the ferric chloride must be used at low pH of wastewater for sugar industry wastewater for a better performance [16]. From the plot in Figure 1, it can be shown that coagulation with PAC only gives 26% of COD and did not reach the target of COD removal which is in range of 40% to 72% of removal [8].

By using PAC, the percentage of TSS removal can reach 71% to 99% [9] and 78% to 81% [8]. In this study, results showed that TSS can be reduced up to 100% at low dosage of PAC. Similarly, 100% of TSS can be removed by using alum as coagulant. This removal is more than a percentage removal achieved by [9], [5] and [16]. In this case, PAC is the most efficient in removing TSS in the wastewater since it can reduce the same amount of TSS as alum at low dosage. In contrast, $FeCl_3$ gives less efficient result of TSS removal with only 40% removal.

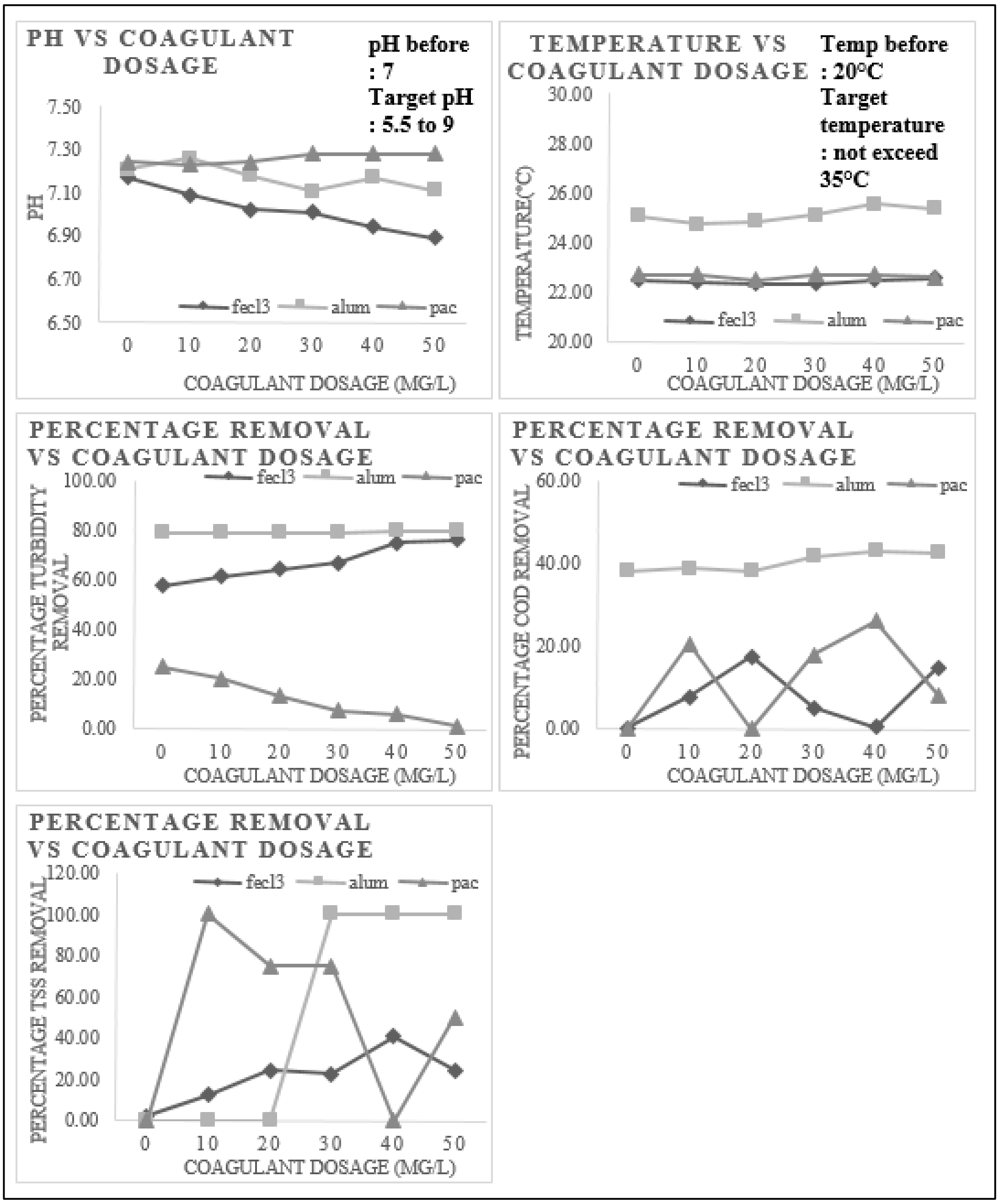

Fig. 1. Effect of $FeCl_3$, alum and PAC at dosage of 10mg/l to 50mg/l on pH, temperature, turbidity, COD and TSS of the wastewater.

In conclusion, at this stage, where pH was set to 7 or neutral and other variables were not optimized yet, alum shows the highest removal efficiency of COD and TSS at all dosages compared to $FeCl_3$ and PAC.

3.3 Optimization of alum condition for sugar industry wastewater treatment

The actual and predicted value for percentage of COD removal using FCCCD are shown in Table 3. Out of 11 runs, the highest percentage of COD removal (27%) was achieved in Run 3, where the pH was 9, 50 mg/l coagulant dosage at 250 fast mixing speed.

Table 3: Actual and predicted value of percentage COD removal

Run	1 : pH	2: Coagulant dosage (mg/l)	3: Fast mixing speed (rpm)	% of COD removal (Actual value)	% of COD removal (Predicted value)
1	7	30	200	12.70	14.28
2	5	10	250	25.68	25.00
3	9	50	250	27.03	21.62
4	5	10	250	24.32	25.00
5	9	10	150	3.85	3.85
6	9	10	150	3.85	3.85
7	9	50	250	16.22	21.62
8	7	30	200	9.52	14.28
9	5	50	150	1.28	1.92
10	7	30	200	20.63	14.28
11	5	50	150	2.56	1.92

The model generated by Design Expert was analyzed using ANOVA. The model F-value of 13.53 (Table 4) implies that the model significant. There is only 0.44% chance that a "Model F-Value" this large could occur due to noise. Values of "Prob>F" less than 0.05 indicate model terms are significant. In this case, fast mixing speed is significant model terms. Values greater than 0.1 indicate the model terms are not significant. If there are many insignificant model terms (no counting those required supporting hierarchy), model reduction may improve the model. The "Curvature F-Value" of 0.146 implies the curvature (as measured by difference between the average of the centre points and the average of the factorial points) in the design space is not significant relative to the noise. There is a 71.51% chance that a "Curvature F-Value" this large occurs due to noise.

Table 4: Analysis of variance for percentage COD removal

Source	Sum of Squares	DF	Mean Square	F-Value	*p-value* Prob > F	
Model	849.873	3	283.291	13.532	0.0044	**significant**
A: pH	1.055	1	1.055	0.050	0.8298	
B : Alum dosage	14.069	1	14.069	0.672	0.4437	
C : Fast mixing speed	834.749	1	834.749	39.873	*0.0007	
Curvature	3.066	1	3.066	0.146	0.7151	**not significant**
Pure Error	125.611	6	20.935			
Cor Total	978.550	10				
R-Squared	0.8712					
Adj R-Squared	0.8068					
C.V.	34.0916					
Pred R-Squared	0.6036					
Adeq Precision	7.483					

The "Pred R-Squared" of 0.6036 is not as close to the "Adj R-Squared" of 0.8068 as one might normally expect. This may indicate a large block effect or a possible problem with the model and/or data. Things to consider are model reduction, response transformation, outliers, etc. "Adeq Precision" measures the signal to noise ratio. A ratio greater than 4 is desirable. The ratio of 7.483 indicates an adequate signal. This model can be used to navigate the design space.

3.3.1 Analysis on effects of pH, coagulant dosage and fast mixing speed on COD removal

From Table 4, for high percentage removal of COD of the wastewater, fast mixing speed was found very significant ($p<0.05$) compared to pH and coagulant dosage. Similarly, by comparison of the line plots shown in Figures 2 to 4, the effect of fast mixing speed is also clearly shown by the large gradient of the slope. Fast mixing speed is preferred in the coagulation process due to the fact that the coagulant can be uniformly dispersed and is helpful in promoting the particle collision. In this study, 250 rpm fast mixing speed contributes to greatest percentage removal of COD which is 16% to 27% (Table 3).

Figures 2 and 3 illustrate that high percentage of COD removal can be achieved at low pH and low alum dosage although the percentage removal is not too high.

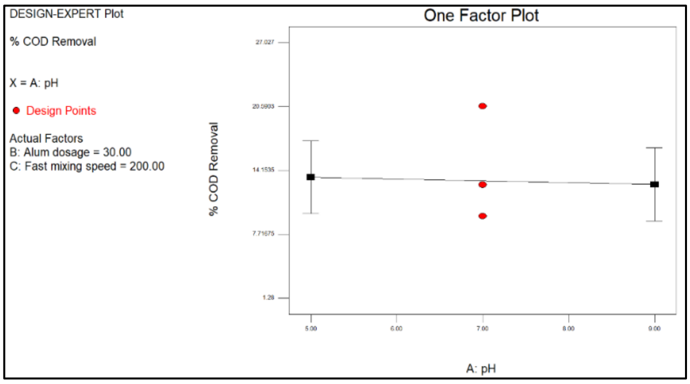

Fig. 2. Effect of pH on percentage of COD removal.

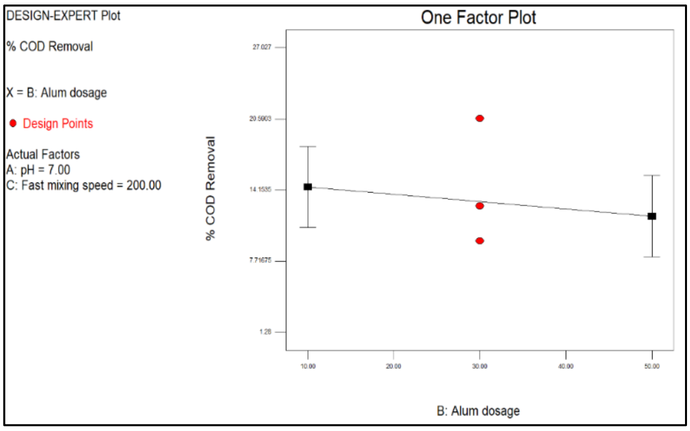

Fig. 3. Effect of alum dosage on percentage of COD removal.

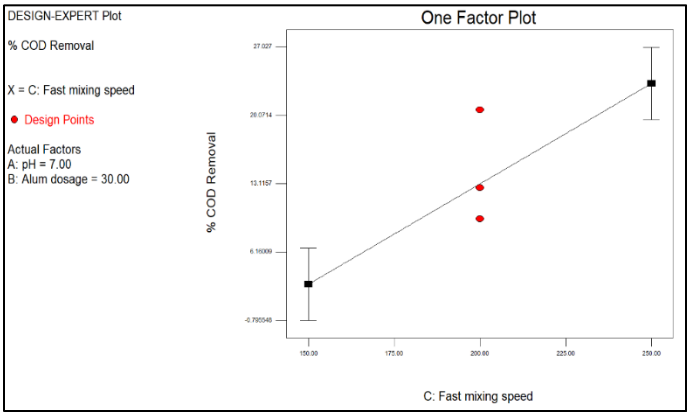

Fig. 4. Effect of fast mixing speed on percentage of COD removal.

3.3.2 Model validation

Model validation generated from Design Expert software was carried out by conducting one experiment out of 10 proposed optimum conditions (Table 5). The validation experiment was conducted at pH 5.21 using alum dosage of 10 mg/l at 250 rpm of mixing speed. The actual value for percentage of COD removal is 25.67% while the predicted one is 24.9%. The small percentage difference (< 3%) indicates that the model has been well-validated.

Table 5: Proposed solution from FCCCD to validate model

No	pH	Alum dosage	Fast mixing speed	% of COD Removal	
1	5.21	10.00	250.00	24.96	**Selected**
2	5.75	10.00	250.00	24.86	
3	5.84	10.00	250.00	24.84	
4	5.99	10.00	250.00	24.82	
5	7.73	10.00	250.00	24.50	
6	8.02	10.00	250.00	24.45	
7	8.58	10.00	250.00	24.35	
8	8.63	10.00	250.00	24.34	
9	5.00	26.39	250.00	23.91	
10	5.00	39.62	250.00	23.03	

4. CONCLUSION

Based on the results from this study on selection of various coagulants for sugar industry wastewater treatment, the following points could be drawn as conclusion points:

- The characteristics of the raw wastewater from sugar industry are high with the average of 588.33 mg/l of COD, 132.5 mg/l of TSS and have turbidity of 186.69 NTU.
- The suitable coagulant to be used to treat this pollutant in sugar industry wastewater was found to be alum with 42.9% of COD removal and 100% of TSS removal compared to ferric chloride and PAC.
- The optimum condition for alum was found to be at pH 5.2, 10 mg/l of aluminium sulphate and at high speed of fast mixing which is at 250 rpm. At this optimum

condition, the purposed percentage removal of COD is 24.961% and the value obtained from the experiment is 25.677%.

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