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Coagulation-flocculation treatment for batik effluent as a baseline study for the upcoming application of green coagulants/flocculants towards sustainable batik industry

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ABSTRACT

The batik industry has been one of the main family businesses in most of the east-coast region of the Malaysian peninsula for many years. However, appropriate water treatment is still a major challenge for this industry. Stringent laws introduced by the Malaysian authorities and the intention to protect the environment are factors that drive researchers to search for suitable, appropriate, affordable and efficient treatment of batik wastewater. Treatment research on batik wastewater is still lacking and coagulation-flocculation treatment using alum was introduced and chosen as a stepping stone toward the selection of green coagulants. This study aimed to determine the best conditions for alum flocculation-coagulation using a standard jar test method. Four main factors were investigated: alum dosage (0.1–3.5 g/L), pH (4–11), settling time (0.5–24 h) and rapid mixing rate (100–300 rpm). Results obtained were further analysed statistically using SPSS software prior to determining the significant effect of variable changes. From this study, the best conditions for batik wastewater treatment using the flocculation-coagulation process were found to be at alum dosage of 1.5 g/L, pH 8, 4 h settling time and a rapid mixing rate of 100 rpm. Chemical oxygen demand (COD), turbidity, colour and total suspended solids (TSS) were removed by 70.7, 92.2, 88.4 and 100%, respectively, under these conditions. This study showed that batik wastewater can be treated by the coagulation-flocculation process using chemical means of alum. This indicates the need for forthcoming developments in natural-based-coagulant-flocculants toward the sustainability of the batik industry.

1. Introduction

The batik industry is considered as part of the textile industry, producing many types of garments with different patterns and motifs

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makes the batik industry one of the main attractions when visiting Malaysia or certain other southeast region countries. The main problem of this industry is a large amount of wastewater produced, which contains contaminants such as wax, grease, heavy metals, surfactants, suspended solids (SS) and dyes (organic or inorganic) [1,2]. Batik is produced via several processes, including pattern making using wax, dyeing, dye fixing and wax removal, as well as final product drying [1]; [74]. Based on a review conducted by Ref. [3]; batik effluent is alkaline (up to pH 12.1), high in colour (1469 ADMI), chemical oxygen demand (COD) (20900 mg/L) and total suspended solids (TSS) (3180 mg/L). According to Ref. [4]; skin irritation, allergic dermatitis and cancer are the toxicity effects due to direct exposure to dye substances used during the dyeing process.

Nowadays, as a matter of fact, the majority of batik industry players are operated as a home-based business, discharging their wastewater directly into waterways. This scenario was also reported by other researchers from the largest batik manufacturer country, Indonesia [5]. Since this wastewater is dye-based, any possible treatment must be able to remove colour, COD and TSS. There are many treatments that have been used to treat batik wastewater. In Indonesia, the most popular treatments studied are electrochemical/electrolysis treatments [2,6]. Other batik and dye-based wastewater treatment approaches studied are physico-chemical treatments such as electrolysis, catalytic treatment and acidification [2,7]. As for biodegradation treatments, plants: 75–95, 93 and 97% removal of colour, TSS and chemical oxygen demand (COD), respectively [8,9], bacteria: 58–92% removal of colour [10,11] and fungi: 50.41% removal of colour [12] were able to treat dye-containing wastewater.

In addition, coagulation-flocculation is known to be a successful and important physicochemical process to treat various types of wastewaters [13]. In these processes, the coagulant used can be either chemical or derived from natural sources. Examples of chemical or inorganic coagulants are alum, lime, ferric chloride and polyaluminium chloride, while natural coagulants are derived from plant parts such as *M. oleifera* seed, *Opuntia* spp., locust bean gum, Rambutan seed and *Tamarindus indica* [14–17], bacteria [18–21] and chitosan [22].

However, coagulation-flocculation process is rarely employed for batik effluent. Only two records utilised the coagulation-flocculation approach to treat batik wastewater. According to Ref. [23]; 73% of COD was successfully removed from batik effluent when alum was used as chemical/inorganic coagulants and lime as coagulant's aid. Higher removals of turbidity (95%) and TSS (86%) were achieved by this approach when a natural coagulant from *Moringa oleifera* seed was employed [24]. Hence, this study will give some added value to the database of batik effluent treatment using the coagulation-flocculation process. To achieve optimum performance, an appropriate amount of coagulant is required; hence, optimum adsorption can occur, charges can be neutralized and particles will flocculate [25]. The dosage of coagulant plays an important role that affecting the overall efficiency of the treatment. Exact dosages will optimize the treatment's efficiency and reduce the cost of operation.

Among conventional chemical/inorganic coagulants, alum is known to be the most effective coagulant for application in the treatment of various types of wastewater, such as yeast industry effluent [26] and dairy wastewater [27]. About 64 and 89% of COD and colour, respectively, were removed from yeast wastewater [26] and 68% of COD were eliminated from dairy wastewater [27]. Additionally, 53 and 92% of COD and colour were respectively removed from biodiesel wastewater [28]. Alum is the least dangerous, corrosive, and expensive of the primary inorganic coagulants, such as ferric chloride and aluminium chloride, making it ideal for usage in small-to medium-sized industries like the batik cottage. In contrast to other coagulants, it is usually possessed stable pH of treated wastewater [29]. Alum usually works well at dosages of 1.5–2.5 g/L [16,30,31]. The optimum dosage of coagulant should be

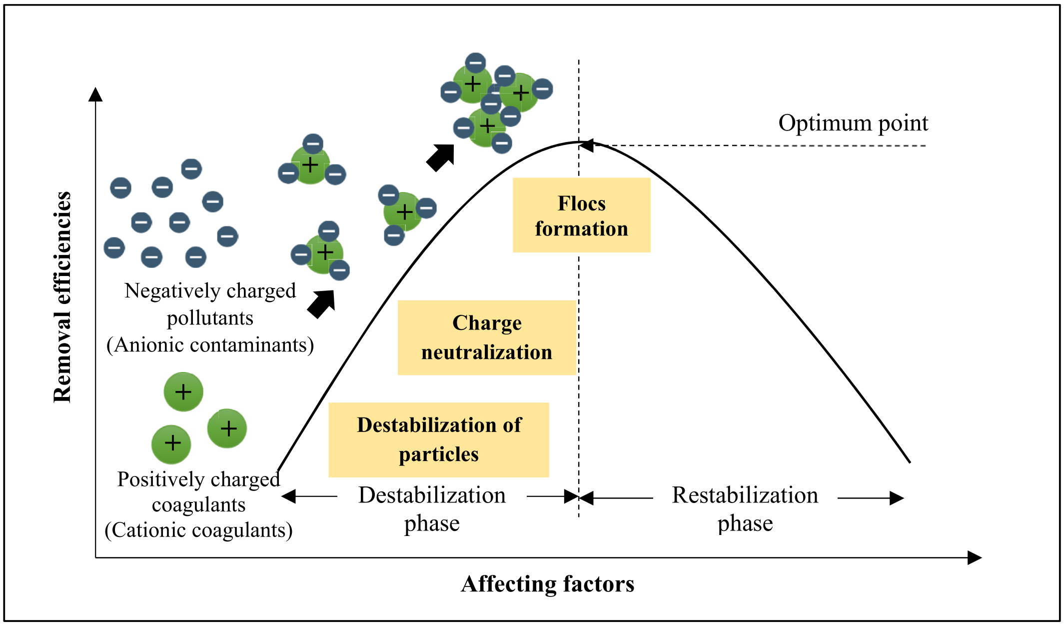

Fig. 1. Schematic diagram of charge neutralization mechanism.

determined since excessive coagulant dosage might affect the cost of treatment as well as increase public health concerns, while too small a dosage might reduce the efficiency of the treatment and render it unable to meet the requirements of environmental quality regulations. As for pH [32], stated that the pH of wastewater has a significant effect on coagulation-flocculation efficiency. According to previous studies, alum coagulation-flocculation is usually efficient at pH 6–8 but depends sometimes on the nature of the wastewater [31,33]. The settling time of the coagulation-flocculation process is one of the important criteria for maximising treatment efficiency. As reported, the coagulation-flocculation performance also increases as settling time is increased [16,34]. However, a longer settling time might lead to another problem: the treatment becoming more time-consuming. An optimized settling time may solve this problem.

Hence, in this study, the commonly used coagulant, aluminium sulphate ($Al_2(SO_4).18H_2O$), known as an alum was chosen to reduce the TSS, chemical oxygen demand (COD) and colour in batik effluent. Additionally, the significant effect of four main factors of the coagulation-flocculation process (alum dosage, pH of wastewater, settling time and rapid mixing rate) and their interactions on the removal of COD, turbidity, colour and TSS were also determined. By using Statistical Package for the Social Sciences (SPSS) tools, the significance of each factor obtained from the overall experiment could be determined. This project is an ongoing research work, aiming to seek naturally derived coagulants that can reduce and replace alum to coagulate-flocculate batik effluent for the sustainability of water resources. A baseline study using alum was carried out for subsequent comparison with the performance of future natural coagulants.

In coagulation-flocculation process, two phases generally involve; destabilization and restabilization phase. As alum is added to wastewater, series of hydrolysis reaction will occur, a range of metal hydrolysis species will be formed and causes pH changes [76]; [35]. This will promote destabilization process of alum and contaminants. In this study, since alum, metallic salts was used, it is expected that the removal mechanism of contaminants from batik wastewater is charge neutralization. Alum added to batik wastewater will be destabilized by forming positively charged ions of Al^{3+} and SO_4^{2-}. As alum and contaminants particles are destabilized, cationic species of alum hydrolysis species tends to adsorb negatively charged particles of wastewater and undergo charge neutralization mechanism [29] as illustrated in Fig. 1. Hence, it is expected that the stabilized contaminants in batik wastewater are attracted to alum to destabilize the charges through charge neutralization process. If optimized conditions of each affecting factors can be achieved, maximum efficiency can be obtained. At this point, any changes on the operating factors will not give any effect on the overall performance [28]. Destabilized coagulant that is not attracted to other particles will be stabilized back in restabilization phase [36]. This assumption was also made by considering the low dosage range used and initial pH of batik wastewater. According to [72], at low dosages, the primary mechanism that may be involved is charge neutralization.

2. Materials and methods

2.1. Source of batik effluent and its characterisation

The batik effluent used in this study was collected from the batik industry located in Kelantan, an east-coast state of Malaysia. The wastewater collected was greenish in colour (Fig. 2) with an alkaline pH of >10. Other characteristics of the wastewater are shown in

Fig. 2. Batik effluent collected from the batik industry.

Table 1. All the parameters are above Malaysian environmental limits, requiring the effluent to be treated.

2.2. Alum coagulation-flocculation process

This experiment was conducted at a laboratory scale. The coagulation-flocculation treatment of batik wastewater was carried out using jar test apparatus (JLT-6, Velp Scientifica, Italy). Fig. 3 illustrates the coagulation-flocculation process of batik wastewater using alum. Wastewater (500 mL) was poured into a 1-L beaker to which alum was added for the coagulation-flocculation process to take place. Alum was added to the beaker, and two stages of rapid and slow mixing were carried out before treated batik effluent can be collected. In this study, a slow mixing rate was maintained at 30 rpm for 30 min. Only one factor was varied at a time while keeping other variables constant. Effect of dosage (A) was conducted first, followed by the effect of pH (B), settling time (C) and rapid mixing rate (D). The process conditions of each analysis are summarised in Table 2. The range of each factor was selected according to previous studies in which alum dosage can work optimally [31]. The alum dosage used may vary depending on the nature of the wastewater. In this study, the alum dosage used was 0.1–3.5 g/L, and a pH range of 4–11 was chosen by considering the initial pH of batik wastewater. The same approach was used to find the suitable range of settling time and rapid mixing rate. Each experimental run was repeated three times to reduce any error.

2.3. Buffer solutions

The buffer solutions that were used to lower or increase the pH value of batik wastewater were 1 M hydrochloric acid (HCl) (Merck, Germany) and 1 M sodium hydroxide (NaOH) (Systerm ChemAR, Malaysia). NaOH solution was prepared by dissolving 40.0 g of NaOH pellets in 1 L of distilled water and then stored in a Duran bottle for later use.

2.4. Analytical methods for water quality analysis

In this study, all parameters such as pH, COD, turbidity, colour and TSS were measured according to APHA Standard Methods for the Examination of Water and Wastewater [38]. For pH and turbidity, a pH meter (Metrohm, Switzerland) and a turbidimeter (HACH 2100AN, USA) were used, respectively. The other parameters COD and colour were determined according to the Reactor Digestion Method (Method 8000) (HACH DR3900, USA) [39,40] and the ADMI Weighted Ordinate Method (Method 10048) [41] (HACH DR3900, USA) while TSS was analysed using the Photometric Method for suspended solids in a liquid (Method 8006) [41].

2.5. Analysis of functional groups of batik wastewater

The FTIR analysis was performed using FTIR (Thermo Fisher Nicolet 6700 Optic). Instruments used in this study have been supported by Makmal Persekitaran, *Department of Chemical and Process Engineering*; Makmal Pencirian Struktur Molekul (MPSM), Centre for Research and Instrumentation Management (CRIM) and Unit Mikroskopi Elektorn, Universiti Kebangsaan Malaysia.

2.6. Statistical data analysis using SPSS tools

The mean values and significant effect of each factor were analysed using SPSS Version 21 for Windows software (SPSS Inc, 1989–2012, Chicago, IL, USA). The significant difference of $p < 0.05$ was based on one-way analysis of variance (one-way ANOVA) [39] with post-hoc multiple comparisons (Tukey and Games–Howell). The homogeneity of variance (Levene's test) of the data was further analysed and discussed.

3. Results and discussion

In this section, the effect of factors on the coagulation-flocculation process of batik wastewater was further analysed. Generally, the

Table 1
Characteristics of batik effluent.

Parameter	Unit	EQA 1974		Value
		Standard A	Standard B	
pH	–	6.0–9.0	5.5–9.0	10.7 ± 0.08
COD	mg/L	80	250	867 ± 45
Turbidity	NTU	–	–	46.1 ± 8.7
Colour	ADMI	100	200	381 ± 29
TSS	mg/L	50	100	72 ± 41

**EQA 1974:* Malaysian Environmental *Quality Act 1974* [37].
*Standard A: applicable to discharges into any inland waters within catchment areas.
*Standard B: applicable to discharges into any other inland water or Malaysian waters.
*COD: Chemical oxygen demand.

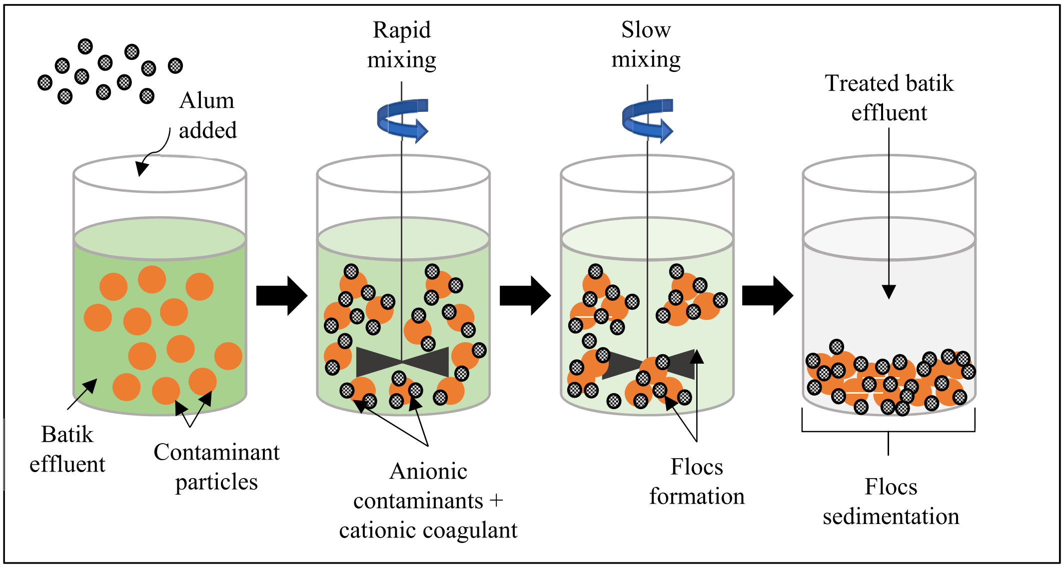

Fig. 3. Illustration of alum coagulation-flocculation process of batik wastewater.

Table 2
Process conditions of each analysis.

Analysis	Factor	Range	Alum dosage	pH	Settling time	Rapid mixing rate
A	Effect of dosage	0.1–3.5 g/L	–	Initial pH of wastewater	24 h	200 rpm
B	Effect of pH	4–11	Best dosage from analysis A	–	24 h	200 rpm
C	Effect of settling time	0.5–24 h	Best dosage from analysis A	Best pH from analysis B	–	200 rpm
D	Effect of rapid mixing rate	100–300 rpm	Best dosage from analysis A	Best pH from analysis B	Best settling time from analysis C	–

coagulation-flocculation process using alum managed to remove turbidity, colour and TSS. This section also discusses the results obtained from the statistical analysis.

3.1. Effect of dosage

In this batch analysis, an alum dosage of 0.1–3.5 g/L was used with pH maintained at the initial pH of wastewater with a rapid mixing rate of 200 rpm and 24 h settling time. The removal efficiencies of COD, turbidity, colour and TSS are demonstrated in Fig. 4. Generally, the effect of dosage was conducted to find the lowest possible suitable dosage for optimum removal efficiency. There are studies using alum reporting dosages as low as 0.04 g/L [75] or as high as 5.5 g/L [26]. According to Fig. 4(a), low COD removal efficiencies were recorded at dosages of 0.1 and 0.5 g/L. The removal efficiency increased when the dosage was increased to 1.5 g/L. However, increasing the dosage from 2.0 g/L up to 3.5 g/L resulted in only small changes in COD removal efficiency. A similar pattern was seen for turbidity removal efficiency, as demonstrated in Fig. 4(b). The low removal efficiency was recorded with alum dosages of 0.1–1.0 g/L. The removal spiked to more than 90% when alum dosage was increased to 1.5 g/L. Increasing the alum dosage from 1.5 to 3.5 g/L showed a similar pattern to COD removal efficiency, with insignificant differences in their removal efficiencies.

For colour and TSS removal (Fig. 4(c) and (d)), maximum removal was recorded at 1.5 g/L alum dosage. Additional alum dosage of up to 3.5 g/L resulted in decreased colour and TSS removal efficiencies. According to Ref. [34]; decreased removal efficiency might occur when the alum dosage exceeds the required level. During the coagulation-flocculation process, when alum is added, colloid particles will form hydrolysis products, which will be destabilized and appear in floc form. Additional alum dosage might result in charge reversal, re-stabilisation of hydrolysis product/particles and cause higher residual turbidity. As a result, the flocs formed might be of smaller size and low density, subsequently decreasing the removal efficiency [34].

Generally, statistical results based on one-way analysis of variance (one-way ANOVA) showed that changes of dosage had significant impacts ($p < 0.05$) on COD, turbidity, colour and TSS removal. However, post-hoc multiple comparisons using Tukey and Games–Howell tests indicated greater significant difference. In dosage effect analysis, the significant effect was analysed based on changes from one dosage to another. From this aspect, evaluation of whether the addition of dosage from one value to another value

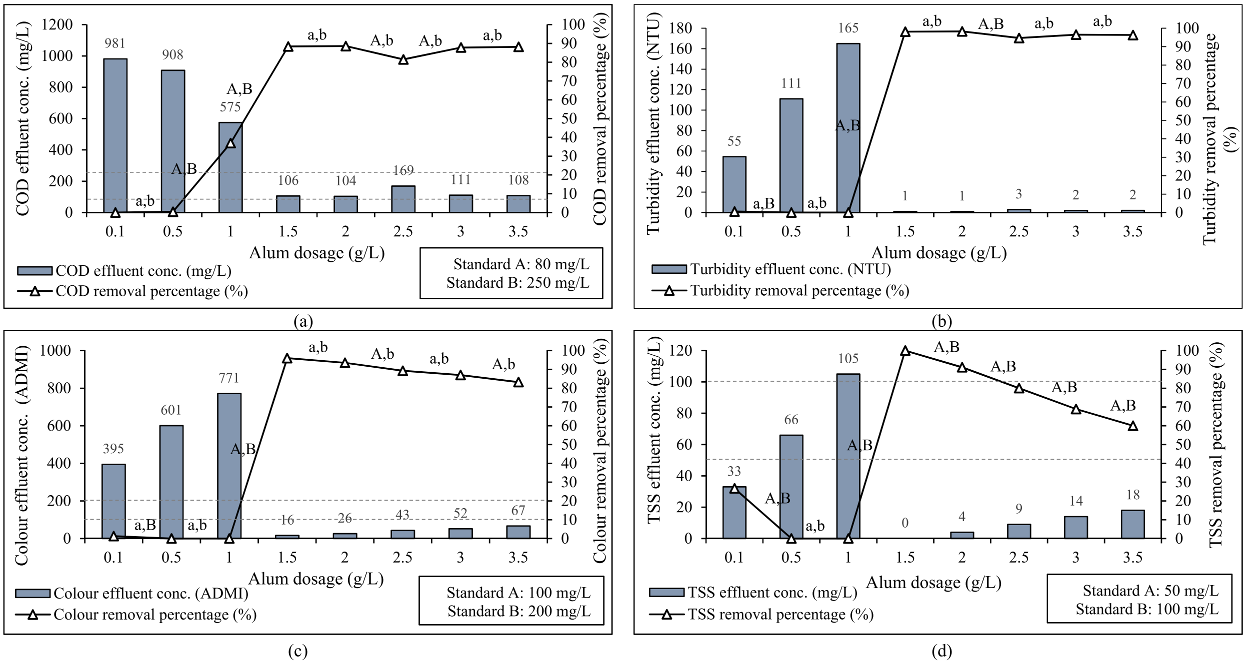

Fig. 4. Effect of dosage on (a) COD, (b) turbidity, (c) colour and (d) TSS removal efficiencies (**A: Tukey significant; a: Tukey not significant; **B: Games–Howell significant; b: Games–Howell not significant). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

was significant or otherwise, can be determined. As can be seen in each Fig. 4(a)–(d), the significant dosage change was indicated using the notation of A (Tukey significant), a (Tukey not significant), B (Games–Howell significant) and b (Games–Howell not significant). From Fig. 4(a), there was a significant effect (both Tukey & Games–Howell) on COD removal when alum dosage was changed from 0.5 to 1.0 g/L and from 1.0 to 1.5 g/L.

Turbidity removals were significantly different when alum dosage was changed from 1.0 to 1.5 g/L (significantly increased) and from 2.0 to 2.5 g/L (significantly decreased) while colour removal efficiency was increased significantly only when alum was increased from 1.0 to 1.5 g/L. Different results were found for TSS and turbidity removal efficiencies, where all dosage changes resulted in significant differences, except for alum increases from 0.5 to 1.0 g/L. Additional alum dosage up to 3.5 g/L significantly decreased the removal efficiencies. It is believed that the additional dosage of alum leads to charge reversal and destabilises the form floc particles [29]. From the overall results, it can be seen that 1.5 g/L alum was able to give high and optimum removal efficiencies [25]. stated that, at optimum dosage, the collision efficiency reaches a maximum value, contributing to optimum performance. Hence, for the analysis of the effect of pH, 1.5 g/L of alum was used.

3.2. Effect of pH

Other than coagulant dosage, the initial pH of wastewater plays an important role in the efficiency of coagulation processes. [13] stated that for the removal of organics, the pH of the wastewater influences the coagulation's effectiveness. A suitable pH may control the production of hydrolysis species. Previous studies recorded that alum works well at certain pH range. According to Ref. [33]; alum usually works well at pH 5–7 while pH 6–8 was found to be the best pH range in a study conducted by Ref. [31]. In this analysis, the variation of pH was selected from a highly acidic region to a highly basic region [42]. Hence, the pH of batik wastewater used was varied from 4 to 11, using its original pH as a control. Alum was added at 1.5 g/L with 24 h settling time and 200 rpm rapid mixing rate.

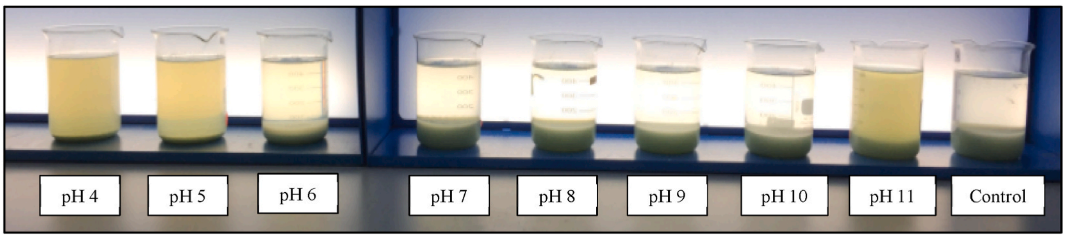

Fig. 5. Coagulation-flocculation of batik wastewater at different pH values.

In the analysis of the pH effect, the multiple difference of post-hoc test was based on changes in initial pH (control) to other values. Based on SPSS analysis, the significant differences when the initial pH (control) was changed to other values were evaluated. Fig. 5 shows the conditions of wastewater after the treatment process, while Fig. 6(a)-(d) illustrates the efficiencies of removal of COD, turbidity, colour and TSS.

Generally, high removals of all four parameters could be achieved at the initial pH of batik wastewater (control), without pH adjustment. However, in real plants, a fixed pH is required to maintain the operation process conditions, since the wastewater characteristics may vary from time to time. Hence, it is important to have one pH value as a guideline for any treatment process involved. Thus, in this analysis, other pH values having similar efficiencies with no significant differences were determined. According to Fig. 6(a), treatment of batik wastewater with no pH adjustment has resulted in 77.4% COD removal, higher than at other pHs. Changing pH from 4 to 11 resulted in 63–76% COD removal. As can be seen in Fig. 6(a), mostly the changes of initial pH to other pH having significant different except for pH 8 and 10. Other than pHs of 8 and 10, COD removals were found to be <71%. pHs of 8 and 10 showed similar high COD removals such as were obtained at initial pH, with 74.3 and 75.8% removals, respectively.

For turbidity, the removal without pH adjustment was 95.0%. As can be seen in Fig. 6(b), at pHs 4 and 5, no turbidity was removed. This might be due to the limited capability of alum to work well in acidic conditions. Naturally, alum is acidic when added to water [43]. As the cation of alum, Al^{3+}, reacts with water, the water tends to be acidic. Hence, sufficient alkalinity of water is needed to achieve effective coagulation. Whenever sufficient alkalinity is present, the reaction of alum as shown in Equation (1) can occur and result in effective coagulation. Hence, when alum is added to acidic wastewater, in this case wastewater with pH 4 and 5, coagulation cannot completely occur or cannot occur effectively [44]. However, as the pH was increased to 6 and up to 10, turbidity removal was increased gradually but decreased again at pH 11. Since the initial pH recorded high turbidity removal, Tukey's and Games–Howell's post-hoc multiple comparisons showed significant differences when the initial pH was changed to values of 4, 5, 6, 7 and 11, while pHs of 8, 9 and 10 showed turbidity removals close to that at initial pH. For colour and TSS removal, alum coagulation-flocculation treatment at initial pH (control) managed to remove 92.7% of colour and 87.1% of TSS.

$$Al_2(SO_4)_3.14H_2O + 6HCO_3^- \rightleftharpoons 2Al(OH)_3 + 6CO_2 + 14H_2O + 3SO_4^{2-} \tag{1}$$

Fig. 6(c) and (d) show colour and TSS removal efficiencies having similar trends. At pHs of 4 and 5, no removal was reported until changing to pH 6 increased removals, which subsequently reached maxima at pH 8, with 90.2 and 83.9% colour and TSS removals, respectively. Increasing pH to 9 and up to 11 decreased the removal efficiencies. From the overall analysis, several favourable pHs were selected for the subsequent tests and are summarised in Table 3. A pH value of 8 can be considered as the most favourable, having similar efficiencies as obtained at the initial pH.

As stated before, the selected or fixed pH is required as a condition guideline in any treatment process, since the characteristics of wastewater may vary from time to time. Even though pH 8 was been selected as the most favourable, further tests were conducted using batik wastewater without pH adjustment and at pH 8 to determine any difference between these two values. Alum dosage was

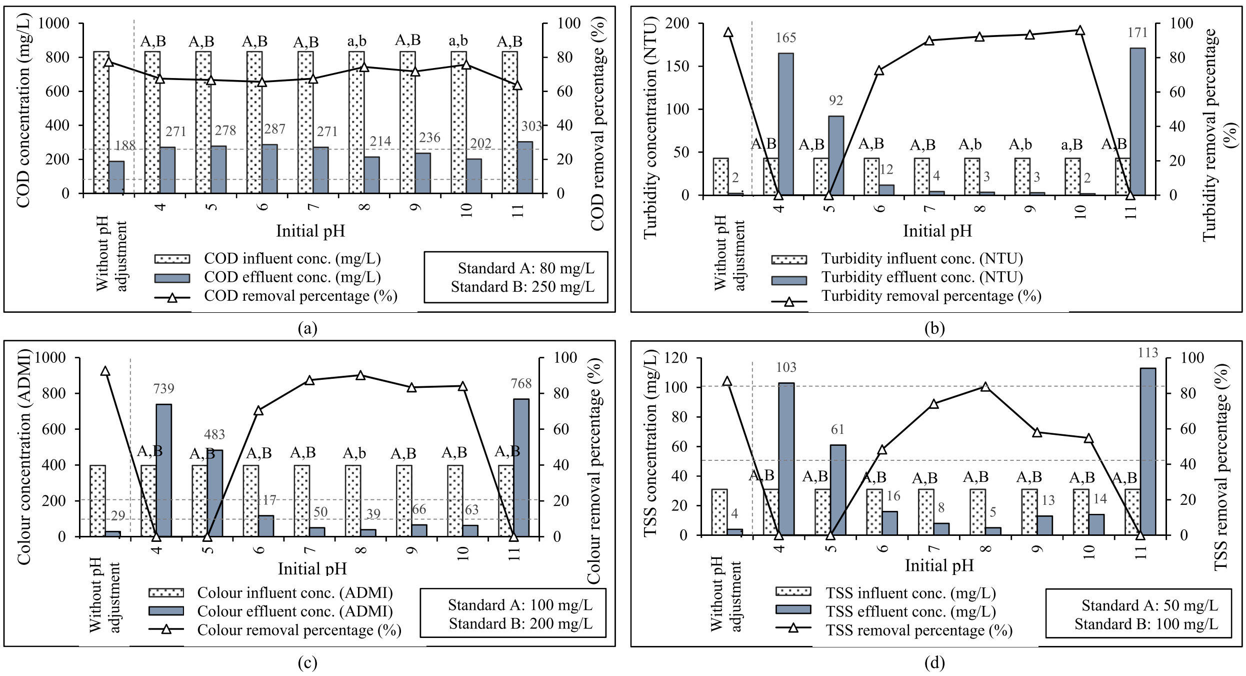

Fig. 6. Effect of pH on (a) COD, (b) turbidity, (c) colour and (d) TSS removal efficiency (**A: Tukey significant; a: Tukey not significant; **B: Games–Howell significant; b: Games–Howell not significant). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

fixed at 1.5 g/L. Their changes during alum coagulation-flocculation were observed physically and visually. Fig. 7 shows the physical observation of the process (a) during treatment, (b) 0.5 h after treatment, (c) 1.0 h after treatment and (d) 24 h after treatment.

In Fig. 7(a), the flocs formed during coagulation-flocculation at pH 8 can be clearly seen compared to the initial pH (10.78). Referring to Fig. 7(b), (c) and (d), the flocs forming at pH 8 settled faster than at the initial pH. The sedimentation at pH 8 remained unchanged from 0.5 to 24 h, indicating that the flocs took less than 1.0 h to settle fully. Compared to the initial pH of 10.78, the flocs formed took some time to fully settle [45]. stated that the formed flocs grew rapidly to a larger size and settled easily. The better performance of coagulation-flocculation at pH 8 can be related to what had been discussed by Ref. [16]. They stated that optimum coagulation-flocculation occurs between pH 6 to 8. When alum was added and a charge neutralization mechanism took place, the positively charged alum predominated and neutralized the negatively charged contaminants in wastewater. Comparing both pHs, it can also be seen clearly that the amount of floc/sedimentation formed at pH 8 was less than at the original pH. This finding was in agreement with what had been found by Ref. [46]. They discovered that from the comparison study that has been made between the charge neutralization mechanism of the coagulation-flocculation process at low pH and high pH, low pH was more advantageous since lower sludge production resulted.

[23] stated that for alum coagulation-flocculation to work well, the initial pH selected should be near to neutral (pH 7). Too acidic or too alkaline pH may increase the solubility of $Al(OH)_3$, forming either positive or negatively charged $Al(OH)_3$ colloids. If the solubility of $Al(OH)_3$ is changed, the ability of $Al(OH)_3$ to adsorb surrounding particles may be reduced, subsequently reducing the efficiency of the coagulation-flocculation treatment [45]. This can be related to what was observed at pH 4, 5 and 11 when efficiencies of removal of turbidity, colour and TSS decreased drastically. Thus, in this analysis, pH 8 was seen to be more favourable than the initial pH of 10.78.

3.3. Effect of settling time

In this analysis, five settling times were selected: 0.5, 1.0, 2.0, 4.0 and 24 h. Other process conditions were fixed: alum dosage of 1.5 g/L, pH 8 and 200 rpm rapid mixing rate. Results of the analysis of COD, turbidity, colour and TSS removal efficiencies are illustrated graphically in Fig. 8. As can be clearly observed in Fig. 8(a), the removal of COD by increasing time from 0.5 to 24 h did not result in huge differences. About 78.1% of COD removal was recorded at 0.5 h. This value decreased to 76.1% at 1.0 h and 73.0% at 2.0 h but increased up to 78.5% at 4.0 h. However, a decrease in the removal efficiency to 74.3% could be seen after 24 h settling time. For turbidity (Fig. 8(b)), coagulation-flocculation for 0.5 h managed to remove 87.6%. This was then increased to 95.9% as settling time increased to 1.0 h and reached 97.1% removal at 2.0 h. A flat trend was observed when settling time was increased to 4.0 h. A trend similar to turbidity was seen for colour removal, as shown in Fig. 8(c). About 85.9% of colour was removed after 0.5 h settling time, which was increased to 91.7% after 1.0 h settling time. According to Fig. 8(d), a flat trend can be seen at 2.0, 4.0 and 24 h settling times with 90.7, 90.0 and 90.2% colour removals, respectively.

However, for TSS removal, a different trend was obtained at short settling times. High removal of 93.9% was recorded at 0.5 h settling time, but this decreased to 78.8% at 1.0 h. As settling time increased to 2.0 h, 100% TSS was removed. This removal was maintained at 4.0 and 24 h settling time. Based on the SPSS analysis, ANOVA analysis showed that variations/changes in settling time had significant effects on all four parameters, with $p < 0.05$. Post-hoc analysis was conducted to determine the significant effect of varying settling time from one value to another. The significance of changes from one settling time to another can be seen in Fig. 8.

Referring to all figures (Fig. 8(a)–(d)), it shows that 2.0 h settling time was adequate for high removal of turbidity, colour and TSS. Increasing the settling time to 4.0 h had no significant effect (Tukey and Games–Howell) on their removals. However, for COD, 2.0 h settling time was not enough for high COD removal; increasing to 4.0 h resulted in higher removals. Referring to Fig. 8, increasing settling time from 2.0 to 4.0 h had a Tukey's significant effect on removal, a significant increase in COD removal efficiency. This comparison led to the selection of a 4.0 h settling time to be used in subsequent analyses.

3.4. Effect of rapid mixing rate

The effect of rapid mixing rate on pollutant removal is discussed in this section. The results of efficiencies of COD, turbidity, colour and TSS removal are shown in Fig. 9, where it can be seen that variation in rapid mixing rate had no significant difference on almost all removal parameters. According to Ref. [26]; the mixing rate may influence the efficiency of a coagulation-flocculation treatment process; however, this analysis gives a contrasting result. Referring to Fig. 9, the removal efficiency of COD at a 100 rpm rapid mixing rate was around 73.1%. This was then reduced slightly to 70.1% as the rate was increased to 200 rpm and continued to decrease to

Table 3
Favourable pH from analysis.

Parameter removal efficiencies	Favourable pH	Removal percentage at each pH	Removal percentage without pH adjustment (Control: Initial pH)
COD	8, 10	74.3% (pH 8) 75.8% (pH 10)	77.4%
Turbidity	8, 9, 10	90.0% (pH 8) 92.2% (pH 9) 93.4% (pH 10)	95.0%
Colour	8	90.2%	92.7%
TSS	8	83.9%	87.1%

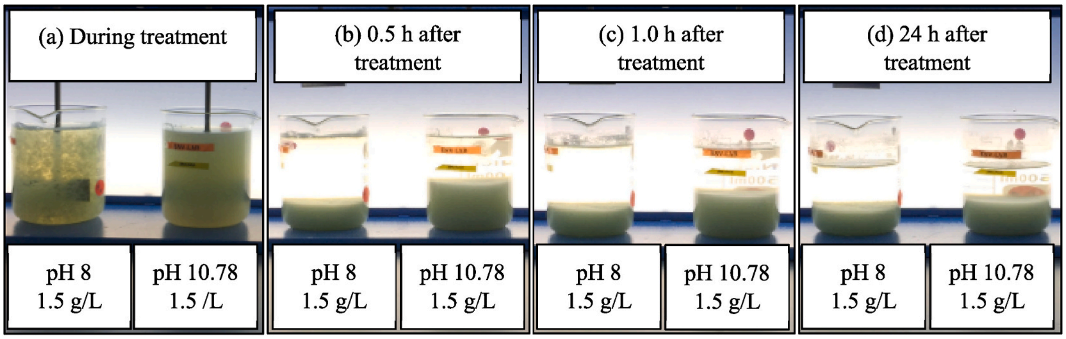

Fig. 7. Observation during treatment, and at 0.5, 1.0 and 24 h after treatment.

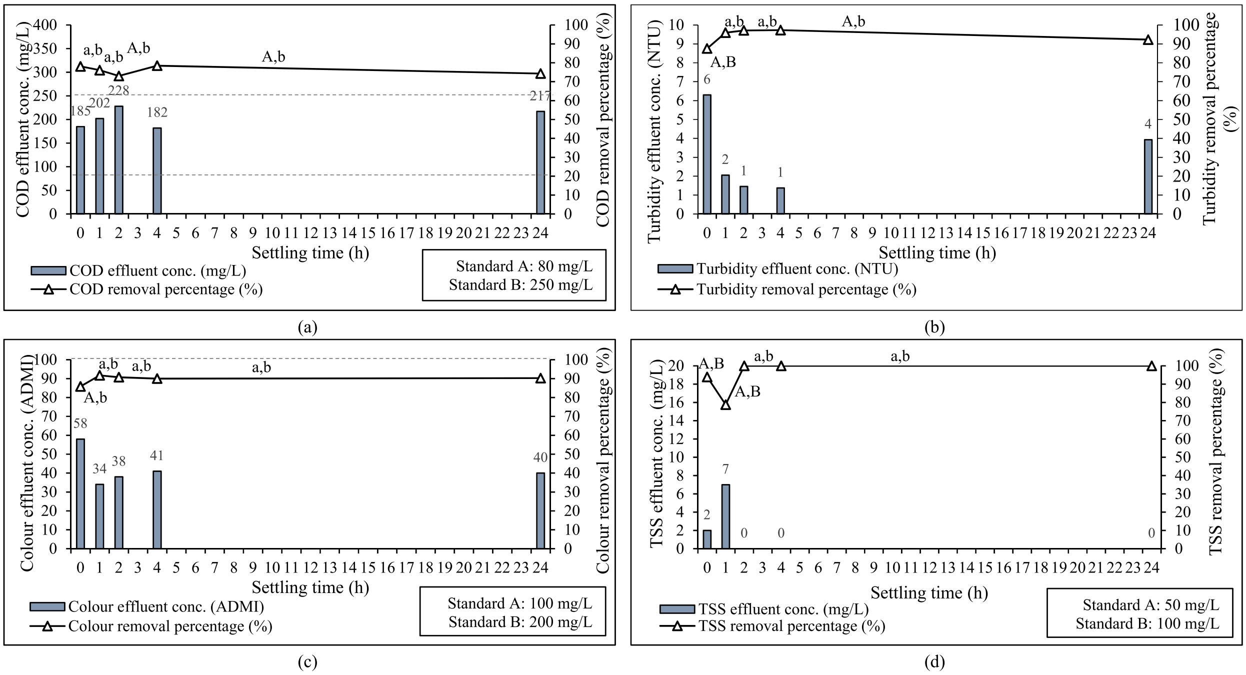

Fig. 8. Effect of settling time on (a) COD, (b) turbidity, (c) colour and (d) TSS removal efficiency (**A: Tukey significant; a: Tukey not significant; **B: Games–Howell significant; b: Games–Howell not significant). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

69.5% at 300 rpm. For turbidity, removal efficiencies at the three rapid mixing rates were not significantly different. Rates of 100, 200 and 300 rpm managed to remove 89.1, 92.2 and 91.5% turbidity, respectively. A similar trend was recorded for colour removal, where removals of 86.1, 88.4 and 87.5% were attained at 100, 200 and 300 rpm, respectively. For TSS removal, changes in rapid mixing rate did not result in great differences in removal efficiency. All three rapid mixing rates analysed managed to remove 100% turbidity.

Based on the ANOVA analysis, the effect of mixing rate only had a significant effect on COD removal efficiency with $p = 0.011$ ($p < 0.05$). Removal efficiency for other parameters, i.e., turbidity, colour and TSS, showed significant probability levels of >0.05 ($p > 0.05$). Referring to Tukey and Games–Howell notation as noted in each graph (Fig. 9(a)–(d)), results of post-hoc tests analysis showed that changes from one mixing rate to another had no significant effect on all parameter removal efficiencies. A study on the effect of mixing rate was conducted by Ref. [31]; and their results showed that the mixing rate did not really affect the performance of the coagulation-flocculation process. From their study, changes in the mixing rate affected only affected the removal efficiency of BOD_5 and not COD or O&G removals.

According to another study by Ref. [16]; alum coagulation-flocculation performed well in a mixing rate range of 100–200 rpm, and increasing the mixing rate showed no significant improvement in turbidity and TSS removal efficiencies. On the other hand [47], stated that the low rate of floc formation can be influenced by an excessively low mixing rate, while an excessively fast rate can promote the breakdown of flocs formed in the coagulation-flocculation process. From these results and comparisons, it can be considered that the mixing rate can still influence the removal efficiency of certain factors but has no effect on the overall alum

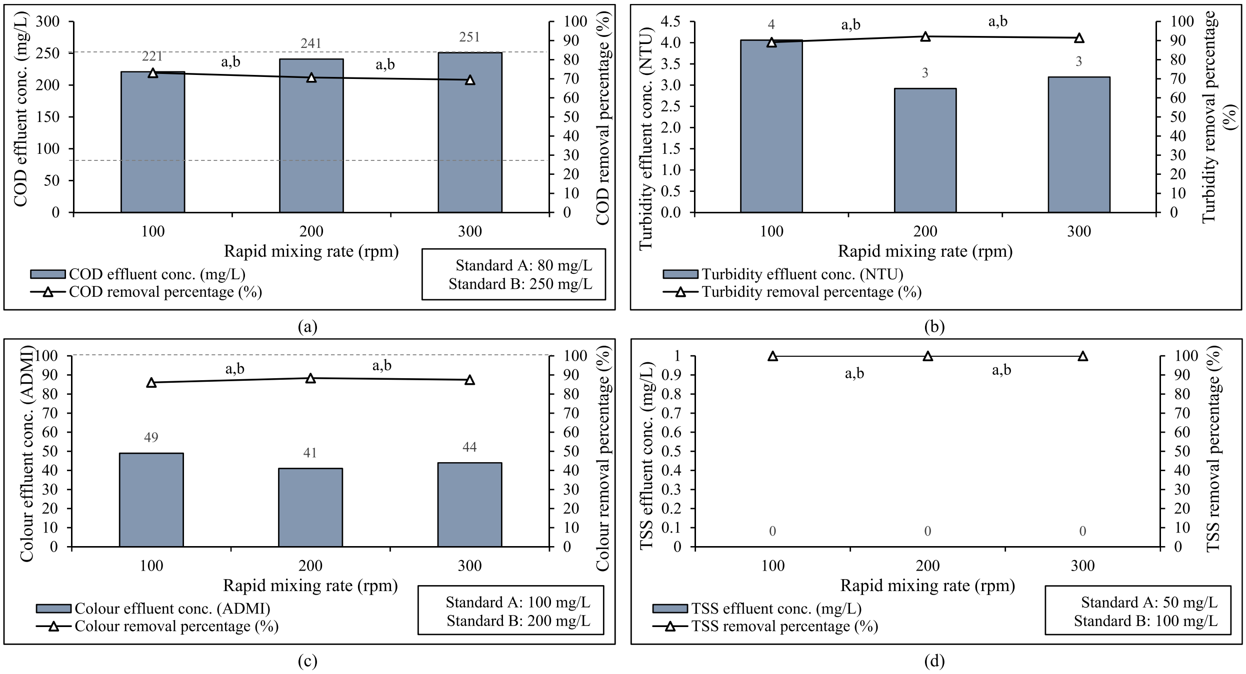

Fig. 9. Effect of rapid mixing rate on (a) COD, (b) turbidity, (c) colour and (d) TSS removal efficiency (**A: Tukey significant; a: Tukey not significant; **B: Games–Howell significant; b: Games–Howell not significant). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

coagulation-flocculation performance. Based on overall removal efficiency, a mixing rate of 100 rpm can be considered the most favourable and sufficient to reduce efficiently the COD, turbidity, colour and TSS. From the analysis of dosage, pH, settling time and rapid mixing rate effect, the overall optimum conditions for coagulation-flocculation process of batik wastewater treatment can be summarised as in Fig. 10.

3.5. *Analysis of functional group groups of batik effluent and expected coagulation-flocculation mechanisms*

FTIR spectrophotoscopy has been very useful for characterization of general functional groups that present in wastewater or contaminants especially organic contaminants [73]. The vibrational functional groups caused by the infrared light can be observed from FTIR spectrum in Fig. 11. The spectrum was referred to Sigma Aldrich IR spectrum (https://www.sigmaaldrich.com/MY/en/technical-documents/technical-article/analytical-chemistry/photometry-and-reflectometry/ir-spectrum-table) to obtain the

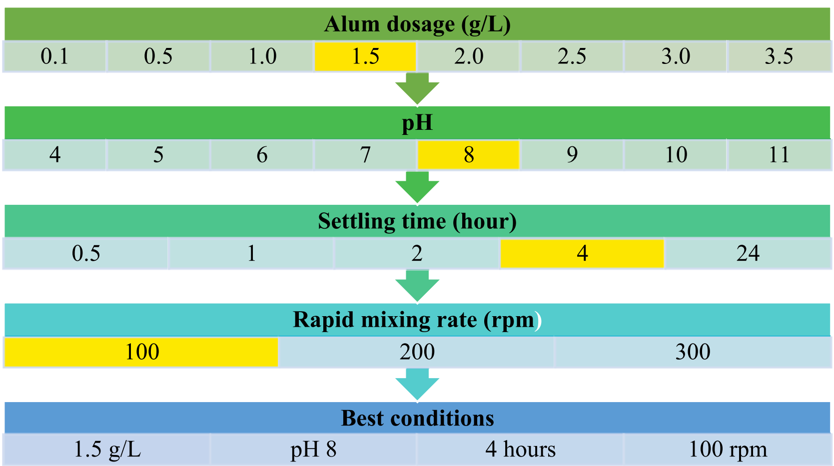

Fig. 10. Summary of overall analysis which led to the selection of best conditions. * the selected optimum conditions were indicated by yellow highlights. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

functional groups of batik effluent. A strong and broad peak of 3394.67 cm^{-1} indicated the O–H stretching of alcohol compound or can be associated with intermolecular bonded of hydroxyl groups. While a sharp and medium peak of 1645.20 cm^{-1} indicates the presence of C═C stretching of disubstituted or cyclic alkene. The broad band at a peak of 673.21 cm^{-1} indicates the Al–O–Si bond group implying the content of sodium silicate used in batik making process [48]. According to Ref. [49]; the hydroxyl groups may contribute to the anionic characteristics of the batik effluent. Hence, by considering the low dosage used (1.5 g/L), alkaline pH (pH 8) and anionic characteristics of batik wastewater, it can be expected that charge neutralization should be the responsible mechanism during alum coagulation-flocculation treatment of batik effluent.

3.6. Comparisons with previous studies

The study of batik wastewater treatment using coagulation-flocculation can be considered new compared to a similar type (dye wastewater) of batik wastewater, that is textile wastewater. In this section, several studies that have been conducted to determine the performance of the coagulation-flocculation treatment process on similar types of wastewaters are discussed. Comparisons between studies focus on the factors and parameters of interest and can be seen in Table 4.

Referring to the table, it can be seen that there are several studies that have conducted research similar to the coagulation-flocculation treatment of batik wastewater; for example, the study by Ref. [23]. In their study, the coagulation-flocculation of batik wastewater was conducted using alum at 1 g/L. Their treatment managed to remove a higher percentage of COD (73.3%) from batik wastewater of higher COD content (6972 mg/L) than that used in this study (<1000 mg/L). However, it should be noted that there was another type of coagulant used in their treatment: about 3 g/L of lime was added, and this might have helped to increase the overall efficiency. In any field of study, direct comparison is not easy. Each study used its own process conditions.

In the case of [23]; the study was conducted using alum and lime for the coagulation-flocculation of batik wastewater at pH 5. Another study on batik wastewater treatment using coagulation-flocculation was conducted by Ref. [24]. Batik effluent of pH 6.05, 3713 mg/L COD, 1306 NTU turbidity and 1248 mg/L TSS was treated by *M. oleifera* seed at a 50 g/L dosage. This seed is an organic coagulant, and this might influence the higher dosage amount used compared to that used for a chemical/inorganic coagulant [53]. applied the same coagulant, alum, to textile wastewater of pH > 10, i.e., close to the pH of this study. However, their COD content was quite high (5744 mg/L). Their treatment managed to remove 58.6% of COD using 5 g/L of alum, at pH 4, with 1 h settling time and 80 rpm mixing rate. Compared to our study, a high amount of alum was used to reduce a high level of COD content. This might relate to that stated by Ref. [16]; that a high dosage of coagulant might be required to reduce a high content of contaminants.

Besides coagulation-flocculation, other treatment processes that have been used to treat batik wastewater are also discussed. Previous treatments for batik wastewater and their performances according to current reports are summarised in Table 5. Many types of treatment processes, including biosorption, phytoremediation, electrolysis, photodegradation and physical treatment using a baffle separation tank, have been used. Based on the data compiled.

(Table 5), it can be seen that most studies focused on COD removal, half of them studied TSS removal and only a few studied turbidity and colour removal. Attracting attention were some green approach treatments that gave high COD removal. These included biofilters and constructed wetlands [54], phytoremediation [56,57] and fungi degradation [58]. However, as stated by Ref. [56]; the treatment performance of phytoremediation was limited to a certain day of treatment and it may drop as dead plants in the system might affect the removal efficiency.

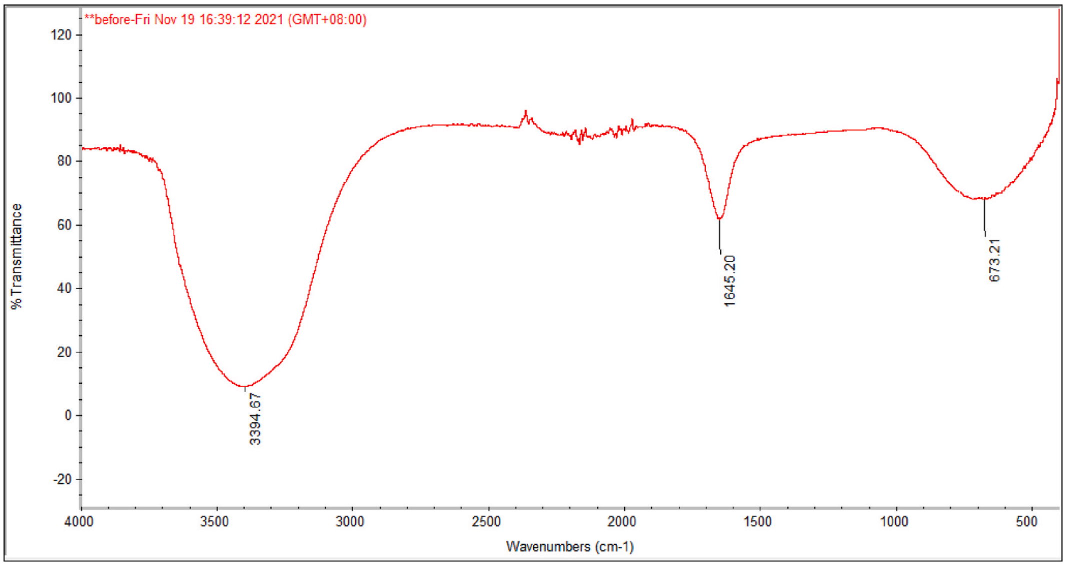

Fig. 11. FTIR spectrum of batik effluent.

Table 4
Comparison of previous studies for batik and textile wastewater treatment through coagulation-flocculation.

Type of wastewater	Wastewater characteristics	Type of coagulant	Interest factors	Process conditions				Removal efficiencies (%)				Reference
				Dosage (g/L)	pH	Settling time (hour)	Mixing rate (rpm)	COD	Turbidity	Colour	TSS	
Batik effluent	pH: 10.62–10.78 COD: 822–912 mg/L Turbidity: 37.4–54.8 NTU Colour: 352–3743 ADMI TSS: 31–113 mg/L	Alum	- Dosage - pH - Settling time - Mixing rate	1.5	8	4	200	70.7	92.2	88.4	100.0	This study (2022)
Batik effluent	pH: 5.0 COD: 6972 mg/L	Alum + Lime	–	1.0 (alum) 3.0 (lime)	5	24	100	73.3	–	–	–	[23]
Batik effluent	pH: 6.05 COD: 3713 mg/L Turbidity: 1306 NTU TSS: 1248 mg/L	*Moringa oleifera* seed	- Dosage - Settling time	50	–	3	100	–	95.3	–	86.4	[24]
Textile effluent	pH: 9.5–11 COD: 1385 mg/L Colour: 4600 PtCo TSS: 210 mg/L	Magnesium chloride ($MgCl_2$)	- Dosage	0.8	–	0.5	200	75	–	94	57	[50]
Textile effluent	pH: 9.5–11 COD: 1385 mg/L Colour: 4600 PtCo TSS: 210 mg/L	Ferric chloride ($FeCl_3$)	- Dosage	0.6	–	0.5	200	84	–	88	65	[50]
Textile effluent	pH: 7 COD: 8720 mg/L TSS: 992 mg/L	*Moringa oleifera* seed powder	- Dosage - Agitation period	0.7	–	–	115	–	–	98.0	–	[51]
Textile effluent	pH: 11.89 COD: 1189 mg/L Turbidity: 213 NTU	Chitosan	- Dosage - pH - Settling time	0.2	4	1	200	58.0	99.0	–	–	[52]
Textile effluent	pH: 10.30 COD: 5744 mg/L Colour: 3840 PtCo	Alum	- Dosage - pH - Settling time	5.0	4	1	80	58.6	–	74.0	–	[53]

Table 5
Type of batik effluent treatments and their efficiency.

References	Type of treatment	Batik wastewater characteristics	Removal (%)				
			COD	Turbidity	Colour	TSS	Others
[54]	Biofilter and constructed wetland of *Hymenocallis littoralis* plant	COD: 12,000 mg/L TSS: 3180 mg/L O&G: 9740 mg/L	72.7	–	–	98.2	O&G: 79.5
[55]	Biosorption process using *S. cinerea and P. ostreotus* baglog waste	pH 8.9 COD: 5004 mg/L TSS: 195 mg/L	77.3	–	–	65.1	–
[56]	Phytoremediation using *Eichhornia crassipes*	pH: 6.55 COD: 359 mg/L TSS: 1336 mg/L	83	–	89	–	–
[6]	Electrolysis process	pH: 5.2 COD: 290 mg/L TSS: 108 mg/L	61.4	–	–	85.2	–
[57]	Phytoremediation using Vetiver *Chrysopogon zizanioides* (L)	pH: 9.8 COD: 1450 mg/L TSS: 268 mg/L	89.1	–	–	–	–
[58]	White-rot fungi *Ganoderma lucidum* degradation	Dye: 100 mg/L	81	–	–	–	Dye: 60.5
[2]	Photodegradation/photocatalysis process	COD: 176 mg/L TSS: 86 mg/L	55.4	–	49.0	86.0	–
[59]	Baffle separation tank	COD: 1390 mg/L	50	–	–	–	–
[7]	Acidification & filtration	COD: 4915 mg/L	78–95	–	–	–	–
[71]	Electroflotation	–	–	69.6	83.3	94.0	–

3.7. Forthcoming research developments towards sustainable batik industry

This project is an on-going research work aiming to seek for naturally derived coagulants that can reduce and replace alum to flocculate suspended solids in batik effluent for the sustainability of water resources. Alum is a metal-based coagulant which can lead to heavy metal contamination in the environment and the batik industry normally consumes chemical dyes which are also toxic to the environment. Thus, a baseline study using alum was carried out for subsequent comparison with the performance of future natural coagulants. With natural coagulants, the toxicity of the resultant sludge from batik industry effluent treatment after going through the coagulation-flocculation process will be much rendered if alum were used instead. Besides, what will be the forthcoming research developments for naturally derived coagulants for batik effluent?

Recently, a review published by Ref. [3] proposed a hybrid treatment system for effluent produced by the batik industry. The physical and biological process of natural adsorption and phytoremediation is proposed as an integrated physical and biological treatment. Besides adsorption, a coagulation-flocculation process using natural resources for the treatment of batik effluent is also seen as having great potential to be employed to remove TSS, turbidity, COD and colour (Table 4). The removal performances for turbidity, colour, TSS and COD were as high as 99, 98, 86 and 58%, respectively. In order to fully utilise biocoagulant-flocculant in a real application, more exploration needs to be done to obtain the maximum removal ability of each natural coagulant. Fig. 12 illustrates the key elements that researchers may look forward to in order to strengthen the fundamental of biocoagulant-flocculant for such a sustainable approach.

Screening of potential biocoagulant-flocculant from natural resources will be the primary action that should be taken by researchers as only *Moringa oleifera* was reported to be able to treat batik effluent (Table 4). Plants-derived [60], microorganisms [42,61] and animals-derived [62] were reported to have the ability to remove turbidity, TSS and colour from wastewater. The utilisation of various natural coagulants could produce different results in removing effluent parameters [63]. For example, combination of plant-derived coagulant; rice-starch and animal-derived coagulant; chitosan, with 1:1 ratio was managed to removed more than 80% of turbidity from domestic wastewater [64]. Next, to study the extraction of active compounds from potential natural sources. Different methods employed during the extraction of active compounds from natural coagulants will result in different removal performance of pollutants during coagulation-flocculation process. Beside water, salt (sodium hydroxide, sodium chloride, sodium nitrate) and alcohol (ethanol) solutions have been widely used as solvents during the extraction of active compounds [65]. No research has been conducted so far to study the efficiency of extraction methods for active compounds from natural resources focusing on the treatment of batik effluent (Table 4).

Furthermore, after determining the critical factors through, the next step is to optimize the actual values of these process factors. The statistical design of experiments is a powerful approach for process conditions optimization. It offers a systematic way of simultaneously evaluating multiple parameter effects and analysing the resulting process outputs. To achieve this purpose, a central composite response surface methodology (RSM) can be used [8,66]. Biocoagulant-flocculant dosage, pH, initial turbidity and stirring speed and time have been reported to closely influence the efficiency and effectiveness of the coagulation-flocculation process by natural coagulants-flocculants [42,63,66,67].

For the purpose of implementation of biocoagulant-flocculant at the industry level, scaling up and techno-economic analysis is highly recommended to be performed. Another area that researchers need to scrutinize is the sludge produced after biocoagulation-flocculation treatment. The sludge produced after coagulation-flocculation process using natural coagulants-flocculants will be much

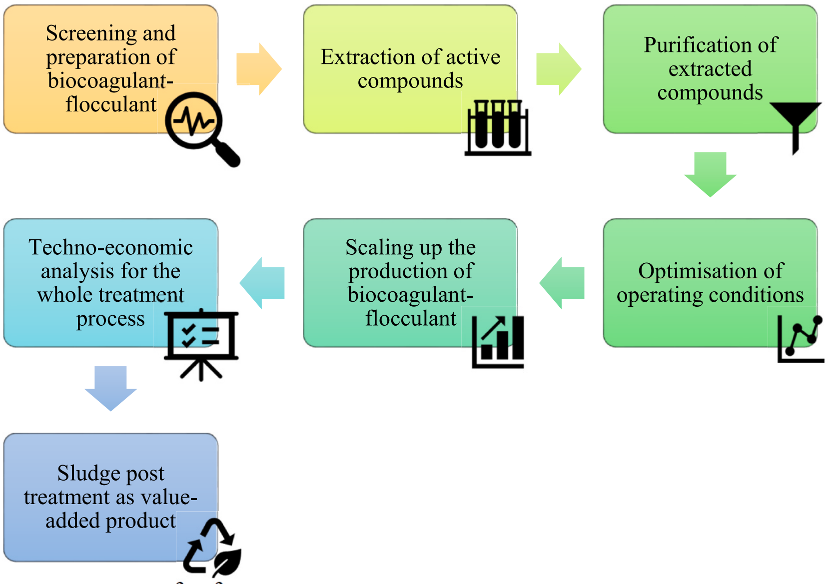

Fig. 12. Key elements for forthcoming research developments of batik effluent applying biocoagulant-flocculant.

less toxic and more biodegradable compared to the generated sludge using chemical/inorganic coagulants-flocculants. The application of the resultant sludge as an alternative raw material in processes may become a secondary resource for various other applications. According to Ref. [68]; sludge produced from textile industry treatment was stabilized after the incorporation with ordinary Portland cement which posed no leachability of toxic metals from sludges. Another study conducted by Ref. [69] proved the ability to recycle textile effluent treatment sludge as a valuable product fuel for energy generation. A lot more time and effort are required to solve an incomplete jigsaw puzzle regarding the ability of biocoagulant-flocculant in treating batik effluent. The last piece of the jigsaw is expected to be resolved as more explorations are done in future.

4. Conclusions

This study was conducted to determine the capability of the coagulation-flocculation process for batik effluent treatment. The aim of this ongoing research was actually to identify a natural coagulant that potentially treats batik effluent. As the first step, the coagulation-flocculation process study using alum, a conventional coagulant was used. The coagulation-flocculation process used in this study managed to treat batik wastewater successfully. The best conditions of the process were found to be at pH 8, 4 h settling time, 100 rpm mixing rate and 1.5 g/L alum dosage. The alum coagulation-flocculation process managed to reduce COD, turbidity, colour and TSS by 70.7, 92.2, 88.4 and 100.0%, respectively. Compared to EQA Standard 1974 [70], the effluent concentration of COD (221 mg/L), TSS (0 NTU) and colour (49 ADMI) were reduced to a level less than the permitted levels. Hence, this treated wastewater is safely discharged to water bodies. TSS and colour were reduced below Standard A (50 mg/L and 100 ADMI, respectively) while COD was reduced below Standard B (250 mg/L). For future study, it is recommended to actively screen potential natural coagulant-flocculant either from plants, microorganisms and animals for batik wastewater treatment to reduce the usage of chemical/inorganic coagulant-flocculant thus ensuring the sustainability of the ecosystem.

Author contribution statement

Nurull Muna Daud: Conceived and designed the experiments; Performed the experiments; Analysed and interpreted the data; Wrote the paper.

Siti Rozaimah Sheikh Abdullah: Conceived and designed the experiments; Analysed and interpreted the data; Contributed reagents, materials, analysis tools or data; Wrote the paper.

Hassimi Abu Hasan; Ahmad Razi Othman: Contributed reagents, materials, analysis tools or data.

Nur 'Izzati Ismail: Analysed and interpreted the data; Wrote the paper.

Data availability statement

Data will be made available on request.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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