

# Colour Removal of Reactive Dye from Textile Industrial Wastewater using Different Types of Coagulants

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**ABSTRACT**— *A study on removal of commercially used reactive dye was conducted to investigate the effectiveness of coagulation-flocculation from textile wastewater. Three types of coagulants were studied; aluminium sulphate (alum), polyaluminium chloride (PAC), and magnesium chloride (MgCl<sub>2</sub>). Polyelectrolyte, Koaret PA 3230 was used as the coagulant aid. The coagulant dosage between 400 mg/L to 5,000 mg/L was studied using jar test. The changes of pH, types and dosage of coagulant, and the addition of coagulant aid on the percentage removal and the concentration of the dyes were determined through colour point. Up to 90% colour removal for reactive dye could be achieved using alum of concentration 6,000 mg/L and pH 2.4. Meanwhile, 99 % colour removal could be achieved using MgCl<sub>2</sub> with concentration of 4,000 mg/L and pH 10.4, whereas 100% colour removal could be achieved using PAC with concentration of 2,000 mg/L and pH 4.1. Among the coagulants used, PAC was the most effective coagulant in treating each dye, with the colour removal up to 100% at the dosage of 800 mg/L. The flocs settling time for the treatment with MgCl<sub>2</sub> was shorter than the treatment with PAC and the treatment with Alum was longer than the treatment with MgCl<sub>2</sub> and PAC. This result can contribute some knowledge on the use of effective coagulants in treating textile industrial wastewater.*

**Keywords**— reactive dye, colour removal, textile industrial wastewater, coagulation-flocculation

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## 1. INTRODUCTION

Dyestuff wastewater from textile industry has been the target of considerable attention in the field of wastewater treatment, not only because of its toxicity, but only because of its visibility. Every year, it is estimated that 280,000 tons of textile dyes are discharged in the textile industry (Jin et al., 2007). According to recent statistics, the annual sewage in China has already reached 390 million tons, including 51% of industrial sewage, and it has been increasing with the rate of 1% every year (State Environmental Water Protection Administration, 2010). The wastewater contains residues of reactive dyes and chemicals, such as complex components. Chromium is among one of the pollutants in wastewater which causes a lot of disposal issues. In the dyeing process, the average dyeing rate is more than 90%. Consequently, the residual dyeing rate in finishing water is about 10%, which contributes to the main source of pollution (Ding et al., 2010).

Synthetic dyes are used extensively in textile wastewater and it is usually treated before it leaves the textile plant. The issue of textile wastewater is generally colour removal as there are complexities in many types of dyes (Saranraj, 2013). Commercial synthetic dyes have excellent binding ability initiated from the formation of a covalent bond between their reactive group and the surface groups of the textile and cellulose fiber. In the textile industry, they are used extensively and their release in the ecosystem caused by increasing environmental pollution danger, because of their toxicity, mutagenicity and non-biodegradability (Asouhidou et al., 2009). Reactive dyes have been identified as the most problematic dye class with respect to treatment among other dyes, as they tend to pass through conventional treatment system (Lazaridis and Karapantosis, 2003).

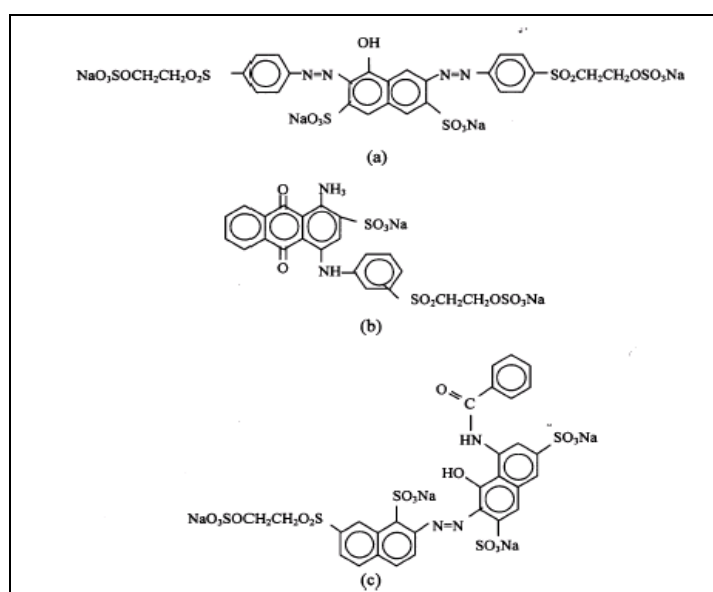
The treatment of textile dyeing wastewater by traditional methods has proven to be ineffective for many wastewater treatment facilities. Conventional textile dyeing system, such liquid-liquid extraction, uses a lot of fresh water, which is then disposed as water containing dye stuff chemicals. The chemicals are water soluble and have relatively low exhaustion rates, therefore, present in high concentrations in the effluent. Some previous research has indicated that biological treatment of textile wastewater has low degradation efficiency due to the presence of biological inert matter combined with high molecular weight dyestuff. On the other hand, physical adsorption is effective in removing non-biodegradable pollutants but it is an expensive method and it is difficult for adsorbent regeneration (Riyanto et al., 2013). Activated carbon adsorption treatment has been proven to be an effective replacement for combined biological and

chemical treatment because it depends on the type of carbon and the characteristic of wastewater (Manaf and Fazlullah, 2005).

Coagulation and flocculation methods are widely used for wastewater treatment, as these methods are efficient and simple to operate (Imen et al., 2010). However, as most of the dyestuffs are designed to withstand environmental conditions such as a slight, pH effect and microbial attack (Manaf and Fazlullah, 2005). Therefore, the aim of this research is to determine the effectiveness of coagulation-flocculation method for the removal of colour from textile wastewater using three different types of coagulants. The study focuses on the effect of pH, coagulant dosage and settling time to obtain the optimum condition of the highest removal of colour reactive dye.

## 2. MATERIALS AND METHOD

Textile wastewater samples were collected from an equalization tank of textile treatment plant of a dye mill in Batu Pahat, Johor, Malaysia. The reactive dyes contained in the textile wastewater were Remazol Black B (RBB), Remazol Brilliant Blue R (RBBR), and Remazol Brilliant Red F3B (RBRF3B). The structures of these reactive dyes are shown in Figure 1 (a), (b), and (c).



**Figure 1:** The structure of reactive dye Remazol Black B (a) Remazol Brilliant Blue (b) and Remazol Brilliant Red F3B (c)

Aluminium sulphate (Alum), polyaluminium chloride (PAC) and magnesium chloride (MgCl<sub>2</sub>) were used as the coagulants, whereas polyelectrolyte, Koaret PA 3230 of commercial grade, was used as the coagulant aid. Sodium hydroxide (NaOH, reagent grade) was used as the pH adjusting agent in the coagulation-flocculation process. Distilled water was used to prepare all the dye solutions, coagulants and coagulant aid solutions.

The experimental work involved treatment of reactive dye conducted in the Environmental Engineering Laboratory, Department of Civil Engineering, College Universiti Tun Hussein Onn Malaysia. The first part involved the treatment of 150 mL reactive dye with 3,000 mg/L to 7,000 mg/L of alum, 1,000 mg/L to 5,000 mg/L of MgCl<sub>2</sub>, and 500mg/L to 4,000mg/L of PAC respectively. Jar test was conducted to determine the effects of solution pH, types of coagulant, coagulant dosage and coagulant aid dosage on colour removal.

A six-paddle stirrer with six beakers were used to conduct the jar test. Each beaker contained 150 mL of the prepared dye solution. NaOH was added to adjust the pH of solution for a period of 1 min at 60-65 rpm. The initial pH for the solutions was measured using a pH meter (ORION 410). Then, the coagulant was added to the beakers and the samples were mixed at 60-65 rpm for 3 min and followed by the addition of polyelectrolyte as the coagulant aid. When the solution height reached half of the beaker and the formed flocs were allowed to settle, the respective settling time was recorded. The final pH for the dye solution was measured using a pH meter and the supernatant was taken for analysis.

The concentrations of dye solutions were measured at a wavelength corresponding to the maximum absorbance (λ<sub>max</sub>) by means of a UV visible spectrophotometer (Shimadzu). The percentage of colour removal was calculated by

comparing the absorbance value of the supernatant to the standard curve obtained by a known dye concentration. The dye concentrations were determined through colour point (pt/Co) measurement using spectrophotometer (HACH DR 2000).

### 3. RESULTS AND DISCUSSION

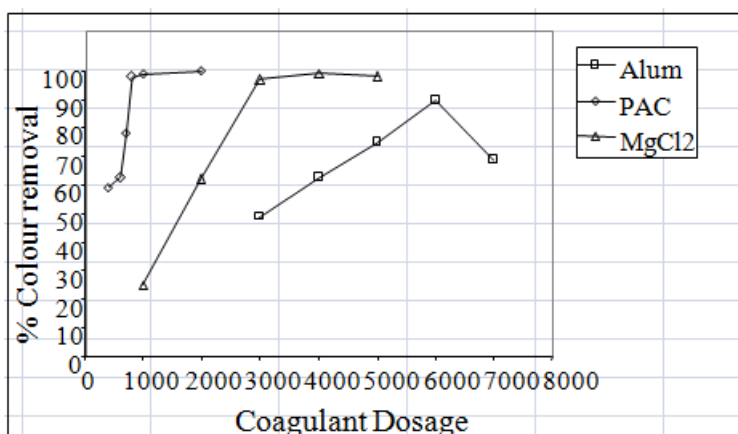
The effectiveness of various coagulation-flocculation treatments for reducing colour in reactive dyes solution was studied. A matrix variable was developed, in which the amount of coagulant and coagulant aid dosages were varied and applied to each 150 mL reactive dye solution. The results for the treatment of raw textile wastewater are summarized in Table 1.

**Table 1:** Treatment of industrial textile wastewater with Alum, MgCl<sub>2</sub> and PAC

Coagulant	Dosage, mg/L	pH	Colour Removal		
			Initial	Final	% Reduction
Alum	3,000	5	12,424	6,336	49
	4,000	4.7	12,450	4,607	63
	5,000	4.3	12,450	3,113	75
	6,000	2.4	12,424	1,242	90
	7,000	4.0	12,450	1,370	89
PAC	400	5.5	12,750	5,228	59
	600	5.6	12,750	4,718	63
	700	4.9	12,750	2,805	78
	800	4.5	17,200	344	98
	1,000	4.3	12,700	127	99
	2,000	4.1	12,250	0	100
MgCl <sub>2</sub>	1,000	11.6	12,750	9,563	25
	2,000	10.1	12,700	1,270	62
	3,000	10.6	12,400	372	97
	4,000	10.4	17,233	173	99
	5,000	10.3	17,233	345	98

#### 3.1 The Effect of Coagulants on Colour Removal

The performance of coagulants (alum, MgCl<sub>2</sub>, and PAC) for color removal is shown in Figure 2. 6,000 mg/L of alum was required achieve 90% colour removal of reactive dye. For alum, an initial dosage of 1,000 mg/L gave a significant increasing linear curve until the dosage achieved 3,000 mg/L, and the curve achieved >90% colour removal. Once the dosage passed the 4,000 mg/L peak, the percentages of colour removal were reduced because during this stage, the dye solution had been removed and had already achieved the optimal dosage. Hence, the suitable dosage of alum to achieve > 90% for colour removal is between the dosages of 3,000 to 5,000 mg/L.



**Figure 2:** Colour removal of different dosages of coagulants

The percentage removal of dye increased tremendously from 25% to 62% when the concentration of MgCl<sub>2</sub> concentration increased from 1,000 mg/L to 2,000 mg/L, and the percentage removal continued to increase up to 97% when 3,000 mg/L of MgCl<sub>2</sub> was used. According to Figure 2, the percentage of colour removed by alum at optimum

condition increased as the dosage was increased from 1,000 to 6,000 mg/L, achieving a maximum of 78.5% removal. However, the percentage of colour removal decreased with the increase of dosage to 7,000 mg/L. This may be due to the restabilization of dye particles by excess alum hydrolysis species (Wong et al., 2007). Further increase in the  $MgCl_2$  dosage could only increase the percentage removal of dye slightly.

The percentage of colour removal of reactive dye achieved >100% at the dosage 2,000 mg/L of PAC. The percentage removal of dye increased tremendously from 59% to 78% as the dosage of PAC increased from 400 mg/L to 700 mg/L. Further increase in the PAC dosage could only increase the percentage removal of dye slightly, and it did not provide a significant increase in the percentage removal. For treatment with PAC and alum, the highest percentage of colour percentage was achieved when 800 mg/L PAC was used, and further increase in the PAC dosage decreased the percentage of colour removal. The treatment of PAC on the dispersed dye also showed a similar trend in the percentage of colour removal as in alum. Further increase in the coagulant dosage after the optimal dosage did not increase the percentage of colour removal as all dyes had been removed at this stage. Excessive coagulant would result in the formation of excessive flocs, which in turn would prolong the settling time. For reactive dye, the same phenomenon occurred in the treatment with PAC and alum. After the highest percentage of colour removal was achieved when 800 ppm of PAC was used, further increase in the PAC dosage decreased the percentage removal of colour.

It can be seen that  $MgCl_2$  and PAC achieved higher colour removal than at low dosage. However, the difference in colour removal efficiency became small with increasing coagulant dosage. Among the coagulants used, PAC was the most effective coagulant in treating each dye with the colour removal up to 100% at the dosage of 800 mg/L. PAC was more effective than  $MgCl_2$  and alum, as >99% colour removal was attained using smaller quantity of the coagulant.

### 3.2 The Effect of Coagulant Dosage on Settling Time

The flocs settling time of different types of coagulants were investigated, and the results are shown in Figure 3. Settling time is very important to the study in order to identify whether the dosage of coagulant used works effectively with the least time required to settle the particles inside the wastewater. The result showed that the flocs settling time increased as the coagulant dosage increased. The flocs settling time for the treatment with PAC was longer than the settling time for the treatment with alum and  $MgCl_2$ . The settling time for PAC dosage of 600 mg/L, alum dosage of 5,000 mg/L and  $MgCl_2$  dosage of 5,000 mg/L were 420 s, 435 s and 495 s, respectively. PAC dosage of 800 mg/L with the removal of 98% had settling time of 352 s. Although higher dosages of PAC gave >98% removal, the settling time was more than 500 s, which is not preferable in the wastewater treatment plant as it would increase the size of the settling tank.

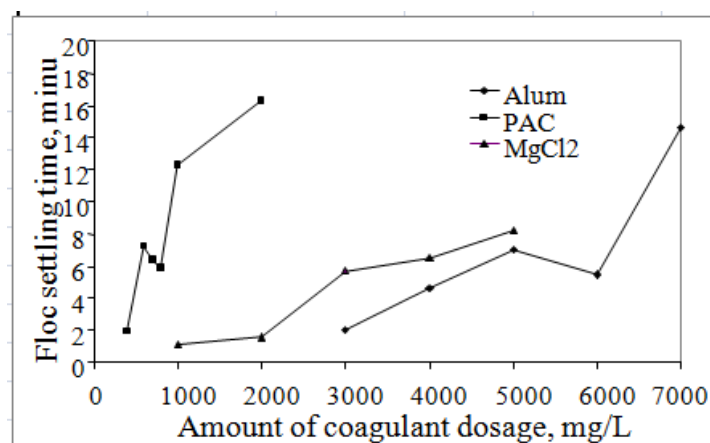


Figure 3: The effect of coagulant dosages on flocs settling time

The settling time for the treatment using  $MgCl_2$  did not exceed 500 s for the coagulant dosages studied. The longest settling time (495 s) of the treatment occurred when 5,000 mg/L of  $MgCl_2$  was used. The flocs formed in the treatment with  $MgCl_2$  were larger and easier to settle compared to flocs formed in the treatment with PAC. The size of the flocs formed using  $MgCl_2$  was observed to be much bigger than those formed using alum and PAC.

It was observed that for alum dosage of 6,000 mg/L, the removal of 90 dye had settling time of 330 s. The increase of alum dosages led to lower removal (69%), with the settling time exceeded 500 s. Even though alum possessed the lower settling time among other coagulants, the percentage of removal was not as effective as PAC. Therefore, PAC had the most effective percentage of removal and also the lower settling time, which is preferable in the wastewater treatment plant (Mohd et al., 2009) also conducted an investigation of reactive dye, and discovered that as the dye concentration

increased, the time required to achieve identical color removal also increased. Excessive coagulant would result in forming of too much flocs, which prolong the flocs settling time.

### 3.3 The Effect of Coagulant Aid on Settling Time

Polyelectrolyte, Koaret PA 3230 was used as a coagulant aid to build a bridge between colloidal particles to form into larger flocs and easier to settle. The optimal pH was kept constant for each dye solution. Figure 4 shows that the colloidal particles formed into large flocs without coagulant aid for alum, which gave a settling time of 295 s. A significant improvement occurred when the dosage of coagulant aid increased to 5.0 mL but the flocs settling time would increase the settling time if 4.0 mL alum was used.

In the case of  $MgCl_2$ , the use of 1.0 mL coagulant aid reduced the settling time to 280 s. The coagulant also needed 4.0 mL of coagulant aid in order to reduce the flocs settling time to less than 50 s. For  $MgCl_2$ , the addition of 4.0 mL of coagulation aid gave a significant improvement when the flocs settling time decreased from 100 s in 3.0 mL of coagulation to 55 s in 4.0 mL of coagulation aid.

From the figure, there was an increase in flocs settling time of about 980 s when 4.0 mL of coagulant aid was used for 600 mg/L dosage of alum and 800 mg/L dosage of PAC. A significant improvement did not occur for further increase of the dosage. In this case, the addition of 2.0 mL coagulant aid reduced the flocs settling time from 258 s to 128 s, but the flocs settling time increased when 5.0 mL of coagulant was added to dye solution.

It was observed that the  $MgCl_2$  without addition of coagulant aid gave higher flocs settling time of 540 s compared to alum and PAC, which gave much lower flocs settling time of 295 s. By using  $MgCl_2$ , shorter flocs settling time was achieved compared to alum and PAC due to the larger size and weight of the flocs formed in the treatment with  $MgCl_2$ , which made the flocs easier to settle compared to flocs formed in the treatment with PAC. The flocs settling time using  $MgCl_2$  was reduced with the increasing amount of coagulant aid. In case of alum and PAC, with the use of additional coagulant aid produced heavy flocs with good settling time velocity. It was found that the coagulation of alum and PAC with additional coagulant aid was better than that of  $MgCl_2$ .

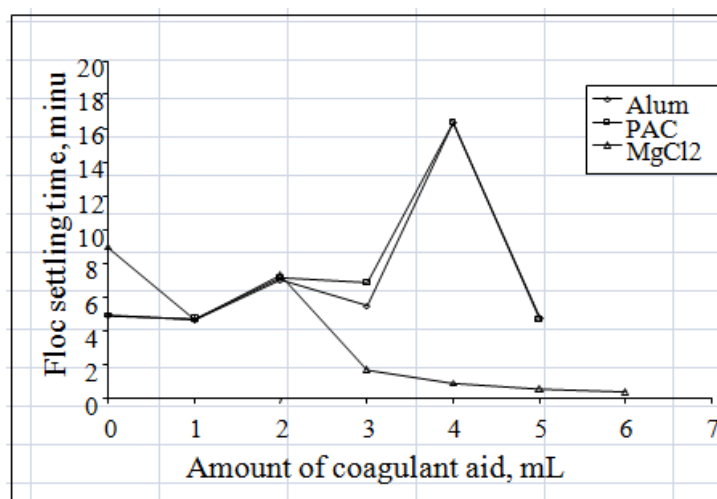


Figure 4: The effect of the amount of coagulation aid on flocs settling time

### 3.4 The Effect on pH

pH effect is practically important in treating dye containing wastewater, since acidic dyeing conditions can lead to acidic discharge wastewater. The percentage of colour removal for reactive dye wastewater is greatly affected by the pH solution with alum and  $MgCl_2$  as shown in Figure 5. The results of pH effects for PAC are expressed in relative to the removal of reactive dye wastewater as shown in Figure 6. The adsorption of the dye solution was barely effected by pH changes and also reduced moderately at alkaline region. The explanation for the negligible effect of pH is probably associated with the electrostatic interactions.

The maximum pH range for  $MgCl_2$  was in the range of 10.0 to 12.0. The removal of dye increased tremendously in the range of 25% to 62% when the  $MgCl_2$  concentration increased from 1,000 mg/L to 2,000 mg/L, and it continued to increase for further addition of  $MgCl_2$ . The result obtained for the treatment using  $MgCl_2$  is in agreement with those of Wong et al., (2007).

The treatment with 1,000 mg/L of PAC gave 99% removal of dye in the pH range of 3 to 5, and a phenomenon of two peaks in the pH range occurred when the concentration of PAC was increased. The peaks were observed, where the first pH range occurred at the acidic zone between 3 to 6, while the second pH range occurred between 6 to 9. The percentage removal of dye in the acidic zone was almost 100% for PAC dosage. The result obtained in Figure 6 for the treatment using PAC is in agreement with those of Omar et al., (2013).

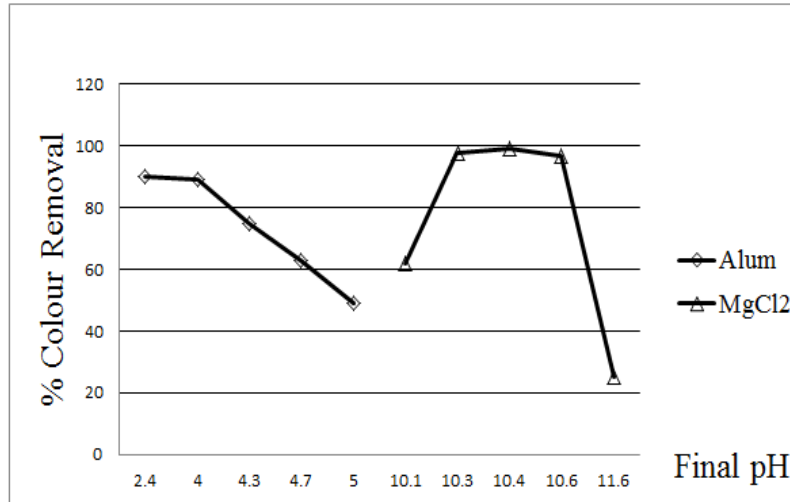


Figure 5: The effects of pH of reactive dye wastewater with MgCl<sub>2</sub> and Alum in the range of 1,000 to 5,000 mg/L

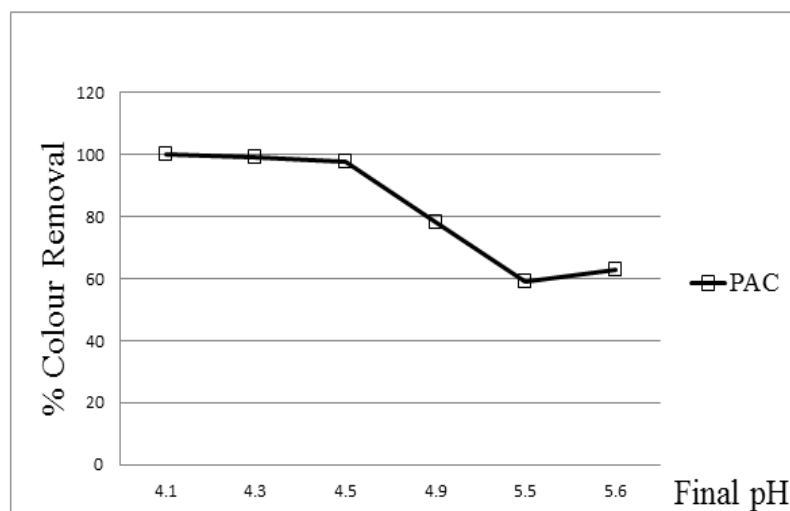


Figure 6: The effects of reactive dye wastewater with PAC in the range of 1,000 to 5,000 mg/L

The result obtained for PAC treatment is in agreement with Maryam (2008), who studied on the effect of coagulant dosage on dye removal efficiency. Different amounts of PAC and alum were added into the dye-containing solutions. Dye concentration was kept constant at 100 mg/L and pH was adjusted to 4 and 5 (optimum pH) for PAC and alum, respectively. Alum and PAC were more effective in a lower pH range but MgCl<sub>2</sub> was more effective in a higher pH range. For MgCl<sub>2</sub>, the colour removal was more effective in the alkaline pH range compared to alum and PAC that were more effective in the acidic range.

The figure above shows that alum was soluble in the pH range from 6.0 to 6.3 but consequently, the coagulation at this pH range converted most of the added alum to solid flocs particles and minimized residual soluble. Alum was more effective in a lower pH range and it fell in the range of 2.0 to 5.0. It is advantageous for coagulation with alum in this pH region for sweep flocs coagulation in order to minimize the residual of aluminium. Furthermore, it helps to utilize a greater fraction of dissolved positively aluminium charged compared to higher pH coagulation. The removal of dye increased tremendously in the pH range of 2.4 to 4.3 when the concentration increased from 5,000 mg/L to 6,000 mg/L. Coagulation below pH 7 means that this highly charged aluminium would be available to destabilize particles by charge neutralization and react with negatively charged colloids.

For the three types of coagulants, the effect on pH adsorption varied as shown in Table 2. Reactive dyes are strongly acidic, thus considerable changes in charge properties are not expected even over a wide range of pH. However, in addition to those electrostatic interactions, other interacting forces are required to explain the different adsorption pattern of this reactive dye. In this case, the removal of dyes at high pH may be caused by competing hydroxide anions. It was found that the lower removal of reactive dye at alkaline pH was caused by the presence of excess OH<sup>-</sup>, which competes with the dye anion.

Table 2 shows that the optimum pH ranges of the PAC treatment are broader than that of the alum treatment. This is in agreement with the result of Aguilar et al., (2005). Therefore, the PAC treatment is less sensitive to pH compared to the alum treatment. This is due to the pre-polymerized species that are partially hydrolyzed and already present in the PAC solution. Hence, the PAC treatment is relatively less sensitive to pH (Aguilar et al., 2005). Table 2 also shows that different ratios of disperse dye to reactive dye in the mixed dye solutions do not have any influence on the optimum pH range for any of the three coagulants studied.

**Table 2:** Optimal pH range for different coagulants

Coagulant	Coagulant dosage (mg/L)	Optimal pH range
Alum	6,000	2.0 to 5.0
PAC	800	4.0 to 6.0
MgCl <sub>2</sub>	3,000	10.0 to 12.0

#### 4. CONCLUSION

Up to 90 %, up to 99% and 100% colour removal of reactive die were achieved using alum of concentration 6,000 mg/L at pH 2.4, MgCl<sub>2</sub> of concentration 4,000 mg/L at pH 10.4 and PAC of concentration 2,000 mg/L at pH 4.1, respectively. Among the coagulants used, PAC was the most effective coagulant in dye treatment with the colour removal up to 100% at the dosage of 800 mg/L. The flocs settling time for the treatment with MgCl<sub>2</sub> was shorter. In addition, the flocs settling time for the treatment with MgCl<sub>2</sub> was shorter than that of PAC, and the flocs settling time for the treatment with alum was longer than of MgCl<sub>2</sub> and PAC.

#### 5. ACKNOWLEDGEMENT

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