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Determination of The Best Available Coagulation/Flocculation Technology with Novel Pre-hydrolysed Coagulants for Colour Removal from Biologically Treated Textile Wastewater

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ABSTRACT

Chemical coagulation/flocculation technology has a complex structure involving various inter -related parameters, since it is very critical to define that how well coagulant will function under given conditions. The literature studies have reported that novel pre-hydrolysed coagulants such as Polyaluminium chloride (PACl), Polyaluminium ferric chloride (PAFCl), Polyferrous sulphate (PFS) and Polyferric chloride (PFCl) seem to give better colour removal even at low temperature and may also produce lower volume of sludge. In this connection, the effectiveness of various novel pre-hydrolysing coagulants for the treatment of textile wastewater have been studied in the literature recently and these studies mostly reported that both they don't need pH correction, additional coagulant aid exct. and they seem to give better colour removal even at low temperature and may also pr oduce lower volume of sludge compared to conventional coagulants. By the way, there are very limited studies carried out on the decolourisation of textile wastewater containing multiple dyes of different classes along with the various chemical additives wh ich are used during textile processing (especially denim washing and reactive dyeing), in the literature. So, the aim of this study is to determine the best available coagulation/flocculation technology with PACl and PAFCl as a novel pre-hydrolysed coagulants for colour removal from an aerobic treated textile wastewater including multiple dyes consists of mostly indigo dye together with reactive dye. According to the results, the colour value of biologically treated textile wastewater was measured as 26,2 m^{-1}/ 23,3 m^{-1}/ 23,7 m^{-1} for 436 nm, 525nm 620 nm. The best colour removal efficiency obtained from 80 mg/L PACl with 2 mg/L Anionic Polyelectrolyte at pH 6,98 was calculated as 74% while sludge prduction rate calculated as 79 kg/day for 1500 m3/day wasteawter. Besides, 10 mg/L PAFCl and 2 mg/L anionic polyelectrolyte at pH 6,98, which is the real pH of the investigated Wastewater Treatment Plant (WWTP) effluent, showed the cost effective result as a coagulant in terms of both colour removal efficiency and sludge production rate with the values 75% and 60 kg sludge/day, respectively,as compared to PACl.

Keywords: Best avalibale coagulation, Novel pre-hydrolysed coagulant, Colour removal, Textile wastewater.

1. INTRODUCTION

On the basis of waste and wastewater (or effluent) generation, the textile mills can be classified into two main groups namely dry processing mills and wet processing mills [1]. Desizing, scouring, bleaching, mercerising, dyeing, printing, and finishing are the main stages of wet processing group. Moerover, the wastewater generated by textile industry generally includes cleaning wastewater, process wastewater, noncontact cooling wastewater, and storm water. The amount of water used varies widely in this industry, depending on the specific processes operated at the mill, the equipment used, and the prevailing philosophy of water use. On account of the involved complexity of different processes at different stages, textile wastewater typically contains a complex mixture of chemicals. Apart from this, large numbers of associated hazards have also been reported by the various chemicals used in different stages of textile processing [2-5).The unused materials from these processes are discharged with wastewater that is high in colour, biochemical oxygen demand (BOD), chemical oxygen demand (COD), pH, temperature, turbidity and toxic chemicals as shown in Table 1.

Table 1. Major characteristics of real textile wastewater studied by various researchers [6]

pH	COD (mg/L)	BOD_5 (mg/L)	TSS (mg/L)	TDS (mg/L)	Colour	Turbidity (NTU)	References
8.8–9.4	595 ± 131	379 ± 110	276 ± 76	–	–	–	El-Gohary and Tawfik, 2009
11.2	2276	660[a]	–	47.9	–	–	Golob et al., 2005
5–10	1100–4600	110–180	–	50	1450–1475(ADMI)	–	Dos Santos et al., 2007
6.5–8.5	550–1000	–	100–400	–	7.50–25.50[b]	15–200	Ciabatti et al., 2010
2.7	7000	–	440	930		2140	Al-Malack et al., 1999
13.56	2968	–	–	–	3586 (C.U)	120	Joo et al., 2007
12–14	1500–2000	–	–	–	Dark blue	–	Gozalvez-Zafrilla et al., 2008
10	1150	170	150	–	1.24^{436nm}	–	Selcuk, 2005
9	750	160	–	–	–	–	Schrank et al., 2007
2–10	50–5000	200–300	50–500	–	>300 (C. U)	–	Lau and Ismail, 2009
8.32–9.50	278–736	137	85–354	1715–6106	–	–	Phalakornkule et al., 2010
8.7 ± 0.2	17900 ± 100	5500 ± 100	23900 ± 50	1200 ± 50	–	–	Rodrıguez et al., 2008
9.30	3900	–	–	–	–	240	Paschoal et al., 2009
7.8	810 ± 50.4	188 ± 15.2	64 ± 8.5	–	0.15^{669nm}	–	Haroun and Idris, 2009
13 ± 1	2300 ± 400	–	300 ± 100	–	–	–	Debik et al., 2010
6.95	3422		1112	–	–	5700	Bayramoglu et al., 2004
7.86	340	210	300	–	>200 (Pt-Co)	130	Merzouk, 2010
7.5 ± 0.3	131 ± 18	–	75 ± 13	1885 ± 80	–	–	Ustun et al., 2007

[a] BOD_7 and effluent is from reactive dye bath.
[b] Integral of the absorbance curve in the whole visible range (400–800 nm), ADMI : American dye manufacturer institute, C.U: Colour Unit.

The widely used methods for dyeing wastewater treatment involve many physical–chemical techniques, such as coagulation, adsorption, membrane filtration, and advanced oxidation, etc. [7–10]. Since Coagulation flocculation is cost effective technology and gives excellent colour removal for wide variety of dyes, it becomes promising technology for decolourisation of textile wastewaterChemical coagulation is a complex phenomenon involving various inter-related parameters, hence it is very critical to define that how well coagulant will function under given conditions. On the basis of effectiveness to decolourise the textile wastewater, chemical coagulants can be categorised in the three parts such as hydrolysing metallic salts, pre-hydrolysing metallic salts and Synthetic cationic polymers, respectively [6]. It has been reported that pre-hydrolysed metallic salts are often found to be more effective than the hydrolysing metallic salts such as aluminium sulphate (alum), ferric chloride and ferric sulphate those are readily soluble in water [11]. Prehydrolysed coagulants such as Polyaluminium chloride (PACl), Polyaluminium ferric chloride (PAFCl), Polyferrous sulphate (PFS) and Polyferric chloride (PFCl) seem to give better colour removal even at low temperature and may also produce lower volume of sludge. In this connection, Gregory and Rossi (2001) have studied more rapid flocculation and strong flocs than that of alum at equivalent dosage [12]. This can be attributed by the fact that these coagulants are pre-neutralised, have smaller effect on the pH of water and so reduce the need of pH correction. Various authors have suggested the most important parameters to be consider in coagulation are pH and concentration of applied metal ions (coagulant) [6] PACl shows better colour removal efficiency in a wider pH range of 7-10. Various researchers have also revealed that the addition of polyelectrolyte generally increases turbidity and volume of settled sludge. This undesired effect may be eliminated if the used concentration of polyelectrolyte is less than 2 mg/L [13]. PAFCl combines the coagulatory advantages of both aluminium and iron salts and hence able to form flocs rapidly with more bulky and rapid sedimentation. This novel coagulant is not widely studied for textile wastewater treatment hence very limited information is available. The principle mechanism of PAFCl is charge neutralisation and bridging [14]. Some of the reported chemical coagulation technology and their performance have been summarised in Table 1 [6].

Table 1. Effectiveness of different chemical coagulants studied by different researchers for colour removal of textile wastewater [6]

Name of coagulant	Optimised dose (mg/L)	Coagulant aids (if any)	Type of dyes present	Optimum pH	% Colour removal	Reference
Steel industry wastewater			Disperse	4.25	99	Anouzla et al., 2009
Potassium ferrate	100	Polyamine based polymer		6.5–8.5	95	Ciabatti et al., 2010
Polyaluminium Chloride (PACl)	10			7.2	99.9	Choo et al., 2007
Poly-epichlorohydrin-diamine	20			7	95	Kang et al., 2007
Alum	200	Polyacrylamide based polymer (Cytec)		5.3	78.9	El-Gohary and Tawfik, 2009
Alum	5000	Copper sulphate as catalyst		4	74	Kumar et al., 2008
Alum	20	Commercial cationic flocculant (Colfloc-RDeCiba)	Reactive and acid	Near to neutral	98	Golob et al., 2005
Alum	7×10^4			5.7–6.5[a]	74	Patel and Vashi, 2010
Ferrous Sulphate	200	Polyelectrolyte	Sulfur	9.4	90	Bidhendi et al., 2007
Ferric chloride	400		Sulfur	8.3	100	Bidhendi et al., 2007
Ferric chloride	293		Reactive and disperse	6	71	Kim et al., 2004
Ferric chloride	56	Cationic polymer		4	92	Suksaroj et al., 2005
Magnesium chloride	400	Polyelectrolyte (Koaret PA 3230)	Reactive	11	85	Tan et al., 2000
Magnesium chloride	120	Lime		11	100	El-Gohary and Tawfik, 2009
Magnesium chloride	800	Hydrated lime	Reactive and disperse	12	98	Gao et al., 2007
Polyaluminium chloride (PACl)	0.1	Poly acrylamide-seed gum	Reactive, acid and direct	8.5	80	Sanghi et al., 2006
Polyaluminium chloride (PACl)	800	Anionic polyacrlamide, Exerfloc 204		7.5	75	Tun et al., 2007
Ferrous sulphate	400	Lime and Cationic polymer	Reactive	12.5	90	Georgiou et al., 2003
Ferrous sulphate	1000	Anionic polyelectrolyte (Henkel23500)		9.5	60	Selcuk, 2005
Ferrous sulphate	7×10^4			5.7–6.5[a]	85	Patel and Vashi, 2010
Ferric sulphate	7×10^4			5.7–6.5[a]	58	Patel and Vashi, 2010

So, the aim of this study is to determine the best available coagulation/flocculation technology with PACl and PAFCl as a novel pre-hydrolysed coagulants for colour removal from an aerobic treated textile wastewater including multiple dyes consists of mostly indigo dye together with reactive dye. According to the results of chemical treatability of this wastewater, the optimum conditions (pH, coagulant dosage, sludge volume exct.) will be described and cost-comparative alternative will be defined for the superior colour removal for this industry.

2. MATERIAL AND METHODS

2.1. Materials

2.1.1. Wastewater

This study was carried out in the wastewater treatment system treating denim washing (70%) and reactive dyeing (30%) wastewater located in Çorlu town of Tekirdag. Treatment system has a 1500 $m^3.day^{-1}$ capacity composed of physical and biological treatment units including conventional activated sludge units. Treated wastewater is discharged in a receiving body called Sinanlı stream. Experimental studies were carried out effluent samples characterized for 1 month of this treatment plant. While the color of the effluent was brownish dark blue which was caused by main dyes – indigo (from denim washing processes) and reactive dyes (from cotton based fabric dyeing processes), the organic content of it low concentrations.

2.1.2. Coagulants and Coagulation/Floculation test procedure

PACl and PAFCl was chosen as coagulant for color removal in biologically treated textile wastewater. Solid PACl product was obtained from Shanghai Sungo Technology&trade Co (Shanghai/ China, $Al_{13}O_4OH_{24}(H_2O)_{24}(H_2O)_{127}^+$ and 10-34,8% purity) and PAFCl was obtained from Henan Allrich Chemical Co., Ltd. (Henan/China, Al2(OH) ncl6- nm Fe2(OH) ncl6- nm(SO4) x and 30% purity) were used as received without further purification. Solid Anionic Polyelectrolyte was supplied from Zhengzhou Jing Lian Water Purification Materials Co., Ltd. (Henan, China, polyacrlamide based, 99% purity). Real textile wastewater was used in this study which was taken from the investigated wastewater treatment plant mentioned above. All coagulation/flocculation experiments were conducted in 1.0 liter glass beakers using a conventional Jar–test apparatus (VELP Scientifica, FC6S) equipped with four beakers with two different pre-hydrolized coagulants which were added to 1000 ml of the wastewater. 1000 ml stock solutions of each polymer were prepared 10 % w/w. 1ml (2 mg/l) anionic polyelectolite was used as coagulant aids for each coagulant. The solutions were stirred rapidly at 200 rpm for 2 min during the addition of coagulants, followed by slow stirring at 45 rpm for 15 min and settling for 30 min. After settling, supernatant samples were collected and filtered using coarse filter for further analysis. Optimum conditions are assesed for Colour and COD removal before treatability experiments were started. Jar test trials were made at efficient pH intervals, mixing rate and flocculant dosage which is determined in treatability pre-studies for used each coagulant. So, the pH of the samples were adjusted to 4, 6.98(real pH), 9, 10 by adding 1N HCl or NaOH solutions for both PACl and PAFCl. After then, supernatant and sludge characterization was carried out on the sampels taken from supernatant and completly mixed phases for the obtained best result beaker, respectiveley.

2.1.3. Analytical Methods

All analyses were performed according to the Standard Methods [15] except COD and Colour removal. The COD and Colour were measured according to ISO 6060 Method [16] and ISO 7887 method [17]. The adjustment and measurement of pH was carried out using a (WTW pH315i) pH meter.

3. RESULTS AND DISCUSSION

3.1. Effluent Characterization

Wastewater characterization of investigated plant is given Table 2.

Table 2. Investigated WWTP wastewater characteristics

PARAMETERS	UNIT	Raw WW	Effluent of WWTP	Discharge Criteria
Total COD	$mg.L^{-1}$	495	106	300**
Soluble COD (0,45μ)	$mg.L^{-1}$	450	80	-
Suspended Solid (SS)	$mg.L^{-1}$	85	6	100**
Conductivity	$\mu mho.cm^{-1}$	-	2843	-
Colour436 nm	CN*(m^{-1})	-	26,2	7***
525 nm	CN*(m^{-1})	-	23,3	5***
620 nm	CN*(m^{-1})	-	23,7	3***
pH	-	6,3	6,98	6-9

*CN: Colour number, **WPCR: Water Pollution Control Regulation, *** discharge criteria defined in the European Standard EN ISO 7887 for receiving environment.

Table 2 shows that colour parameter as a pollution parameter does not achieve the discharge 4 criteria defined in the European Standard EN ISO 7887 for receiving environment although organic content and pH values are enough low for aquatic media . So, it need to remove with a best avalilable treatment technology before discharged.

3.2. Jar test

3.2.1. PACl experiment

The most important parameters in coagulation are pH and concentration of applied coagulant. According to the pre-studies, PACl shows better colour removal efficiency in a wider pH range of 7-10 [13]. So, in this study, the pH values were arranged as 4, 9, 10 for PACl and 4, 8, 9 PAFCl, respectively. Coagulation carried out also at pH 6.98, because this pH is the real wastewater level. The tests carried out with PACl at pH 10 was canceled because of poor floc formation. Best removal was achieved with 80 mg/l PACl dosage at pH 4 and pH 6,98. As a coagulant aid, anionic polyelectrolyte dosage was choosen 1 ml (1/500, 2 mg/l). Same PACl dosage gave sufficent floc formation for each pH, except pH 9. Especially, polyelectrolyte addition caused reduction in color removal efficiency at pH 9. Table 3, Figure 1 and Figure 2 show that colour removal efficiencies and sludge production rates obtained from jar tests carried out with PACl coagulant at which best coagulation results observed, respectively. According to the Table 3, Figure 1 and Figure 2, the best colour removal rate (74%) was evaluated for PACl (80 mg/L) with Anionic Polyelectrolyte (2 mg/L) at pH 6,98, which is the real pH of the WWTP effluent, although sludge prduction rate appears 21% higher than pH 4 test result, since it will require pH adjustment before discharge to receiving environment. These results have been found consistent with the literature [18,19].

Table 3. Colour removal and Sludge production determined for PACl coagulant

		Conductivity	**COLOUR (Colour Number, m^{-1})**			**Solid Matter Percentage of the sludge (kg SS/100 L)**	**Sludge Production rate**	
Parameters	**pH**	**µS/cm**	**(436 nm)**	**(525 nm)**	**(620 nm)**	**%**	**kg/day**	**m^3/day**
Raw WW	6,98	2013	26,2	23,3	23,7	-	-	-
80 mg/L PACl (%10)	4,00	2193	10,2	7,7	6,0	*	*	*
	Colour Removal (%)	**	61	67	75			
	6,98 (Real)	2036	13,8	11,7	10,0	0,54	98	18
	Colour Removal (%)	**	**47**	**50**	**58**			
	4,00	2184	11,7	7,7	5,8	0,25	56	23
	Colour Removal (%)	**	**55**	**67**	**76**			

80 mg/L PACl (%10) + 2 mg/L A.Poly.	6,98 (Real)	2047	8,2	6,6	6,10,29	79	28
	Colour Removal (%)	**	**69**	**72**	**74**		

*No settling, ** No removal

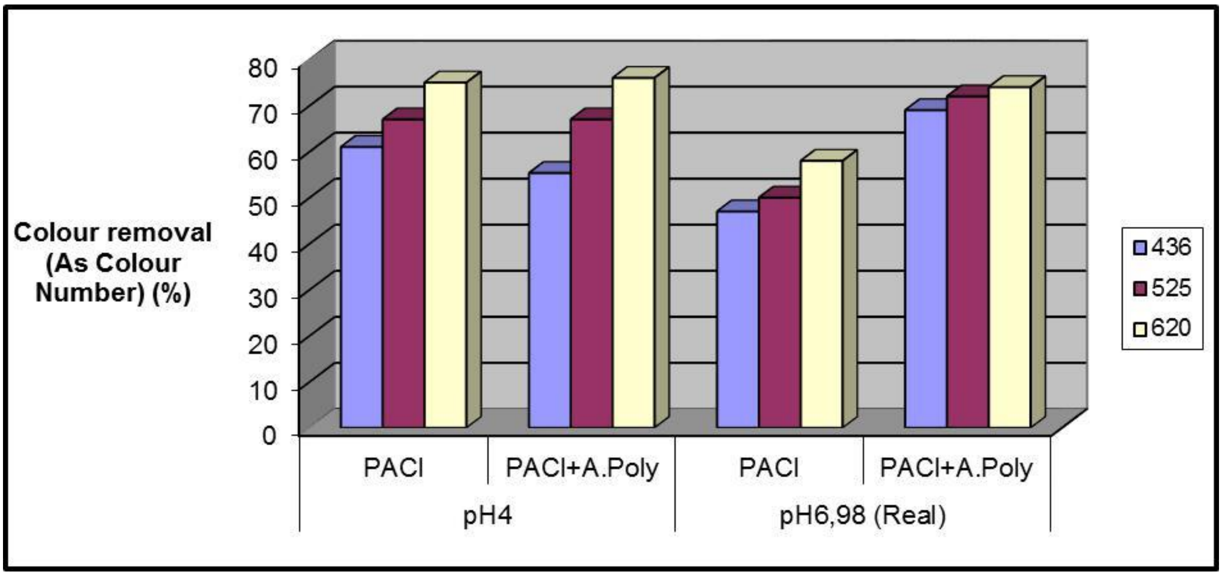

Figure 1. Colour removal efficiencies determined for PACl experiment

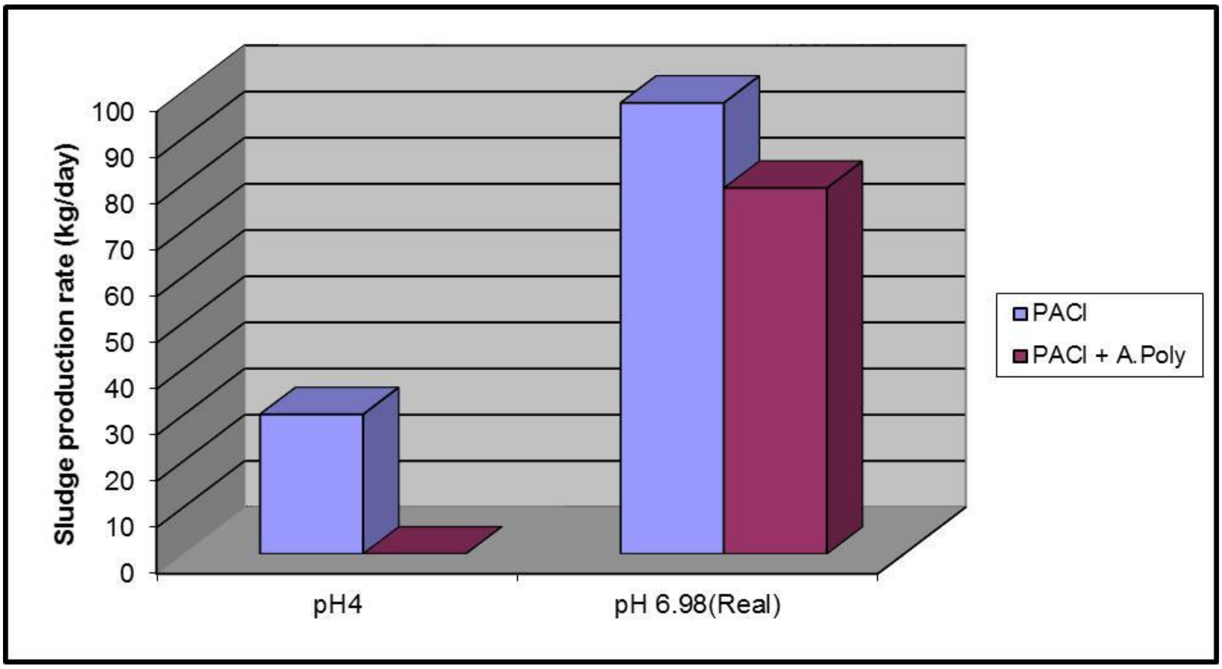

Figure 2. Sludge production rates determined for PACl experiment

3.2.2. PAFCl experiment

PAFCl is not widely studied for textile wastewater treatment hence very limited information is available [20]. Gao et al. (2001) have investigated PAFCl for color removal from petrochemical wastewater [21]. It is reported that PAFCl gives better turbidity removal in pH 7.0-8.4 and better colour removal than other coagulant such as PFS and PACl. Table 4, Figure 3 and Figure 4 show colour removal efficiencies and sludge production rates obtained from jar tests carried out with PAFCl coagulant, respectively. According to theses results, the best coagulation was evaluated as pH 4 and pH 6,98 for both PAFCl and PAFCl with Anionic Polyelectrolyte alternatives, since smaller amount of coagulation was observed at pH 8 and 9. Furthermore, for both alternatives, it is observed that depending on the increased pH, colour removal efficiecy decrease. But, the best colour removals and sludge production rates were obtained from PAFCl with Anionic Polyelectrolyte at pH 4 as 97% , 81 kg sludge/day and at pH 6,98 which is the real pH of the WWTP effluent as 75%, 90 kg sludge/day. At this point, it can be said that since there is no anourmous difference between sludge production rates and colour removal value of both alternatives provides the European Norm EN ISO 7887 discharge criteria determined for colour parameters as 7 m^{-1}/ 5m^{-1}/ 3 m^{-1} for receiving environment, the best coagulation was obtained at pH 6,98 with 10 mg/L PAFCl, 2 mg/L A. Poly. dosages. These results show that although PAFCl gives better colour removal at pH 4 in contrast to the result which was found for petrochemical industry wastewater in the literature [21], 75 % colour removal at pH 6,98 obtained from this study also can be acepted as consistent with literature since it provides European standart.

Table 4. Colour removal and Sludge production determined for PAFCl coagulant

Parameters	pH	Conductivity µS/cm	COLOUR (Colour Number, m^{-1}) (436 nm)	(525 nm)	(620 nm)	Sludge Percentage (kg SS/100 L) %	Sludge amount kg/day	m^3/day
Raw WW	6,98	2013	26,2	23,3	23,7	-	-	-
10 mg/L PAFCl (%10)	4,00	2210	4	2,5	1,9	0,23	68	39
	Colour Removal (%)	**	**77**	**84**	**88**			
	6,98 (Real)	2065	4,8	3,8	3,4	0,18	66	38
	Colour Removal (%)	**	**73**	**76**	**78**			
10 mg/L PAFCl (%10) + 2 mg/L A.Poly.	4,00	2200	1,5	0,8	0,5	0,23	71	32
	Colour Removal (%)	**	**92**	**95**	**97**			
	6,98 (Real)	2072	5,3	4,4	3,9	0,17	60	26
	Colour Removal (%)	**	**70**	**72**	**75**			

*No settling, ** No removal

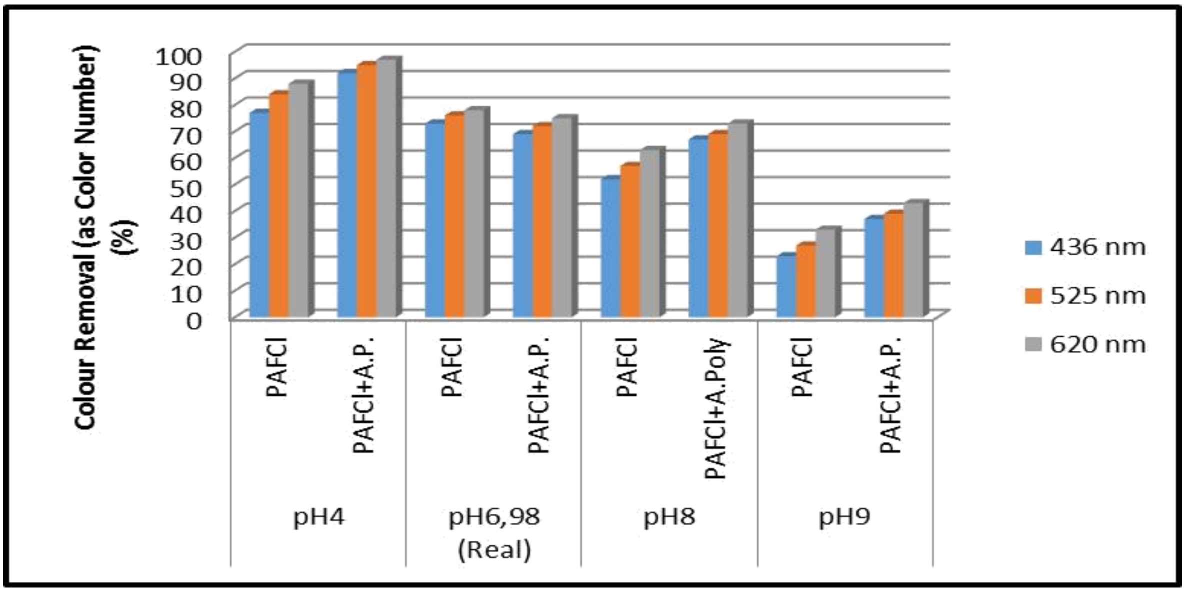

Figure 3. Colour removal efficiencies determined for PAFCl experiment

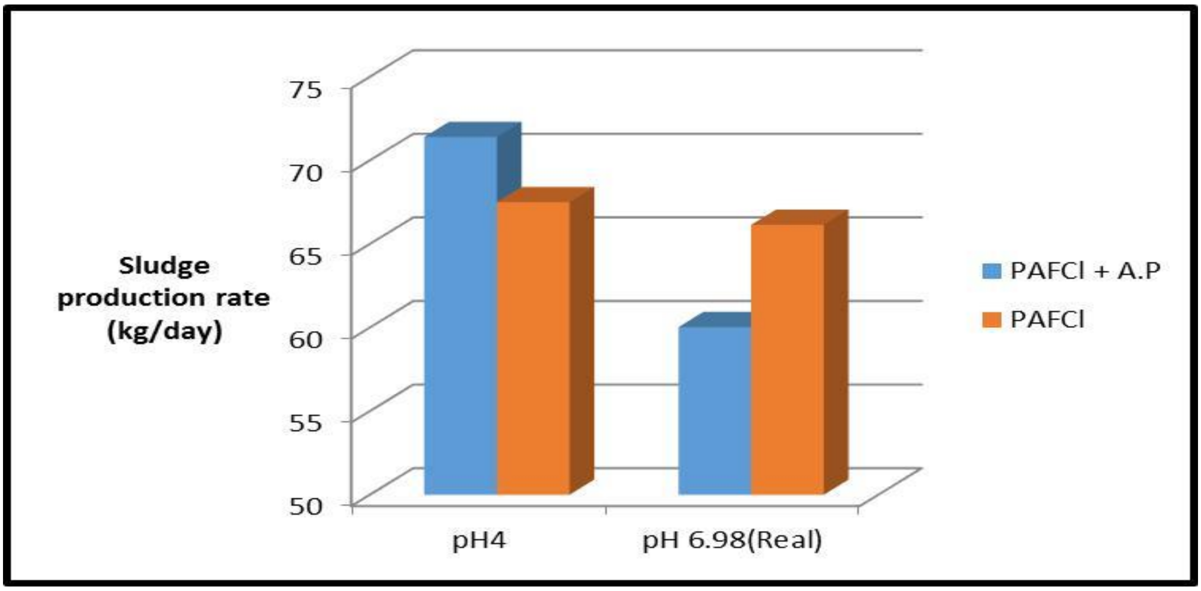

Figure 4. Sludge production rates determined for PAFCl experiment

Lastly, when we compare the best results of both coagulants (PACl and PAFCl), the optimum results (in terms of pH, coagulant dosage, sludge volume) can be summurized at Figure 5 and Figure 6.

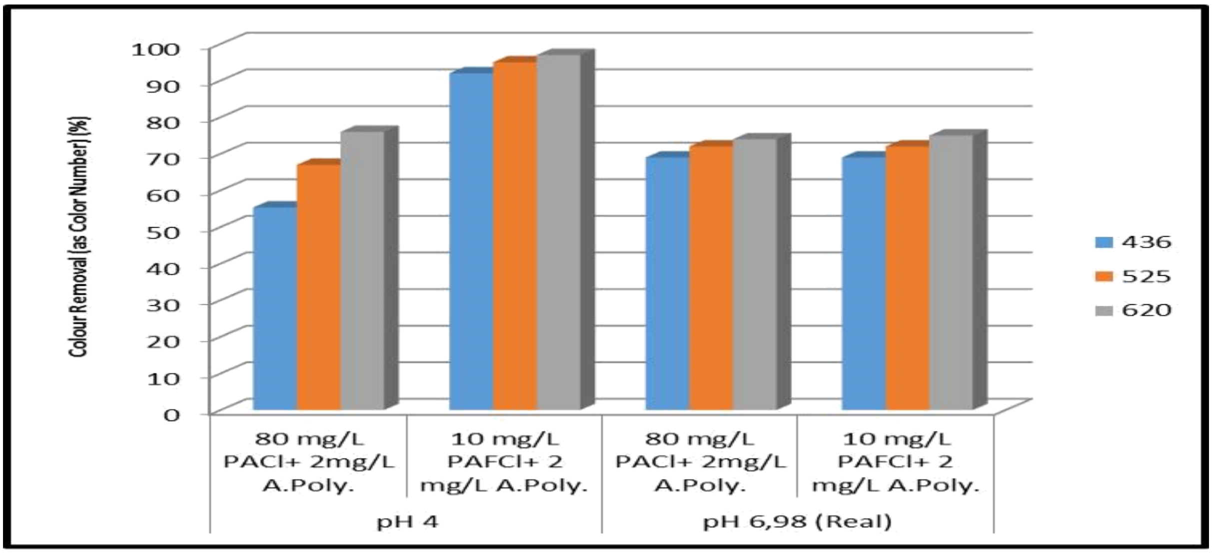

Figure 5. Comparation of the best results of colour removal efficiencies determined for PACl and PAFCl experiment.

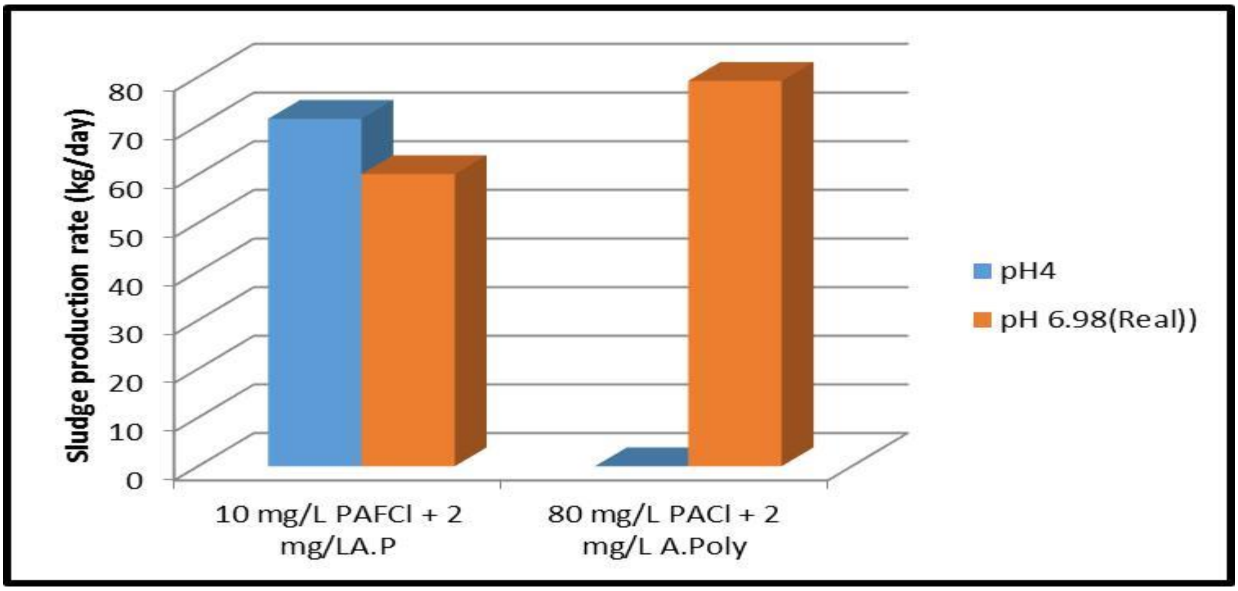

Figure 6. Comparation of the sludge production rate determined for PACl and PAFCl experiment.

According to these results, the best colour removal and sludge production rate were obtained from PAFCl with Anionic Polyelectrolyte at pH 6,98 which is the real pH of the WWTP effluent as 75 % and 60 kg sludge/day.

3.2.2. Cost assesment

Cost assesment for every coagulant implementation should be made to decide the best suitable one for the colour removal of investigated plant wastewater. The cost asses ment containing Colour removal efficiecy and Sludge production rate for both Novel pre-hydrolysed coagulants used in this study has been calculated as 14-24 $ for PAFCl and 32-63 $ for PACl at pH 6,98 which is the real pH of the WWTP effluent, respectively. So, it can be said that PAFCl coagulant is the cost effective option as compared to PACl both color removal efficiency and sludge production rate for this aerobic treated textile wastewater including multiple dyes consists of mostly indigo dye together with reactive dye.

3. CONCLUSION

Waste water of textile industry includes high COD and colour. The colour of textile waste water results in an esthetic problem as well as toxicity for receiving bodies So, the colour in waste water must be removed before discharge to any surface water . The colour removal of textile waste water can not be achieved by conventional treatment processes, completely. So, it can be performed pratically using two methods. First one is intensification of colour in the sludge. The second one is the decomposition of colourful molecules. It is very important to remove the colour from wastewater using suitable, efficient and cost effective method for environment. In this study, first of all, removal of dyestuffs from the wastewater treatment ,which is biologically treated but colour removal is not achieved according to the discharge criteria defined in the European Standard EN ISO 7887 for receiving environment, was investigated by using best avalibale chemical coagulation technology using novel pre-hydrolysed coagulants such as PACl and PAFCl. As a result of the study, PAFCl coagulant was found the cost effective option as compared to PACl both color removal efficiency and sludge production rate for this aerobic treated textile wastewater including multiple dyes consists of mostly indigo dye together with reactive dye.

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