Analysis of WTP sludge usage as Wastewater Quality improvement in Pulp industry with Coagulation-absorption Method

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Abstract

In the global warming issue, water problems are an issue everywhere, including industries along the Pengabuan River. The cycle 10-year long dry season has lead declination of water quality significantly. The level of turbidity increase100-200 NTU become 3500-4000 NTU and conductivity increase from 100 μs become 800 μs, these issues triggered by increased sedimentation and sea water intrusion increase in salt content (NaCl). The pulp industry operating in this river is also impacted. Water plays an important role in controlling quality and production costs, the pulp washing process and the filtrate evaporation process produce wastewater containing lignin compounds which give a brownish colour effect to wastewater effluent even though it was treated in the wastewater treatment plant. The amount of wastewater produced is ± 25 m3/ton of pulp. An alternative method to do breakthrough how this large of wastewater can be reused to reduce dependence of surface water usage and reduce the level of pollutants in the water. Usage of Water Treatment Plant blowdown sludge is one of alternative can be used to capture lignin and reduce colour intensity from 200-300 Pt-co to below 50 Pt-co and parallel reducing COD to below 100 ppm with application of Poly Aluminium Chloride. This research needs to be further developed to looking for process alternatives after lignin captured and the optimum dose of sludge, time, and PAC.

Keywords: *Lignin, Colour, Water reused, Long dry season*

1. Introduction

Water is a basic need in life, the existence of water is so important for all living creatures on this earth. The availability of clean water sources such as rivers, lakes and wells is very much needed. Apart from fulfilling the primary needs of living creatures, water is also needed for other activities such as industry, households, laundry, restaurants, hotels, and others.

To create a healthy environment so that it can support balance in maintaining the epidemiological triangle, physical environmental parameter conditions are needed, such as temperature, humidity, compound content in the air, COD, BOD, TSS and others. In Indonesian Government Regulation No. 7 year 2004 concerning water resources in article 3 it is stated that water resources are managed in a comprehensive, integrated and environmentally friendly manner with the aim of realizing sustainable benefits of water resources for the greatest prosperity of the people. Water is more than just a combination of the chemical's hydrogen and oxygen. Water is a commodity that humans need for various purposes. Water is used for drinking water, industrial raw materials, supporting materials for agricultural, plantation, fishery, and tourism activities, as an energy source for steam and hydropower generating centers. Based on Indonesia's Statistic Center (BPS) clean water needs in Indonesia based on the 2014-2019 survey published in 2020 is as follows:

Tabel 1.1. Treatment Water distribution (PDAM) year 2019

No	Segmen	Jumlah pelanggan	Volume air yang disalurkan (Juta m^3)
	Non Business	13.843.720	2.679,3
	Business and Industry	914.543	476,9
	Social	196.904	103,9
	Specific	30.777	175,1

Source: Indonesian Statistic centre (BPS) year 2020 (Publish no: 05330.2004)

Each water treatment plant having a specific characteristic of raw water. The characteristic of raw water is reference to design the plant size, type, and technology. Pengabuan River is one of raw water source for several industry operated at District Tanjung Jabung Barat. This river has a low and high tide period, during normal season characteristic of the water is acceptable for the treatment water plant as annual basis, but every 10 year cycle recorded an abnormal condition during long dry season, conductivity and turbidity increase significantly. It was recorded in 2004, 2015 and 2023.

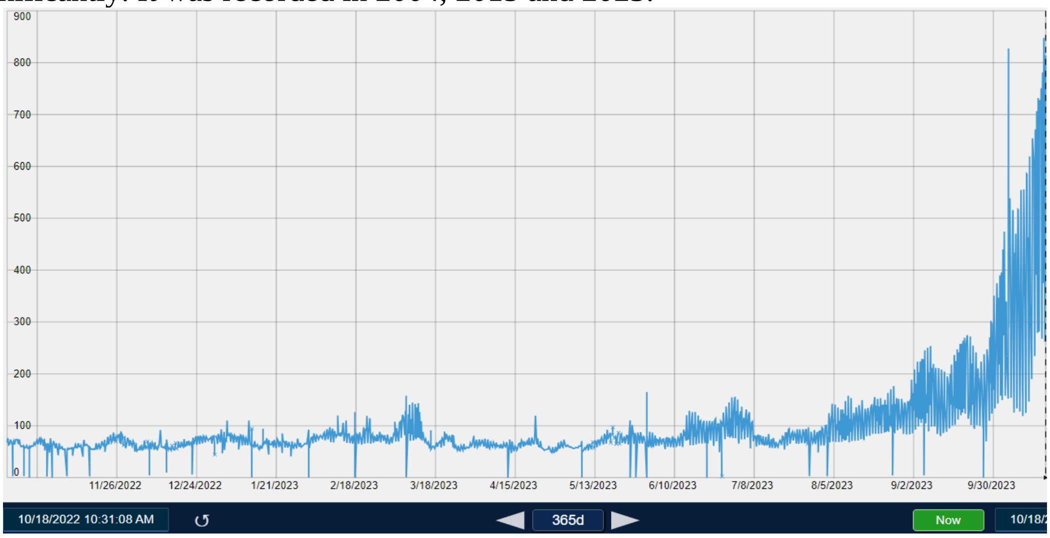


Figure 1.1 Change of Pengabuan River's water conductivity during long dry season 2023, conductivity increase 80 times from normal condition (Source online sensor Pengabuan River's conductivity monitoring Pulp Industry)

Sludge is a residual resulting from water treatment process. Sludge is produced in the sedimentation process and then the sludge processed in the WWTP processing plant before used for other purpose. Disposal of sludge to WWTPs creating additional burden on the COD degradation process by microorganisms and some elements contained in sludge such as Cl- have a toxic effect on microorganisms. The entry of sludge into the wastewater treatment plant will increase the volume of waste, thereby reducing the residence time in the aerated lagoon. And the entry of sludge into the inlet does not have a significant impact without the presence of coagulants/flocculants and does not have much of an impact on reducing the colour level of 200 – 300 Pt-co

The raw water source for the pulp company domiciled in Tebing Tinggi District, West Tanjung Jabung Regency is the Pengabuan River which is 90 km from the coast. The Pengabuan River have high tides and low tides cycles, within 10-year cycle there is a long dry season so that sea water intrusion reaches the raw water intake point so that the raw water contains high levels of salt and processing by sedimentation is ineffective because a small portion of the Chloride element is successfully removed. A high Cl content will lead to corrosion in the piping system. In demineralized water process also facing problem which is leaded by availability of chloride content in the raw water feed will impact to Column ion exchange resin performance. High NaCl content will lead to resin's saturation condition because a lot of Cl elements must be removed. Saturated resin will lead chloride carry over to demineralize water product, the performance of the resin quickly decreases, and regeneration (reactivation of the resin) is required. This regeneration process requires supporting raw materials such as NaOH and HCl. As regeneration is carried out more frequently, damage to the resin structure occurs more quickly and must be replaced immediately, chemical consumption increases, productivity is low due to Output between Regeneration (OBR) and production time becomes smaller.

In facing a situation like the one above, saving water use is a necessity to be implemented to the maximum extent possible, apart from that, the reuse of alternative sources of raw materials needs to be explored, including the reuse of waste, which is important so that the large dependence on raw water sources can be reduced. The impact of long dry season and tidal cycles causes several problems, such as:

1. Companies have difficulty getting clean water sources during long dry seasons on a 5- or 10-year cycle because raw water sources experience an increase in salt content and high turbidity in raw water.
2. High sludge volume in influent wastewater treatment plant creating several problems as such increasing.
3. chloride level from Poly aluminum chloride which can become toxic to microorganism and increasing turbidity will increase sludge blowdown frequency and finally increase influent volume.

2. Methodology

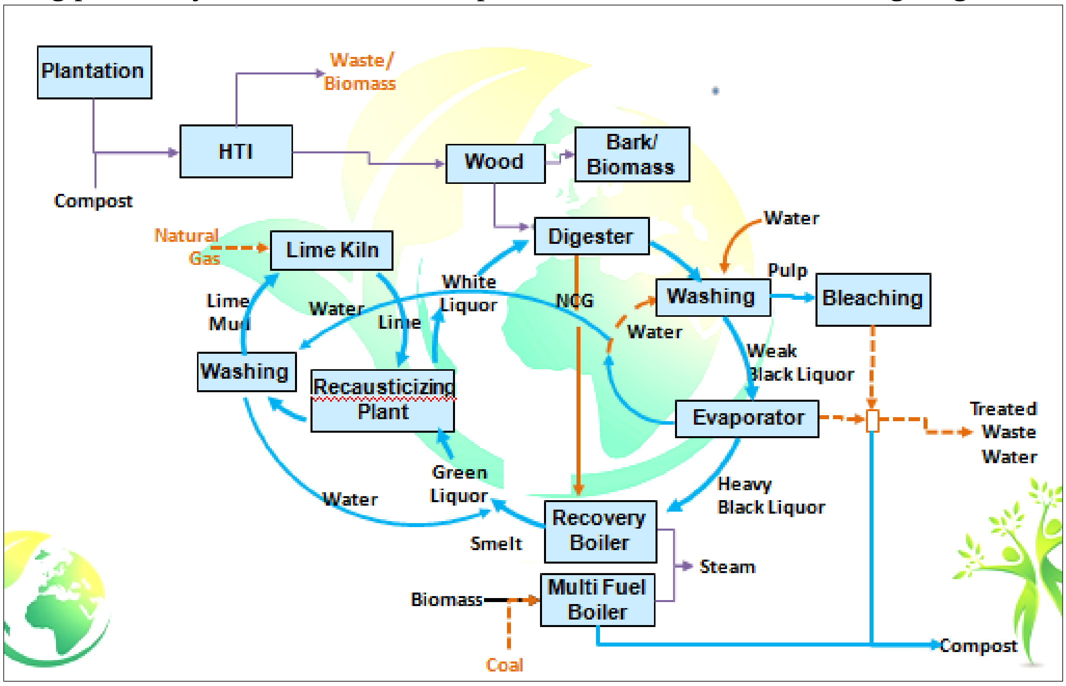
Utilization of water for the various purposes above produces wastewater which can pollute the environment, including clean water sources, if not handled properly. Therefore, various efforts have been made to reduce the level of pollution and the implementation of provisions governing quality standards for wastewater discharge into river bodies as regulated under Indonesian Minister of Environment Regulation No. 5 of 2014 and Jambi Governor Regulation No. 20 of 2007.

Table 2.1. Standards for wastewater discharge from the bleached paper pulp industry

No	Paramater	Unit	Standard
1	pH	-	6 – 9
2	COD	Ppm	< 350 (bleached pulp)
3	BOD	Ppm	<100
4	TSS	Ppm	< 5

Source: Indonesian Minister of Environment No. 5 of 2014 and Jambi No. 20 of 2007

The pulp industry using the bleaching process method is an industry that uses quite a lot of water, each ton of air-dry pulp requires around 20-25 m3 of water. Generally, pulp processing has its own water treatment processing and waste processing system (integrating) to meet wastewater standards that have been regulated in statutory regulations. The paper pulp making process cycle and its water footprint can be seen in the following diagram.



Source: Kraft Recovery Short Course (Douglas W. Reeve, 1999)

Figure 2.1. Pulp Making Process Flow Diagram

The culmination of the process of washing and bleaching paper pulp is in the form of liquid waste which contains components of colloidal organic compounds, hemicellulose Fibers, decomposing agents as well as cellulose, lignin, and others. The content of these compounds causes high levels of turbidity, colour, and high levels of Chemical Oxygen Demand (COD) in wastewater. So that the content of these compounds does not cause pollution to the surrounding environment, a series of wastewater treatment processes are carried out starting from separating solid materials such as Fibers and other solid particles with physical and chemical processes through coagulation and filtering processes. Followed by biological processing to reduce the organic content in the solution so that the COD content is obtained in accordance with the requirements mandated by law. Biological processes using aerobic processes and the use of activated sludge can reduce COD with an efficiency of up to 77%. Meanwhile, further processing has not been carried out for colour because it is not required by statutory regulations. Measurement data shows that the colour intensity of pulp industry wastewater is around 200 – 300 Pt-co. The high intensity of colour in wastewater is caused by the presence of lignin. Lignin in wastewater is a complex organic compound that is difficult to degrade and is a stable colloid.

Rahmadini (2016) the content of suspended particles which are stable colloids and the presence of lignin content results in a blackish brown colour, the use of fly ash coagulant at a dose of 10 gr with a value of 65 gr/l can remove colour up to 94.49%. Kristina Wijaya (2008) using Oxone at a dose of 1.6 g/l with 0.4 g/l α-Mn_2O_2 at a temperature of 50°C, was able to reduce wastewater COD from 1059 mg/l to 344 mg/l.

Ardhi Ristiawan (2015) conducted research to degrade lignin from pulp and paper industry waste using the advanced oxidation process (AOP) method with a combination of ozone and hydrogen peroxide. Increasing the ozone feeding rate and adding H2O2 can increase COD and lignin removal. However, high concentrations of H2O2 can reduce the removal efficiency due to H2O2 excess can be a scavenger (inhibitor) in the process. Optimum results were achieved at an ozone feeding rate of 26.7 ppm/sec and an H2O2 dose of 1000 mg/l for 2 hours at the original pH of the wastewater sample (7-8). Under optimum conditions, COD removal was 28% and lignin removal was 88%. The kinetics of the COD removal reaction follows zero order with a reaction rate constant of 2.5 mg/l min^{-1} and lignin is included in the first order reaction with a reaction rate constant value of 0.0196 min^{-1}

In general, pulp and paper industry wastewater contain extractive products, carbohydrates, and lignin. Extractive products are compounds dissolved in organic solvents such as resins, fatty acids, triglycerides, waxes, sterols, and phenols. Lignin and chlorinated organic compounds are the main compounds that are of concern for potential environmental pollution (Ugurlu and Karaoglu, 2009). The bleaching process is a production process that uses chlorine-based chemicals as a bleaching process which can produce chlorinated organic compounds (Pokhrel and Viraraghavan, 2004 in Oller et al., 2010). Pulp and paper industry wastewater also has the potential to contain toxic Organic compounds that can pollute the environment. Chlorine compounds used in the bleaching process can produce organic chlorinated (Kumar et al., 2011).

The presence of colored compounds on surface waters is aesthetically undesirable and causes disruption of the aquatic biosphere due to reduced penetration of sunlight and depletion of dissolved oxygen. Some colored compounds are toxic and mutagenic and have the potential to release carcinogenic amines. Because they are difficult to degrade, colored compounds can also contribute to the failure of biological processes in wastewater treatment plants. In some applications, colored treated water is also not suitable for water reuse. In addition, color generally increases throughout the biological processing system, which may be due to the organic material being converted into smaller chromophoric units rather than mineralized. This review will focus on lignin as the main dye and its treatment using coagulation/flocculation. Before that, the structure and classes of lignin will be discussed.

2.1. Sludge Characteristic

The sludge from water treatment plant is being dumped into water bodies after going through the processing process at the Wastewater Treatment Plant (WWTP). The large water treatment capacity of up to tens of thousands of cubic meters per day with an average discharge of 6-9% of the volume creates its own problems in managing this residual sludge. Residues from the water treatment process can be reused by utilizing the remaining coagulant remaining in the settled sludge. There are two uses for this clean water processing residue (Mahes R Gadekar, 2017), firstly it is used as a coagulant in wet form where some of the sludge is returned to the beginning of the water treatment process, secondly in dry form it is used as a colour absorbent in the processing of paper waste containing paper colouring agent (SS Mogaddam, 2011).

The characteristics and discharge volume of sludge produced from clean water processing activities vary, predominantly influenced by the source of the water raw material. The sludge produced by the clean water processing company Pulp and Paper which is domiciled around the Pengabuan River, Tungkal Ulu-Tebing Tinggi sub-district has characteristics as shown in table 2.2 below.:

Table 2.2 Characteristic of WTP sludge in Pulp Industry.

No	Paramater	unit	Value
1	pH (1%)	-	6,2
2	Conductivity	µs/cm	193,3
3	Organic	%	7,8
4	Anorganic	%	92.2
5	Metal Content in sludger		
	MgO	%	3,5
	Fe2O3	%	13,8
	Al2O3	%	44,5
	Na2O	%	0,7
	K2O	%	3,5
	CaO	%	0,9
	SiO2	%	2,0
	Others (Mn,Co, Zn, Ba, As,Sr, Cu, Pb, Cr dll)	%	<0,5

Source: Laboratories Research and Development, Preliminary analysis of clean water treatment sludge content, Ref. QC/TM/1-44, QC/TM/4-01, Tools: ICP

2.2. Lignin

Lignin is an abundant natural raw material and the most abundant natural aromatic polymer compound. Lignin is a natural polymeric product from enzymes that undergo dehydrogenative polymerization of 3 initial polymers. The following structure is the structure of lignin. In general, lignin has an unfavourable effect when it gets into water. (Yaser A.Z., 2014).

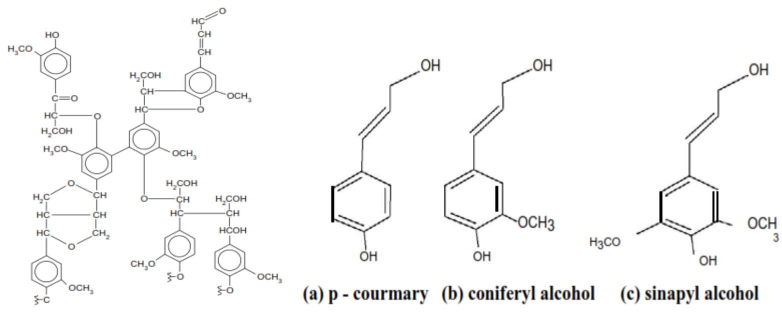


Figure 2.2 Lignin structure and the three basic constituents of lignin

2.3. Flocculants and Coagulant

Coagulation and flocculation are one of the important process stages in wastewater processing units and clean water processing. Coagulation is the destabilization of particles using coagulant materials, coagulants are grouped into 3 categories according to their constituent materials.

a. Coagulant with inorganic base
b. Coagulants with organic base ingredients

c. Coagulant with hybrid base ingredients

Coagulants function to overcome factors that support particle stability and form clots or flocs. Flocculation is a process of destabilized particles or particles formed from the destabilization process being forced to stick together to form larger clumps. In general, coagulants used for liquid waste containing lignin use coagulants with aluminium and iron bases, both coagulants were proven effective in separating organic materials.

The floc produced from the flocculation process is removed through a gravity sedimentation process. The main purpose of sedimentation is to produce filtrate that has undergone a clarification process, but it is also necessary to produce sludge with a solid concentration that can be handled and processed easily. The solid phase of the flocculated sludge must settle freely under still or laminar flow conditions when introduced into the settling zone (S.S. Wong, et al, 2006).

The basic objectives of wastewater processing and purification technology can be carried out using one method or combining physical, chemical, and biological methods (Metcalf and Eddy, 2003). Adsorption is the use of solid media to remove substances from gas or liquid solutions, this has been used extensively widespread. This process involves nothing more than the separation of a substance from a liquid or gas phase onto the surface of a solid medium. Where the absorbing medium is called an adsorbent, and the absorbed substance is called an adsorbate.

2.4. Framework

Based on a literature review and theoretical basis, the level of colour improvement in wastewater is influenced by, firstly, the presence of materials used to absorb colour from wastewater (Mahesh R Gadekar, 2017) and secondly, how the absorbent material and colour are separated to produce colourless water. Particles The lignin contained in liquid waste from the pulp industry is in colloidal form, the coagulation process is not effective because of the low level of suspended particles, so in this research the enrichment of suspended particles was carried out by adding sludge originating from the clean water processing process and adding coagulant/flocculant (Abu Zahrim Yaser, Current Review on the Coagulation Flocculation of Lignin Containing Wastewater 2014). It is hoped that the sludge particles will bind the lignin in colloidal form in the solution and settle together with the mud so that there will be a decrease in the level of colour, COD and TSS in liquid waste originating from pulp washing activities and other activities in the pulp and paper industry environment.

2.5. Experimental Design and Data Analysis

2.5.1. Effect of Treatment to wastewater Quality

The first this experiment was designed to see the effect of various treatment of wastewater with sludge, coagulant, and combination sludge-Coagulant. The research was designed using True Experimental Design with Pretest-Posttest Control Group Design by controlling all external variables that influence the course of the research Through this experiment effect of the treatment will be deeply studied to find out process optimization and evaluate the cost.

Percent removal of the response (COD, turbidity, and colour) from wastewater calculated as follows:

$$\% \, Removal = \left\{ \frac{Yo - Yi}{Yo} \right\} \quad \text{.......} \quad (1)$$

Where, Y_o and Y_i are the initial and final value of the respective parameters (Smita, 2015)

2.5.2. Process Optimization

The second phase of experiment is to looking optimum treatment, experiment designed with Response Surface Methodology Box Behnken (RSM Box Behnken), Range and level of design expressed as below.

Table 3.1 Range experiment and level of independent variable

Design Variable	Factor	Range and level		
		-1	0	1
X1	Sludge, gr	50	100	150
X2	PAC, ml	0,5	2,75	5
X3	time, minute	30	60	90

The range and level of variables in the code units from the RSM study are given in table 3.1. For statistical calculations, variable X is given the code xi, according to the following equation:

$$\boldsymbol{xi = \frac{Xi - Xo}{\Delta X}} \quad \text{..........} (2)$$

Where Xi is the undetermined value of variable i, Xo is the value of X at the midpoint of the observed range and ΔX is distance of change in X value

Data analysis was carried out using the RSM Box Behnken (RSM) Design. RSM is used to develop and optimize processes and to evaluate the level of significance of several influencing factors in the presence of complex interactions. The response of each treatment to wastewater quality is independent of the factors of amount of sludge, PAC dose and residence time on the dependent factors of colour, turbidity, and COD. Quadratic regression is used to analyse the causal relationship between independent factors and dependent factors (Noushin, 2014)

$$Y = \beta o + \sum_{i=1}^{k} \beta i X i + \sum_{i=1}^{k} \beta ii X i^2 + \sum_{i=1}^{k-1} \sum_{j=2}^{k} \beta ij X i X j + \varepsilon \quad \text{..........} (3)$$

Where,

Y = Estimate response

X = Independent Variable

β = Regression coefficient

ε = Error rate

i = Linier coefficient

ii = Quadratic coefficient

ij = Interaction coefficient

k = Number of factors studied and optimized in research

2.6. Results and Discussion

2.6.1. Effect Coagulant or sludge to wastewater quality

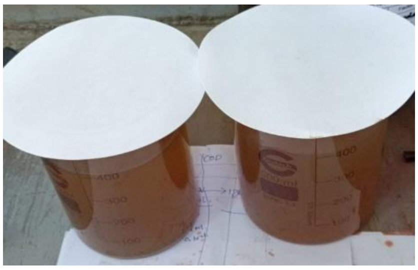


Figure 1 treatment with coagulant and sludge

There was no significant change of colour, COD, and turbidity.

Addition Coagulant without sludge does not give impact due to no floc formed (SS Wong, 2006) another additional sludge without coagulant no impact as lignin in wastewater in stable condition. Coagulant needed to destabilized lignin and form floc.

2.6.2. Effect combination Sludge and coagulant to wastewater quality

Combination of sludge 25% (v/v) and various dose of coagulant in this experiment we are using industrial grade PAC1130 where this product used in water treatment plant. Showing a quality improved on treated wastewater, the quality (Colour, COD) improved. Treatment no 1, 75% of wastewater mixed with 25% of WTP sludge and addition PAC 0.1 ml/ltr as shown in the picture turbidity high. Picture no 2 with composition 75% (v/v) plus 25% (v/v) WTP sludge and added 0.25 ml/ltr the visual of colour slightly improve compare than first treatment. In the last treatment combination 75% (v/v) wastewater with 25% (v/v) with 0.5 ml/ltr PAC dosage creating clear filtrate and solid settle easily. Measuring colour using spectrophotometer which is calibrated using standard solution Pt-Co at wavelength 450-465 nm,

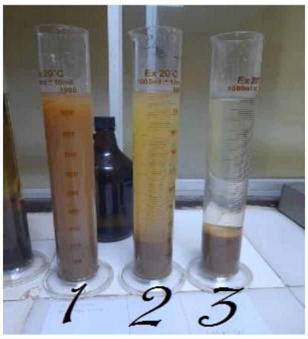


Figure 2. Combination wastewater and wtp sludge

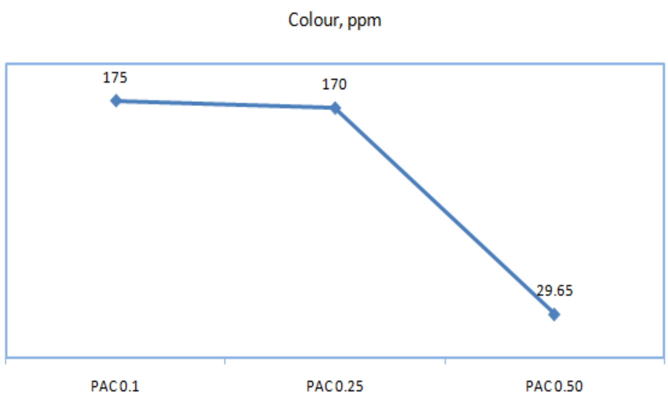


Figure 3 Colour vs PAC dosage

Another parameter checked (color) showing positive impact, initial color of wastewater 199 ppm removal color changes according to changes in PAC dosing. Increasing PAC dosage impacts increasing color removal from wastewater. The color removal calculated with formula (1) shown as below table. The data showed that PAC with Aluminum base coagulant able to overcome factors which can support particle stability and form floc. Flocculation is a process of destabilized particles or particles formed from the destabilization process being forced to stick together to form larger clumps. Coagulants used for liquid waste containing lignin use coagulants with aluminum and iron bases, both coagulants were proven effective in separating organic materials. The presence of mud particles binds the destabilized lignin particles and precipitates them.

No	PAC Dosage per liter	Colour ppm	Removal, %
1	-	199	-
2	0.1	175	12%
3	0.25	170	15%
4	0.5	29.65	85%

Table 1 Effect WTP sludge and PAC to color remocal

Along with the removal of lignin from liquid waste, this has a direct impact on reducing COD levels due to reduced lignin content.

2.7. Conclusion

Combination sludge and coagulant give impact to wastewater quality improvement. Increasing PAC dosing increase Colour removing, removal colour is inline with lignin removal it means COD removal also increases. Next step is to find out the optimum doses of sludge and PAC to get economical treatment cost.

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