Surface Water Treatment with Natural Starches as Coagulant via Coagulation Process

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Abstract. In this study, natural polymers in the forms of starch from sago and tapioca were used as coagulant. The removal of turbidity and chemical oxygen demand (COD) using coagulation process from river water was examined. The treatment on the river water sample was evaluated via a series of Jar Test experimental works with rapid mixing speed of 120 rpm for 1 minute, slow mixing speed of 30 rpm for 20 minutes and settling time of 60 minutes. The tapioca starch removed 93.7\% of turbidity at dosage of 1000 mg/L. While, sago starch successfully removed 96.4\% of turbidity under optimum dosage of 2000 mg/L. pH 12 was optimum pH for turbidity removal by using sago and tapioca starch whereas the highest percentage of COD removal occurred at pH 4 using sago and tapioca starch. At pH 4, tapioca and sago starch removed 22.5\% and 25\% of COD respectively. Overall tapioca starch showed better removal percentage of turbidity compared to sago starch throughout the experiment study. Generally, the usage of natural polymers as coagulants is good for turbidity removal; however, it did not work well on COD removal.

1. Introduction

Environmental issues related to water cleanliness are big and important issues that need to be considered by everyone not only in developing countries but all over the world. Good and healthy water brings about healthy and sustainable ecosystems, thus leading to better human wellbeing. One of the biggest water resources is surface water. Surface water is water on the surface that has in the Earth; which is including river, wetland, lake or ocean. This study will focus on the treatment of river water. Commonly, river will be the victim of indiscriminate dumping of untreated wastewater and chemical wastes by irresponsible person or management. It is very vital to make sure the cleanliness and quality of river water is secure as the river is one of the top resources for drinking water and also a popular place for recreation.

Coagulation is an important process in the treatment of surface water. This process involved the removal of dissolved chemical species and turbidity from the wastewater and water. Coagulation is carried out by addition of ions that has opposite charge with the colloidal particles charge. Coagulant which is usually positively charged provides positive electric charges to reduce the negative charge of the colloids. The particles collide against each other to form bigger particles.

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A coagulant which is water dissolved electrolytes can be divided into two main which are the inorganic coagulants and polyelectrolytes [1]. Common synthetic coagulants which is aluminium salts and inorganic coagulant such as alum and ferric chloride are the most used in water and wastewater treatment. However, this chemical-based coagulant comes with many drawbacks, such as Alzheimer’s disease and similar health related problems associated with residual aluminium treated water, the most serious case that it can cause harm to human health and environment. Aluminium and iron salts are normally used as chemical coagulant [2]. Studies conducted by American Water Works Association (AWWA), showed that there are some serious problems of using the aluminium salts as coagulant in water and wastewater treatment such as Alzheimer’s disease and similar health related problems associated with residual aluminium treated water. It is therefore, a natural coagulant was introduced as a substitute for commercial coagulant due to others advantages of natural coagulant over chemical coagulant.

In this study, natural starch which are sago and tapioca starch were used as coagulant to replace the chemical coagulant. Beside, jar test was used to study water treatment in the laboratory through coagulation and flocculation process.

2. Materials and methods
2.1. River water sampling
Surface water samples were collected from the Sungai Perlis, Kangar. The samples were transferred into 5.5 L plastic container and then close, sealed tight and label to avoid any oxidation and contamination before being transport to the laboratory. The samples were stored in a dark place to minimize the surface water sample from experiencing further biodegradation due to microbial activities. Characterization of samples is followed in accordance to the Standard Methods for the Examination of Water and Wastewater. The samples are characterized in terms of turbidity and COD. Before the jar test is conducted, the sample thoroughly agitated for re-suspension of settled solid.

2.2. Chemical and analytical methods
COD test is performing according to Method 5220D (Closed Reflux, Colorimetric Method) of the Standard Methods for the Examination of Water and Wastewater. Thus, COD was measured using HACH DR3900 Portable Spectrophotometer while turbidity test is performing using HACH 2100AN Laboratory Turbidity Meter with measurement in Nephelometric Turbidity Unit (NTU).

2.3. Experimental plan
The jar test is the usual method used to carry out the coagulation process. To determine optimum dose of coagulation process, 500 mL of river water samples were poured into each of the 6 beakers with fixed pH at 7 and place under jar test apparatus. The desired amount of coagulant (dose of 0 (as control), 1000, 2000, 3000, 4000 and 5000 mg/L) were added to the suspension and stirred at rapid mixing (120 rpm) for 1 minute. Then, followed by slow mixing (30 rpm) for 20 minutes to keep flocs particles uniformly suspended. At the end of the 20 minutes of slow mixing, the stirrer was turned off and flocs were allowed to settle for 1 hour. The experiment for other coagulant concentration (6000 mg/L) was repeated. After that, the supernatant was collected by using a dropper to be used in the determination of the turbidity and COD using the standard method and thus optimum dosage of the natural coagulants was recorded at 1000 mg/L for tapioca starch whereas for sago starch at 2000 mg/L [3]. Next, in order to determine optimum pH coagulation process, the procedure was repeated by varying the pH of the river water as 2, 4, 6, 8, 10 and 12 at optimum dosage which are at 1000 mg/L for tapioca starch whereas at 2000 mg/L for sago starch. The pH of river water samples were controlled by adding 1.0 M hydrochloric acid or 1.0 M sodium hydroxide.
3. Results and discussion

3.1. River Water Characterization

The river water samples collected from Sungai Perlis, Kangar were analyzed. The river water characteristics were as follows: (Turbidity = 44 – 183 NTU) and (COD = 300 – 1400 mg/L).

3.2. Effect of Dosage on Coagulation-Flocculation Process

The dosage used for both sago and tapioca starches ranged from 0 mg/L to 6000 mg/L with interval of 1000 mg/L. In order to determine the optimum coagulant dosage, other variables such as pH, mixing speed and settling time have to keep constant throughout the experiment. The pH of river water sample was fixed at 7 for both sago and tapioca starches.

The effect of sago starch at various doses was shown in Figure 1. A consistent increase for turbidity removal was observed and reached highest removal (optimum dosage) at dosage of 2000 mg/L with efficiency of 78.5%. Beyond this dosage, the turbidity removal decreased gradually with increasing dosage. Initially, the removal pattern for COD was similar to turbidity removal. The optimal dose of sago starch at 2000 mg/L managed to reduce the COD value as much as 31.4%. A decreasing trend was observed at dosage of 3000 mg/L followed by an increase when the dosage was increased to 6000 mg/L.

![Percentage of removal vs. Sago starch coagulant dosage](image)

**Figure 1.** Percentage of turbidity removal against sago starch coagulant dosage

Next, according to figure 2, the highest removal efficiency for tapioca starch (optimum dosage) was observed at dosage of 1000mg/L with efficiency of 88.7%. A decreasing trend was observed at dosage 2000 mg/L until 5000 mg/L followed by an increase when the dosage was increased to 6000 mg/L. At optimum dosage of tapioca starch which is 1000mg/L, the COD removals reached it maximum value at 53.5%. The COD removal efficiency was decreased when the dosage was increased from 2000 mg/L to 6000 mg/L.
The removal efficiency of parameter could be enhanced with further increase of dosage until an optimum value is obtained. The removal efficiency is decreased after the optimum dosage due to restabilization of colloids when the coagulant was used in excessive amount. Overdose the coagulant can cause a complete charge reversal and restabilize the colloid complex. Hence, the excessive starch itself can contribute to the turbidity and the turbidity will increase. From the result, sago and tapioca only required small amount of dosage as an optimum to remove the turbidity.

COD removal efficiency is decreased with the addition of more dose starches. The lower removal percentages of COD for both sago and tapioca starch were probably due to increased organic load from biological components of starches and subsequently contributed to increased COD concentration in the river water sample [3].

3.3. Effect of pH on Coagulation-Flocculation Process

From figure 3 and 4, the highest percentage of turbidity removal by using tapioca and sago starch as coagulant both was reached at pH 12 with the value of 93.7% and 96.4% respectively.

The decreasing trend was observed at range pH 3 to pH 8 for turbidity removal by using sago (figure 3.). Then at pH 10, it is become slightly increased and reached its highest removal efficiency for turbidity at pH 12. This result is compatible with research by [4] that showed the trend of increasing turbidity removal at pH 9 and reached its highest turbidity removal at pH 12. Initially, the removal pattern for COD showed some up and down in its value, and then it continue to decreasing at pH 9.
According to figure 4, the removal efficiency of turbidity for tapioca is increased gradually starting at pH 8 to pH 12. However, the removal efficiency of COD declining in a dramatically order as the pH increasing from pH 6 to pH 12.

Turbidity removal occurred at two pH conditions which are at low pH and at high pH. Turbidity removal performance is not very good at other than these pH ranges. It can be observed from both figure 3 and 4 that more than 80% of turbidity at pH 2 and pH 12 could be removed from river water sample by using sago and tapioca starch as coagulant. The reduction of turbidity might be due to the some particle or inorganic/organic compound solubility (that available in the river water) which is observed to be highly influenced by pH. As the particle solubility increases, the turbidity of the river water decreases.

COD removal efficiency is higher at low pH value. The reduction of COD concentration might be due to the particle or inorganic/organic compound solubility in river water sample, as the particle solubility increased, the COD concentration decreased in the river water sample.
4. Conclusion
River water from Sungai Perlis was treated by using natural starch namely sago and tapioca starch as coagulant. Based on the optimum dosage condition, highest turbidity removal was 96.4% using sago starch as coagulant at 2000 mg/L. While, tapioca starch successfully removed 93.7% of turbidity under optimum dosage of 1000 mg/L. pH 12 was optimum pH for turbidity removal by using sago and tapioca starch whereas the highest percentage of COD removal occurred at pH 4 using sago and tapioca starch. It can be concluded that the usage of natural polymers as coagulants is good for turbidity removal. However, it did not work well on COD removal but quite the reverse, the sago starch is expected to be natural organic polymers anionic that able to remove heavy metal ion. The reasons for this occurrence require further research.

Acknowledgements
The authors acknowledge the Faculty of Engineering Technology, Universiti Malaysia Perlis (UniMAP) for providing equipment and technical assistance for this project.

References
[1] Duan, J., & Gregory, J. (2003). Coagulation By Hydrolysing Metal Salts. *Advances in Colloid and Interface Science, 100–102*, 475-502.
[2] Shilpa, B., Akanksha, K., & Girish, P. (2012). Evaluation of cactus and hyacinth bean peels as natural coagulant. *International Journal of Chemical and Environmental Engineering, 3*.
[3] Idris, J., Mad Som, A., Musa, M., Ku Hamid, K., R.Husen, &Rodhi, M. (2013). Dragon Fruit Foliage Plant Based Coagulant For Treatment Of Concentrated Latex Effluent: Comparison Of Treatment With Ferric Sulfate. *Journal of Chemistry*.
[4] Khalid, n. f. (2005). Zinc,Iron And Turbidity Removal From Semi-Aerobic Leachate Using Coagulant Process.