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Potential of Fruit Peels in Becoming Natural Coagulant for Water Treatment

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Abstract: The effectiveness of chemicals as coagulants such as alum and ferric chloride is well recognized. However, there are many disadvantages associated with the usage such as high operational costs, detrimental effects on human health, production of large sludge volumes and the fact that it is significantly affect pH of treated water. It is therefore desirable to replace these chemical coagulants with natural-based coagulants such as from fruit peels to counteract the aforementioned drawbacks. Therefore, the aim of this study is to investigate the efficiency of fruits peels as natural coagulants in treating water over the use of alum. In terms of selection of natural coagulants, this study focused on the local waste materials, which are banana peels, orange peels and mango peels. These peels were collected from neighborhood and local stall and market. These peels were prepared by washing, drying, grinding and finally sieving, thus becoming powder of natural coagulants ready to be used. A series of jar test was then performed to determine the effect of individual natural coagulants on the efficiency of turbidity removal and coagulation activity under various operating factors such as type and concentration of solvents, pH and coagulant dosage. From the findings, optimum solvent concentration of 2.0M of sodium hydroxide (NaOH) reacted with mango peels at optimum pH and coagulant dosage of pH 2 and 110 mg/L, respectively showed greatest turbidity removal of 92.7% compared to the other tested natural coagulants including chemical coagulant, which was alum. Therefore, it can be concluded that the natural coagulant in specific mango peels pose a great potential in replacing chemical coagulant for the treatment of water.

Keywords: Natural coagulant, fruit peels, yield, pH, coagulant dosage, turbidity removal

1. Introduction

Many impurities in water and wastewater are present as colloidal solids, which do not readily settle. Finely dispersed suspended and colloidal particles that produce turbidity and color of the water cannot be removed sufficiently by ordinary sedimentation. Colloidal particles generally carry a negative electrical charge. These particles surrounded by an electrical double layer (diffuse layer and Stern layer) thus, preventing contact between each other [17]. Adding a coagulant (generally positively charged) and mixing the water causes compression of the double layer and thus neutralization of the electrostatic surface potential of the particles. The resulting destabilized particles stick together upon contact forming solids known as 'flocs'.

Coagulation is an important stage in conventional water and wastewater treatment process [1]. Coagulation is a process of combining small particles into larger aggregates (flocs) and simultaneously adsorbing dissolved organic

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matter on to particulate aggregates so that these impurities can be removed in subsequent solid/liquid separation processes. Chemical coagulants such as alum (aluminium sulphate), ferric chloride and cationic polymers are the common coagulant agents that been used in the treatment process [2]-[4]. However, there are many disadvantages associated with the use of these coagulants such as high operational costs, detrimental effects on human health and the fact that it is significantly affect pH of treated water. The use of chemical coagulants also tends to produce more end sludge product and mostly, this sludge is toxic and hazard. Due to such matter, proper treatment of the sludge before disposal is important. This sludge will normally be disposed to Secured Landfill, which then require greater cost.

The alternative of chemical coagulant, which is natural coagulant, has been used in many countries as an effective coagulant. These coagulants were produced from various materials such as plant seeds, leaves and roots [5]. Among the known natural coagulants are *Moringa oleifera, Jatropha curcas, Strychnos potatorum, Hibiscus sabdariffa* and many more [6]. These natural coagulants are sustainable because there are no health hazards and the cost of producing these natural coagulants were less expensive than the chemical coagulants since it is locally available. The natural coagulants can treat water of high turbidity and having remarkable removal efficiency [7].

Natural coagulants in water and wastewater treatment process also give high numbers of economic advantages. When effluent conductivity is low in water recycling processes, the corrosion of the facilities can be avoided thus, the maintenance cost can be reduced. This can only be achieved by applying natural coagulant rather than normal chemical coagulant. The use of natural coagulant also provides higher coagulant efficiency that allows more active colloidal aggregation and more consistent floc compared to conventional coagulant. This occurrence helps to reduce the necessary quantity of coagulant and consequently, able to reduce the operational cost.

To date, the chemical coagulants are becoming less sustainable in treating water and wastewater due to various adverse effects. It is now desirable to replace these coagulants with more natural-based coagulants such as from fruit peels that can counteract the aforementioned drawbacks. Therefore, the aim of this study is to investigate the efficiency of fruits peels as natural coagulants in treating water over the use of alum.

2. Methodology

2.1 Materials

Water sample was collected from Water Treatment Plant, Taman Impian Emas, Johor Bahru. In terms of selection of natural coagulants, this study focused on the local waste materials specifically the fruit peels. The collected peels are banana peels, orange peels and mango peels. These peels were collected from neighborhood and local stall and market.

2.2 Analytical Methods

The collected peels that have been prepared were weighted for yield and for determination of moisture contents. Eq. (1) and Eq. (2) show the calculation to determine the percentage of yield and moisture content of the prepared fruit peels as natural coagulant.

$$\text{Percentage of Yield} = \frac{\text{Yield}}{\text{Initial Weight}} \times 100 \tag{1}$$

$$\text{Moisture Content} = \frac{\text{Initial Weight - Dry Weight}}{\text{Initial Weight}} \times 100 \tag{2}$$

The effect of these fruit peels, as natural coagulant was determined based on the performance of coagulation activity and turbidity removal. Coagulation activity was conducted based on Kukiĉ et al. [8] while water turbidity, expressed in nephelometric turbidity units (NTU) was determined using HACH 2100Q. Eq. (3) shows the calculation to compute the percentage of coagulation activity.

$$\text{Coagulation Activity } (\%) = \frac{T_b - T_s}{T_b} \times 100 \tag{3}$$

where; T_b is turbidity of the blank and T_s is sample turbidity.

2.3 Experimental Procedures

For the purpose of developing natural coagulant, three types of waste plant materials are chosen. There are banana peels (*Musa sapientum*), orange peels (Citrus reticulata) and mango peels (*Mangifera indic L.*). These peels were collected from neighborhood and local stall and market.

Fig. 1 illustrates the process of preparing the coagulants. Based on the figure, after collecting the materials, only good quality of fruit peels was selected. Fruit peels were washed thoroughly with deionized water to remove the adhering dirt. Selected peels are then kept at room temperature for 2 hrs before oven dried for 24 hrs at 103 to 110 °C. After drying, the collected fruit peels were weighed to obtain the dry weight, and the moisture content can then be computed. Then, the peels were grinded into powder form in order to be used in treating water. The weight of powder form for each of the developed natural coagulant was recorded as yield. The powder coagulants were sealed using plastic seal and stored in cold room to prevent aging effect.

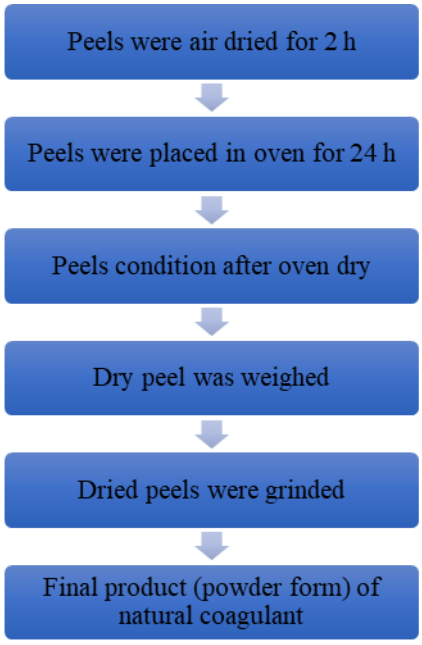

Fig. 1- Process of preparing natural coagulant

For these further experiments, Jar test was used in order to determine the best values of the operational factors, which are types and molarity of solvents, coagulant' dosage and pH, that can determine the success of water treatment. As to investigate the effect of solvent upon the efficiency of developed natural coagulants, two solvents were used, which are sodium hydroxide (NaOH) and sodium chloride (NaCl), while deionized water was used as control. The molarity of 0.1M, 0.2M, 0.5M, 1.0M and 2.0M were used for both solvents. 10 mg of each prepared natural coagulant powder was suspended in 200 mL of each solvent and deionized water. The suspension was stirred using a magnetic stirrer for 10 min at room temperature (20±1°C) to extract the active coagulant components. The optimization on the best solvent was determined based on the greater performance of coagulation activity and turbidity removal.

Upon the efficiency of developed natural coagulants in treating water, the effect of pH and coagulant dosage are also investigated. As to determine the effect of dosage, the pH of water sample is kept constant at pH 7.5. The natural coagulants' dosage is varied between 10 mg/L to 130 mg/L. Meanwhile, in order to determine the effect of pH in treating water, the optimized dosage of the coagulant is kept constant. The pH is varied between 2 to 12. Both experiments testing on the effect of pH and coagulants' dosage are analyzed towards responses of coagulation activity and turbidity removal.

The study is then continued for determining the efficiency of selected natural coagulants in comparison to the used of alum as a representative of chemical coagulant. In this study, the best coagulant among three types coagulants that been tested previously, which resulted in maximum percentage of coagulation activity and turbidity removal was selected. This coagulant was experimented using best solvents with optimized molar concentration, pH and coagulant dosage that obtained in previous experiments. The alum that been compared are also experimented with similar pH and dosage as natural coagulant. The efficiency between these two natural and chemical coagulants can then be determined based on maximum results of coagulation activity and turbidity removal of the treated water.

3. Results and Discussion

3.1 Physical Properties of Natural Coagulant

Table 1 shows the initial weight, yield and moisture content for each of the prepared natural coagulant. Based on Table 1, the highest initial weight of the collected waste plant materials are orange peels with 700 g, followed by banana peels and mango peels with about 374 g and 143 g, respectively. After dried and grinded, the final weight, which is the yield of the plant materials are about 152 g for orange peels, 140 g for banana peels and 36 g for mango

peels. This makes the overall percentage yield powders as compared to initial weight materials are 37.4 % followed by 25.2 % and 21.7 %, for banana peels, mango peels and orange peels, respectively.

Table 1 - Weight and moisture content for all coagulants

Coagulants	Initial Weight (g)	Dry Weight (g)	Yield (%)	Moisture Content (%)
Banana peels	374	140	37.4	62.6
Orange peels	700	152	21.7	78.3
Mango peels	143	36	25.2	74.8

Based on Table 1, it clearly seen that banana peels contributed highest yield compared to the orange peels and mango peels. This is because pretreatment of certain fruit material by microwave heating led to a considerable increase in the yield [9-10]. Apparently in this study, microwave pretreatment of fresh banana peels led to destructive changes in the plant tissue. These changes in the plant tissue after a microwave pretreatment gave an opportunity for the considerable increase in the yield of extractable pectin and improvement of its parameters like molecular mass and gel strength.

Table 1 also shows the trend of moisture content for each of the coagulant. From the table, moisture content in banana peels is the lesser among the other coagulants, thus also contributing to the greater yield as the final powder product. On the other hand, orange peels comprised the highest moisture content of 78.3%, hence resulted in lowest yield among all tested coagulants in this study. Orange peels contain high moisture contents due to its functional properties. According to Kammoun Bejar et al. [11], orange peels have high water holding capacity that allows the peels to retain water into the peels.

3.2 Effect of Solvents on the Efficiency of Natural Coagulant

Three types of solvents - sodium hydroxide (NaOH), sodium chloride (NaCl) and deionized water were tested in this experiment. Fig. 2 and Fig. 3 each shows the turbidity removal and coagulation activity of the tested natural coagulants when deionized water is used.

Based on Fig. 2, it shows that orange peels resulted in higher turbidity removal of 26.5% followed by mango peels and banana peels with removal of 24.5% and 22.7%, respectively. The overall turbidity removals using the respective natural coagulants are low, which is less than 30%. This because the solvent used is the deionized water. Deionized water has not enough capability to extract fruit peels as compared to the solvents such as NaOH and NaCl. The trend of turbidity removal is aligned with the results of coagulation activity as indicated in Fig. 4. Based on the figure, coagulation activity for orange peels, mango peels and banana peels were 13.5%, 11.2% and 9.0% respectively. Orange peels, mango peels and banana peels were able to remove the turbidity from 40.57 NTU to only 29.8 NTU, 30.6 NTU and 31.4 NTU, respectively.

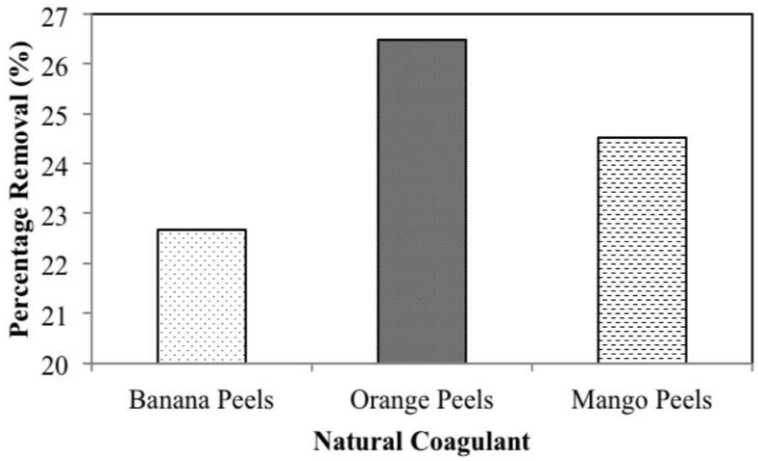

Fig. 2 - Turbidity removal of the natural coagulant using deionized water

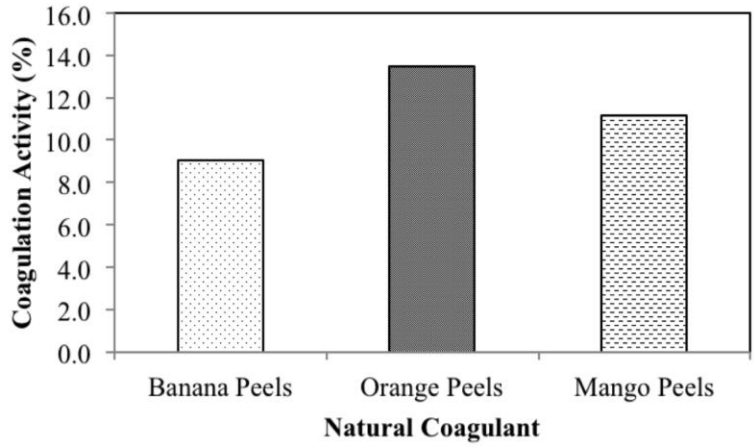

Fig. 3 - Coagulation activity of the natural coagulant using deionized water

Instead of deionized water, the experiment also tested on two different types of solvents named NaCl and NaOH on the natural coagulants. Different concentrations of NaCl and NaOH were used in this experiment. The concentrations used were 0.1M, 0.2M, 0.5M, 1.0M and 2.0M to react with natural coagulant. Fig. 4 and Fig. 5 show the turbidity removal and coagulation activity of natural coagulant under various concentration of NaCl, respectively.

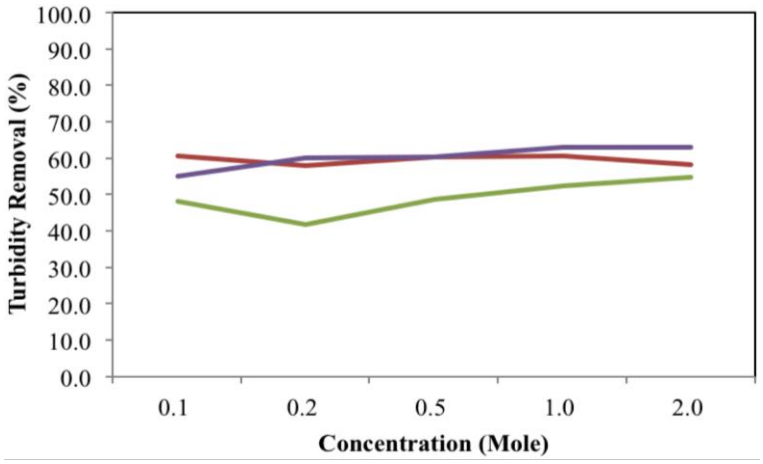

Fig. 4 - Removal of turbidity when natural coagulant reacted with NaCl (Banana peels; Orange peels; Mango peels)

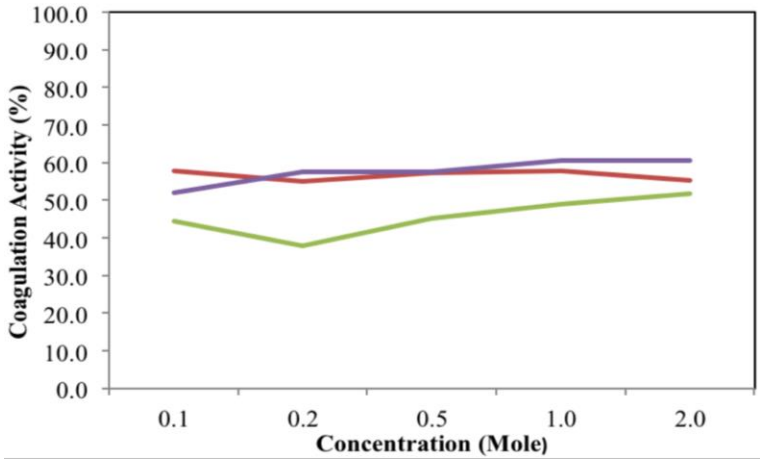

Fig. 5 - Coagulation activity when natural coagulant reacted with NaCl (Banana peels; Orange peels; Mango peels)

Based on Fig. 4, the optimum molar concentration of NaCl for all of the natural coagulants was recorded at 1.0M. At this optimum molar concentration, mango peels showed the highest turbidity removal of 63.2%, followed by banana peels and orange peels with removal of 60.6% and 52.1%, respectively. This observation is aligned with the coagulation activity (as shown in Fig. 5) at which mango peels showed greater coagulation activity of 60.8%, followed by banana peels and orange peels with coagulation activity of 58.1% and 49.0%, respectively.

In comparison to the other available solvents, NaCl seemed does not sufficiently able to extract the coagulant agent of the used natural coagulants therefore, resulting in the least impact of turbidity removal. It can be justified that NaCl is not a suitable solvent to be used in combination with this kind of natural coagulant to treat polluted water. Due to that justification, another solvent, which is NaOH was used. Fig. 6 and Fig. 7 show the trend of turbidity removal and coagulation activity for all the natural coagulants using NaOH as extraction solvent.

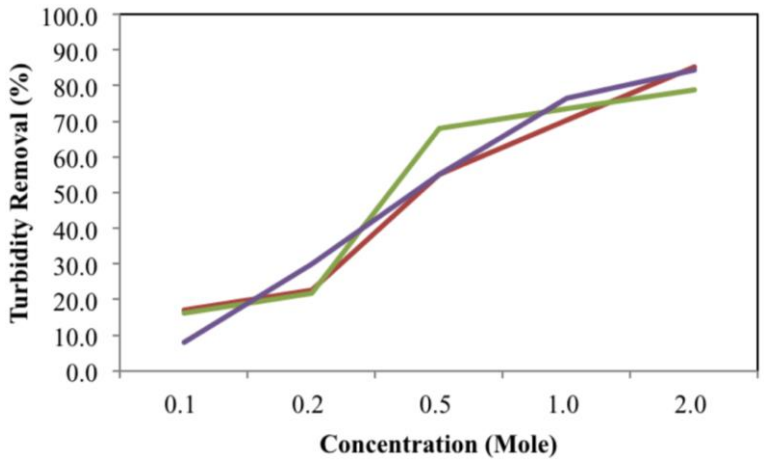

Fig. 6 - Removal of turbidity when natural coagulant reacted with NaOH (Banana peels; Orange peels; Mango peels)

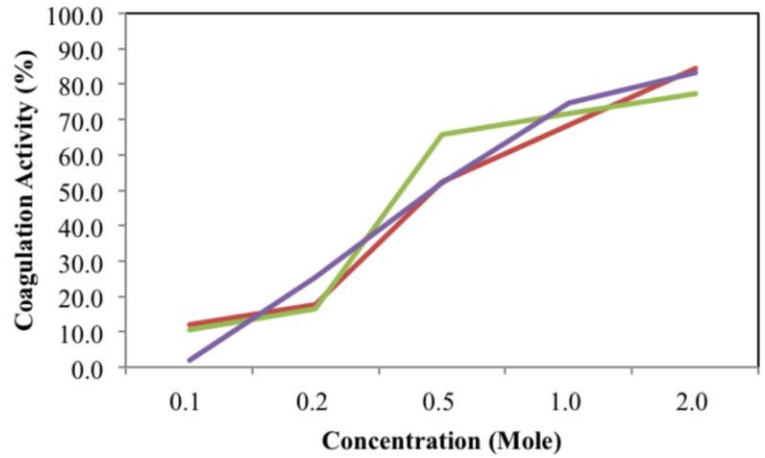

Fig. 7 - Coagulation activity when natural coagulant reacted with NaOH (Banana peels; Orange peels; Mango peels)

Based on Fig. 6, the optimum molar concentration of NaOH for all the natural coagulants was recorded at 2.0M. At this optimum molar concentration, banana peels showed the highest mean turbidity removal of 85.4%, followed by mango peels and orange peels with mean removal of 84.4% and 78.9%, respectively. This observation is aligned with the coagulation activity (as shown in Fig. 7) whereby the banana peels showed greater coagulation activity of 84.5%, followed by mango peels and orange peels with coagulation activity of 83.4% and 77.5% respectively.

In comparison to the other solvent, NaOH was sufficiently able to extract the coagulant agent of the used natural coagulants therefore, resulting in high impact of turbidity removal. However, this finding is contradicted with the findings reported by Muthuraman and Sasikala [12]. In that reported study, NaCl of 0.5M was the best solvent compared to NaOH in extracting natural coagulant of *Moringa oleifera* seeds, which then contributed to the greatest turbidity removal of about 90%. This contrast could be due to the different type of natural coagulant. In this study, fruit peels were used while in the previous reported study, seeds were used.

Although mean turbidity removal and coagulation activity was resulted by banana peels, the consistent increment in the removal and coagulation activity at further increment of NaOH concentration was indicated by mango peels. Turbidity removal was found to increase as the concentration increased up to 2.0M. This means that as the concentration of NaOH increased, more coagulant agent can be extracted from these peels thus, dissolved in the extracting solvent. This phenomenon is known as the salting-in effect [13]. When the concentration increased the solubility of the coagulant agent, then, more coagulation activity can occur, thus leading to a higher percentage removal of turbidity

3.3 Effect of Dosages on the Efficiency of Natural Coagulant

Coagulation dosage does affect efficiency of water treatment. It is best to obtain optimum coagulant dosage that is sufficiently able to well treat the water and at the same time does not adversely affect the quality of the water. In this study, the coagulant dosages were varied at 10 mg/L, 30 mg/L, 50 mg/L, 70 mg/L, 90 mg/L, 110 mg/L and 130 mg/L. These variations were conducted at constant pH of 7.5. Fig. 8 and Fig. 9 show the turbidity removal and coagulation activity for all the natural coagulants, respectively under various dosages.

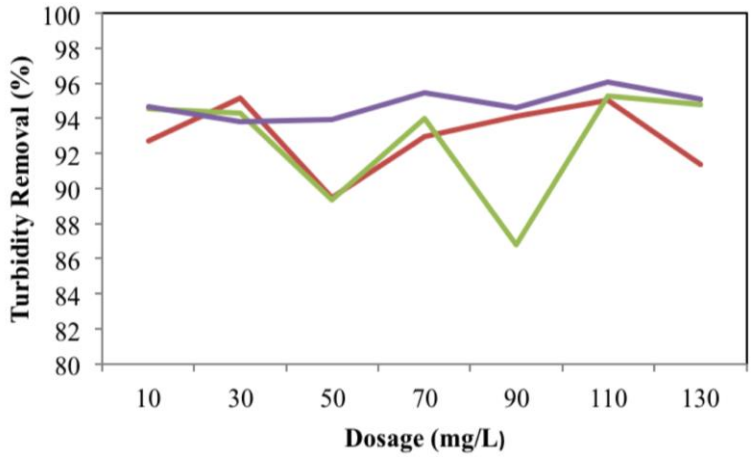

Fig. 8 - Effect of various coagulant dosages on turbidity removal at constant pH of 7.5 (Banana peels; Orange peels; Mango peels)

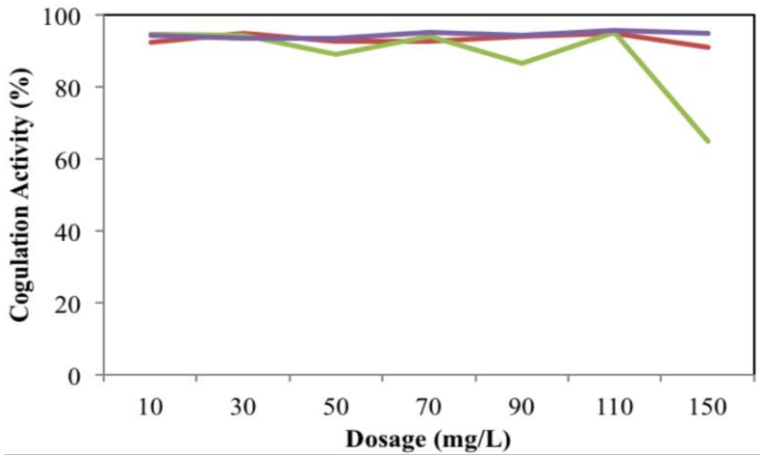

Fig. 9 - Effect of various coagulant dosages on coagulation activity at constant pH of 7.5 (Banana peels; Orange peels; Mango peels)

Based on Fig. 8, all three natural coagulants showed fluctuation in removing the turbidity. Among the three coagulants, mango peels were quite consistent with less fluctuation compared to banana peels and orange peels. Based on the figure, coagulant dosage of 110 mg/L contributed to the highest turbidity removal for all of the natural coagulants, thus this dosage is taken as the best coagulant dosage to be used in later work. At 110 mg/L, mango peels indicated highest turbidity removal of 96.1%. This followed by orange peels and banana peels which each of it obtained turbidity removal of about 95.3% and 95.0%, respectively. The results of turbidity removal are aligned with coagulation activity of these natural coagulants as shown in Fig. 9. Mango peels resulted in higher turbidity removal

compared to the other two natural coagulants could be because of high adsorption potential that has high content of cellulose, pectin (galacturonic acid), hemicelluloses and lignin [14].

After 110 mg/L, all three natural coagulants caused the removal to decrease. This is because all attractive charges of the particles have been attracted to each other prior settling. If high dosage is used, the excess coagulants will simply get added thus, cause turbidity in water. In particular of banana peels, the analysis showed that at lower dosage of 30 mg/L, these peels had resulted in greater turbidity removal of about 95.0% compared to orange peels and mango peels with removal of about 94.0% and 93.0%, respectively. This is because banana peels have high tendency of adsorbent occurred mostly at lower dosage concentration [15]. This could be the reason of such low dosage of banana peels able to result in high turbidity removal among all other tested dosages

3.4 Effect of pH on the Efficiency of Natural Coagulant

Besides coagulant dosage, pH is also among the operational parameter that contributes to the efficiency of natural coagulant in treating water. Fig. 10 and Fig. 11 show the turbidity removal and coagulation activity, respectively for all of the natural coagulants tested under various pH. Based on the figures, pH 2 contributed to the highest turbidity removal for all the natural coagulants, thus this pH is taken as the optimum pH.

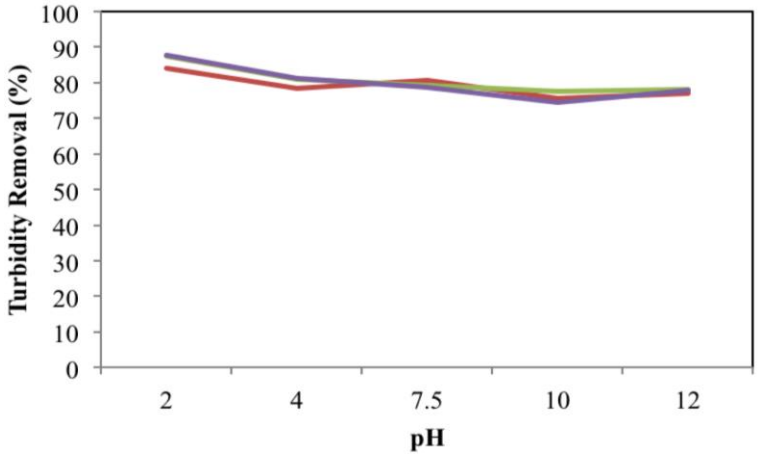

Fig. 10 - Effect of various pH on turbidity removal at constant dosage of 110 mg/L (Banana peels; Orange peels; Mango peels)

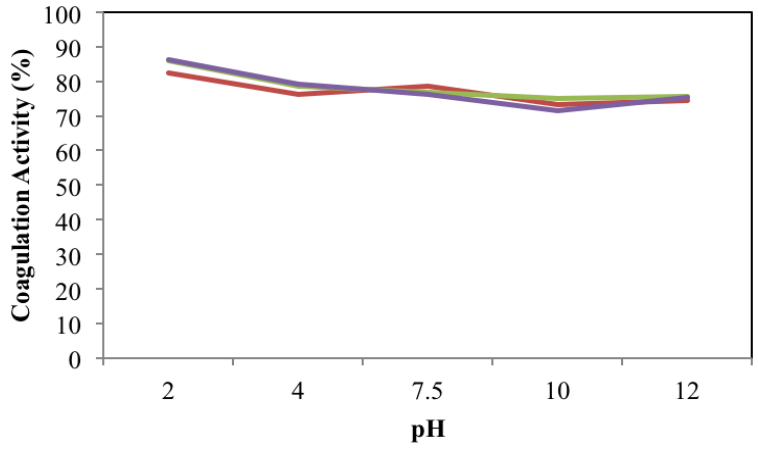

Fig. 11 - Effect of various pH on coagulation activity at constant dosage of 110 mg/L (Banana peels; Orange peels; Mango peels)

Based on Fig. 10, all three natural coagulants exhibited quite similar trends of removal and coagulation activity. At pH 2, mango peels indicated highest mean turbidity removal of 87.9%, followed by orange peels and banana peels which each of it obtained turbidity removal of 87.5% and 84.2%, respectively. The results of turbidity removal are aligned with coagulation activity of these natural coagulants as shown in Fig. 11. All peels resulted in higher turbidity removal at pH 2 could be due to acidic condition of the solution. This acidic solution may aggravate the coagulating agent comprised in the natural coagulant, thus resulting in greater coagulation activity and turbidity removal. At low

pH, more amine groups of natural coagulants were produced. It seems that decreasing the initial pH of the solution had a positive impact on the amount of natural coagulant to be added to the solution for efficient turbidity removal [16].

Further after pH 2, all three natural coagulants caused the removal to decrease. After pH 10, the turbidity removal starts to increase but not as much as in pH 2. This is because at pH 2, natural coagulant was completely active in coagulating the particles therefore ensure efficient flocculation process that resulting in better removal of turbidity.

3.5 Comparison between Natural and Chemical Coagulant

In this study, the efficiency of natural coagulant is investigated by comparing the turbidity removal and coagulation activity of the best-selected natural coagulant with chemical coagulant (alum). Table 2 summarizes the optimization of the solvents, coagulant dosage and pH that are used in this particular investigation.

Table 2 - Summary of optimization of the solvents, coagulant dosage and pH

Responses	NaOH (2.0M)	Dosage (110 mg/L)	pH (2)
Turbidity removal (%)			
Banana peels	85.4	95.0	84.2
Orange peels	78.9	95.3	87.5
Mango peels	84.4	96.1	87.9
Coagulation activity (%)			
Banana peels	84.5	91.4	85.1
Orange peels	77.5	92.1	85.2
Mango peels	83.4	93.5	87.0

Based on Table 2, mango peels are chosen as the best coagulant that exhibited greater turbidity removal and coagulation activity compared to banana peels and orange peels. Fig. 12 and Fig. 13 show the turbidity removal and coagulation activity of mango peels versus alum. Both coagulants, mango peels and alum was tested under specified conditions, which are at dosage 110 mg/L and at pH 2. Under these conditions, the mango peels outperformed alum in term of turbidity removal and coagulation activity

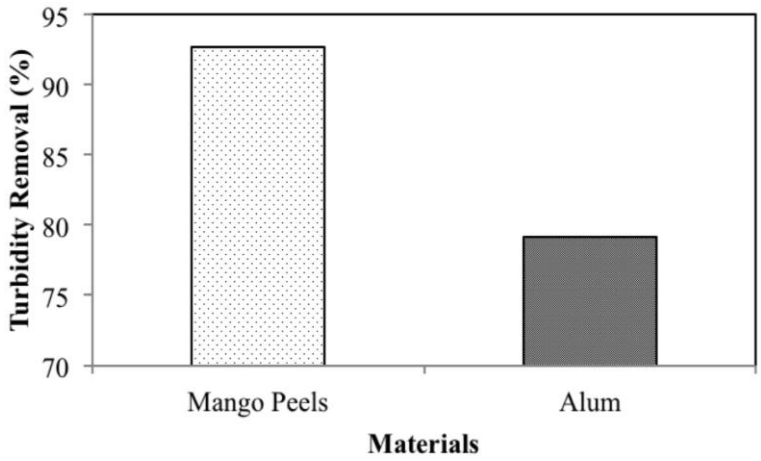

Fig. 12 - Comparison performance of turbidity removal between mango peels and alum

Based on Fig. 12, mango peels resulted in higher turbidity removal of 92.7%, compared to alum of only 79.2%. This observation is aligned with the coagulation activity (as shown in Fig. 13) at which coagulation activity of mango peels was 73.7%, better than alum with coagulation activity of only 25.0%. According to Suarez *et al.* [18], better results indicated by natural coagulant (mango peels) is due to the ability of a combination natural coagulant protein to decrease the viability of gram-negative or gram-positive bacterial cells and to mediate the aggregation of negatively charged particles in suspension, such as bacterial cells, clay, or silicate microspheres. It can then be justified that mango peels, which are a representative of natural coagulant can be used to treat turbid water more effectively than alum as chemical coagulant.

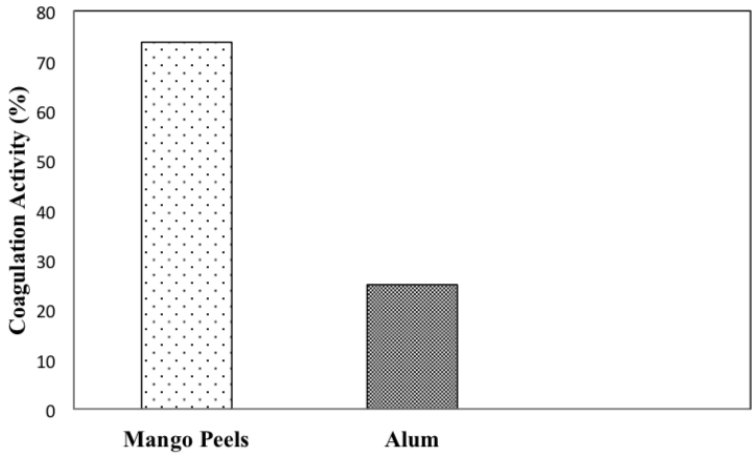

Fig. 13 - Comparison of coagulation activity between mango peels and alum

4. Summary

Plant-based natural coagulants have earned growing interests over the years due to their biodegradability and environmentally friendly nature. The variety of plant-based coagulants includes banana peels, orange peels and mango peels as discussed in this study have provided promising coagulation activity and turbidity removal in treating river water. The following below are the summary that been derived from the result of this study.

- Banana peels contributed the highest yield compared to the orange peels and mango peels. The observation is aligned with the trend of moisture content at which banana peels contributed to the least moisture content of about 63% compared to the orange peels of about 78%.
- NaOH is the most effective solvent in extracting the coagulant agent of the natural coagulant. The optimum molar concentration of NaOH was at 2.0M. At this optimum molar concentration, banana peels showed highest mean turbidity removal of 85.4%, followed by mango peels and orange peels with mean removal of 84.4% and 78.9%, respectively.
- The optimum coagulant dosage was 110 mg/L with pH 2. At this optimum condition, mango peels showed greater turbidity removal and coagulation activity of 92.7% and 73.7%, respectively.
- Under optimum conditions, mango peels outperformed alum at which mango peels resulted in higher turbidity removal and coagulation activity of 92.7% and 73.7%, respectively compared to alum of only 79.2% and 25.0%, respectively. These findings concluded that mango peels can treat turbid water more effectively compared to alum.

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