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The Combination of Papaya Seed and Orange Peel as Natural Coagulant for Greywater Treatment

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Abstract. The study utilized a natural coagulant composed of a combination of papaya seed and orange peel to treat greywater. The surface of papaya seed powder was rough and porous, while orange peel powder had an irregular, aggregated structure with non-uniform pores. The papaya seed coagulant was most effective in removing turbidity (86.3%), total suspended solids (50.3%), and chemical oxygen demand (61.4%) at a pH of 7.5, a dosage of 50 mg/L, rapid mixing at 170 RPM for 25 minutes, and slow mixing at 60 RPM for 20 minutes. The orange peel coagulant was most effective in removing 80.5% of turbidity, 64.6% of TSS, and 54.6% of COD at a pH of 7.5, a dosage of 10 mg/L, rapid mixing at 100 RPM for 5 minutes, and slow mixing at 40 RPM for 5 minutes. The ideal conditions for removing turbidity, TSS, and COD with the papaya seed and orange peel coagulant mixture were a pH of 7.5, a dosage ratio of 70:30, rapid mixing at 170 RPM for 25 minutes, and slow mixing at 60 RPM for 20 minutes.

INTRODUCTION

Water is an indispensable element for the survival and growth of living organisms. However, the rise in urbanization and industrialization has caused an increase in the amount of wastewater generated [1]. Domestic wastewater can be broadly classified into two categories based on their sources: blackwater and greywater. The fundamental difference between the two lies in the organic loading of blackwater being higher than that of greywater [2]. Blackwater comprises a combination of feces, urine, flush water, anal cleansing water, or dry cleansing materials [3]. Greywater, on the other hand, refers to any non-sewage domestic wastewater that comes from sources such as sinks, washing machines, dishwashers, showers, and baths, but excludes wastewater that has come in contact with human waste [4]. Greywater may contain traces of oils from cooking, soap, and some food items. Although greywater usually does not contain high concentrations of pathogens like blackwater, it may harbor some toxic contaminants that can cause illness if consumed [5-10].

The orange peel powder and papaya seed powder both had uneven, clumped structures and varying pore sizes. Using 50 mg/L of papaya seed coagulant, rapid mixing at 170 RPM for 25 minutes, and slow mixing at 60 RPM for 20 minutes at a pH of 7.5 resulted in the elimination of 86.3% of turbidity, 50.3% of total suspended solids (TSS), and 61.4% of chemical oxygen demand (COD). On the other hand, at a pH of 7.5, a dosage of 10 mg/L, rapid mixing at 100 RPM for 5 minutes, and slow mixing at 40 RPM for 5 minutes, orange peel coagulant decreased turbidity by 80.5%, total soluble solids by 64.6%, and chemical oxygen demand by 54.6%. Using a mixture of papaya seed and orange peel coagulant at a pH of 7.5, a dosage ratio of 70:30, rapid mixing at 170 RPM for 25 minutes, and slow mixing at 60 RPM for 20 minutes, resulted in 85.8% of turbidity eliminated, 58.0% of total suspended solids (TSS)

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eliminated, and 60.1% of chemical oxygen demand (COD) eliminated. The addition of 0.25 g of a combination of lemon and banana peels to wastewater reduced biochemical oxygen demand values by 89 to 96%, and eliminated 95.89% of the turbidity [11-14]. Apart from that, the papaya seeds coagulant showed the highest turbidity removal of 95.5% in treating wastewater from Sultan Ismail Water Treatment Plant at optimum condition of 130 mg/l dosage and pH 7.5 [13]

The coagulation process can be influenced by various factors, such as the coagulant dosage, mixing speed, wastewater pH, and coagulant type [15-16]. For water turbidity elimination, papaya seeds are recommended at a dose of 100 seeds per 100 liters, whereas the ideal dosage for orange peel is 1 mg/L. According to Kristianto et al. [17], orange peel coagulant was able to remove 97% of turbidity in dairy effluent when added at a concentration of 0.2 g/L and a pH of 7.5. In addition, papaya seeds were able to remove 84.7% of drimarene dark red dye from textile effluent at a coagulant dosage of 0.57 g/L and a pH of 1.07. Therefore, the aim of this study is to assess the effectiveness of orange peels and papaya seeds as coagulants for greywater treatment.

MATERIALS AND METHODS

Greywater Sample Collection

In this study, greywater sample was taken at a cafeteria located in Universiti Tun Hussein Onn Pagoh, Malaysia. The greywater samples were collected by grab sampling using bottles from a drain at the cafe that flows dishwashing and cooking waste from the sinks as shown in Figure 1. Then, the bottles were store in a chiller at the laboratory prior to be use for experiments.

FIGURE 1. Location of greywater sample collected from a café drain

Preparation of Orange Peel and Papaya Seed Coagulant

Firstly, the collected orange peel and papaya seed were wash thoroughly with distilled water to remove any dirt. Next, the peels and seeds were sun dried for three hours before it was cut into little pieces using knife. Then, the peel and seed were dried in an oven at 110˚C for 24 hours as shown in Fig. 2 [18]. After that, the dried peels and seeds were grinded into powder by using planetary ball mill and sieved using 600 µm mesh. The obtained orange peels and papaya seeds powder were kept in a separate airtight container as shown in Fig. 3.

FIGURE 2. Oven dried (a) orange peel and (b) papaya seed

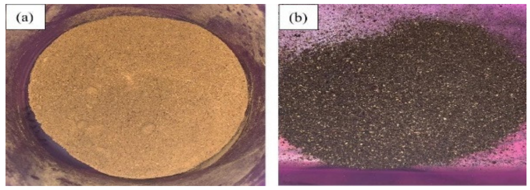

FIGURE 3. (a) Orange peel powder and (b) papaya seed powder

Coagulation Test

In the experiment, orange peel and papaya seed were evaluated as natural coagulants for cleaning greywater samples. The greywater sample was initially tested for its acidity, carbon dioxide (COD), dissolved oxygen (DO), and total suspended solids (TSS). The original 1 litre of greywater was divided among six clean beakers to assess its turbidity. Of the six beakers, five were treated with orange peel coagulant at concentrations ranging from 10 to 50 mg/L to examine the impact of dosage, while the sixth served as the control. The greywater samples treated with the optimal concentration of orange peel coagulant were also tested with pH levels ranging from 3 to 12. Table 1 lists the ideal dosage, pH settings, and mixing rates employed in the experiment. We mixed the samples at 100 rpm for the first five minutes and then decreased the mixing speed to 25 rpm for the remaining 20 minutes to assist flocculation. The flocs were allowed to settle for 30 minutes before 50 ml of the cleared water was piped out. The turbidity of the treated water was monitored throughout the jar test and recorded both at the beginning and end. The reduction percentage in turbidity was calculated using Eq. 1.

$$\textit{Turbidity removal (\%)} = [(T_0 - T)/ T_0]100 \qquad (1)$$

Where T_0 and T represent the initial and final turbidity (NTU) of water, respectively.

The steps were repeated by using papaya seed coagulant and combination of both orange peel and papaya seed coagulants at different mixing speed, pH and coagulant dosage as shown in Table 1.

TABLE 1. Parameter tested for orange peel, papaya seed and combination of orange peel and papaya seed coagulant in jar test experiment.

Jar no.	1 (control)	2	3	4	5	6
Coagulant dose (mg/L)	0	10	20	30	40	50
Papaya seed : orange peel ratio	-	10:90	30:70	50:50	70:30	90:10
pH	-	3	5	7	10	12
Effect of mixing speed						
1	Rapid mixing speed:100 rpm for 5 minutes Slow mixing speed: 25 rpm for 25 minutes					
2	Rapid mixing speed: 130 rpm for 5 minutes Slow mixing speed: 40 rpm for 5 minutes					
3	Rapid mixing speed: 170 rpm for 25 minutes Slow mixing speed: 60 rpm for 20 minutes					

RESULTS AND DISCUSSION

Surface Morphology

The surface morphology of papaya seed powder and orange peel powder is shown in Figure 3 and Figure 4, respectively. Papaya seed powder was agglomerated with non-uniform pore morphology and particle size [19]. In general, rough surfaces of orange peel powder provides high porosity and surface areas that are favourable for molecular diffusion and the adsorption of contaminants [20]. Moreover, the coagulant's larger surface area allows for greater adhesion for interparticle bridging or charge neutralisation.

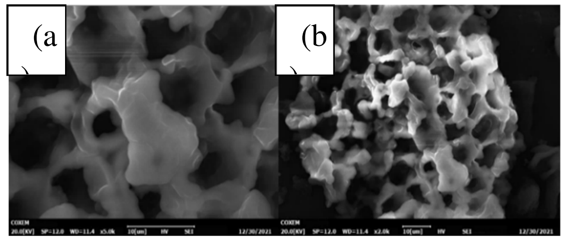

FIGURE 4. SEM morphology of papaya seed at (a) 5000x and (b) 2000x magnification

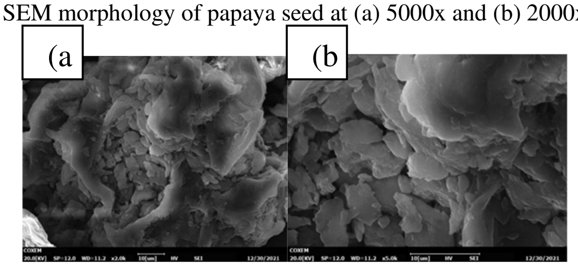

FIGURE 5. SEM morphology of orange peel powder at (a) 5000x and (b) 2000x magnification

Fourier Transform Infrared Spectroscopy (FTIR)

Figure 6 displays the FTIR spectra of papaya seed and orange peel powder. Papaya seed powder's -OH group was identified at 3275.69 cm^{-1}, while the aliphatic C-H stretch absorption was found at 2922.43 cm^{-1} and 2853.16 cm^{-1}. The FTIR spectrum also revealed the presence of functional groups such as C=O (Carboxyl) at 1743.82 cm^{-1} and N-H bend (1 Amines) at 1621.80 cm^{-1}. Additionally, N-H (secondary amides) band was found at 1526.06 cm^{-1}, -CH2 (alpha-methylene) bending alkane band at 1457.53 cm^{-1}, and Si-CH3 band at 1236.00 cm^{-1}. Moreover, the 1096.79 cm^{-1} band was assigned to secondary cyclic alcohols. These findings confirm the presence of various functional groups, such as alcohol, amines, and carboxyl, as reported by Kristianto et al. [17]. Papaya seed powder's OH, C=O, and C-O functional groups on the surface suggest its potential as an effective polyelectrolyte.

Similarly, FTIR analysis of orange peel powder revealed the -OH group's presence at 3279.67 cm^{-1}, while the CH2 asymmetric stretching vibrations and CH2 symmetric stretching vibrations were identified at 2918.05 cm^{-1} and 2860.51 cm^{-1}, respectively. Additionally, a weak alkyne (C=C) group was found at 2114.24 cm^{-1}, and (C=O) group was detected at 1733.00 cm^{-1} and 1602.57 cm^{-1}. The FTIR spectrum also identified the methylene bending alkane at 1418.64 cm^{-1} and tetramethylsilane at 1232.37 cm^{-1}. Furthermore, the stretching vibration of the C-O group was observed at 1010.65 cm^{-1}. Munagapati et al. [20] identified the presence of alcohols, alkynes, and esters in orange peel powder. Functional groups such as OH and C=O, which can neutralize both positively and negatively charged pollutants in water, may facilitate the coagulation-flocculation process.

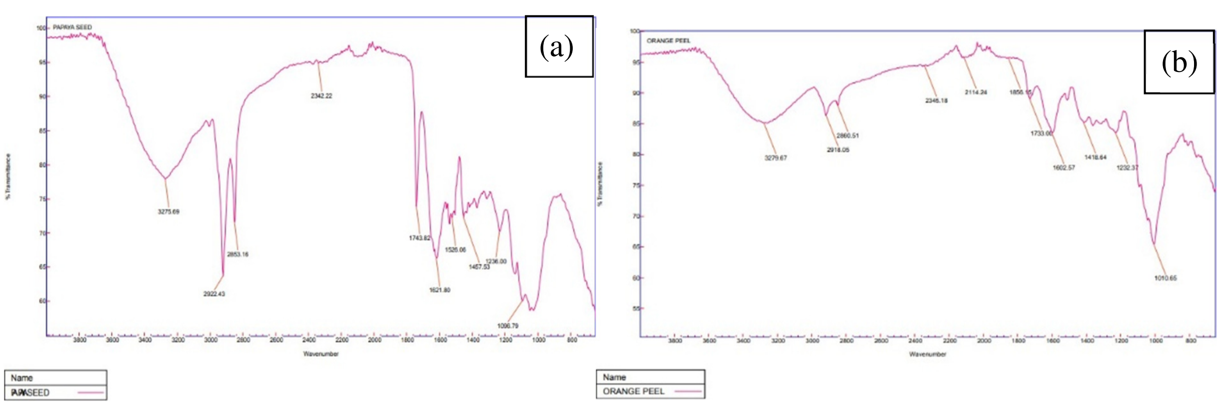

FIGURE 6. The FTIR spectrum of (a) papaya seed powder and (b) orange peel powder

Effect of Coagulant Dosage

In Figure 7, the relationship between coagulant dosage and turbidity reduction is shown. The research concluded that the most effective coagulant dosage was 50 mg/L of papaya seed coagulant, which resulted in a turbidity clearance of 81.71%. Muda et al. [13] found that increasing the coagulant dosage from 10 mg/L to 130 mg/L for papaya seed coagulant improved turbidity removal performance from 92% to 95.5% due to the attractive charges of the particles which caused them to settle together.

For orange peel coagulant, 10 mg/L dosage is the optimum dosage with 76.54% turbidity removal. The lowest turbidity removal recorded was 4.24% at 50 mg/L dosage. This is because high coagulant dosage sometime can lead to excess suspended particle in water that causing turbidity in the water.

As for the combination of papaya seed and orange peel, the optimum turbidity removal was 82.06% at the optimum dosage of 70:30 papaya seed:orange peel. The lowest turbidity removal was 61.96%. for the ratio of 10:90 combination papaya seed and orange peel coagulant.

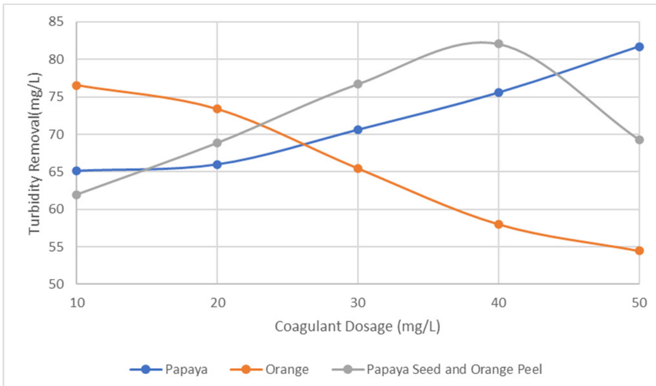

FIGURE 7. Effect of coagulant dosage on turbidity removal

Effect of pH

The effect of pH toward turbidity removal is shown in Figure 8. All three types of coagulant show the highest turbidity removal at pH 7.5. Papaya seed, orange peel and the combination of papaya seed and orange peel able to remove 84.81%, 82.50% and 85.43% turbidity, respectively in greywater at pH 7.5. All three coagulants also show the lowest turbidity removal at pH 12 with 74.72%, 72% and 73.79% for papaya seed, orange peel, and combination of papaya seed and orange peel coagulant, respectively.

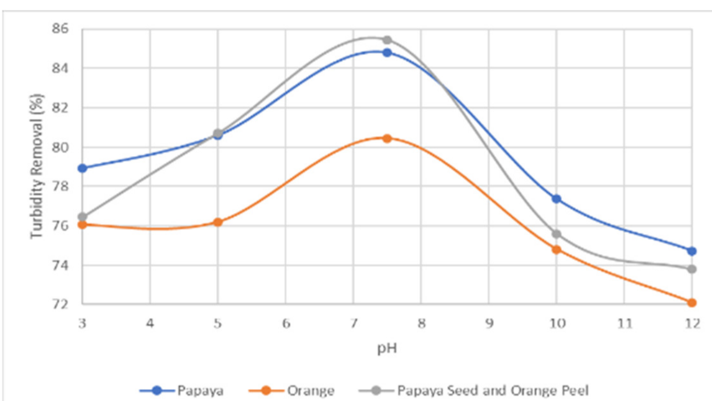

FIGURE 8. Effect of pH on turbidity removal

Effect of Mixing Speed

The coagulation process involves two distinct phases of mixing, and the coagulant dosage and mixing speed can greatly affect the outcome. Figure 9 demonstrates the impact of mixing speed on turbidity removal. For optimal turbidity removal when using papaya seed coagulant on greywater, a combination of fast mixing (170 RPM for 25 minutes) and moderate mixing (60 RPM for 20 minutes) is required. However, the lowest turbidity removal percentages (75.65% and 66.7%, respectively) were observed with quick mixing at 130 RPM for 5 minutes and slow mixing at 40 RPM for 5 minutes. The use of a coagulant made of papaya seeds and orange peels resulted in the best turbidity removal (85.78%) with rapid mixing at 170 RPM for 25 minutes and slow mixing at 60 RPM for 20 minutes, while the worst turbidity removal (67.83%) was obtained with rapid mixing at 130 RPM for 5 minutes and slow mixing at 40 RPM for 5 minutes. The optimal mixing conditions for orange peel coagulant were mixing at 100 RPM for 5 minutes and 25 RPM for 25 minutes, resulting in the best turbidity removal (80.4%), whereas the worst turbidity removal (65.09%) was achieved with mixing at 130 RPM for 5 minutes and 40 RPM for 5 minutes.

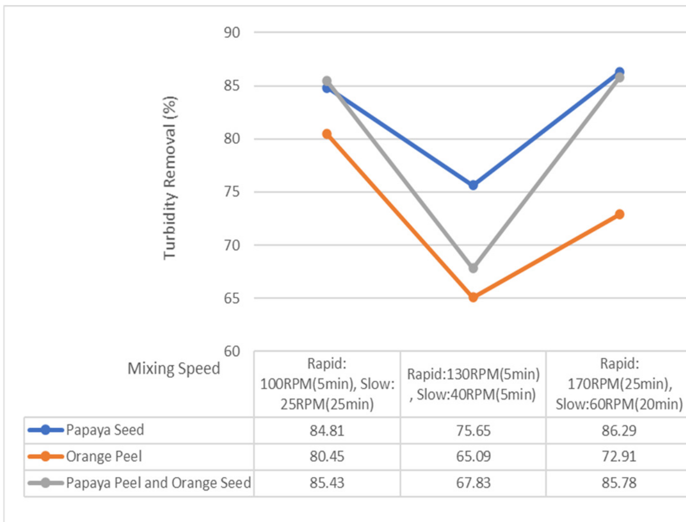

FIGURE 9. Effect of mixing speed on turbidity removal

Effect of Optimum Coagulant Dosage, pH, and Mixing Speed on Greywater Parameters

The investigation of greywater parameters, including total suspended solids (TSS) and chemical oxygen demand (COD), was conducted, and a table was generated to identify the most effective coagulant dosage, pH level, and mixing speed. The TSS and COD values were compared with Environmental Quality (Sewage) Regulations (Standard B) since the location of the greywater sample was not listed in list of catchment areas where Standard A applies as mentioned in Third Schedule of Environmental Quality (Sewage) Regulations.

Total Suspended Solid (TSS)

The initial TSS of greywater was 181 mg/L. TSS removal at optimum coagulant dosage, pH and mixing speed using papaya seed, orange peel and the combination of papaya seed and orange peel coagulant is shown in Figure 10. Orange peel coagulant display the highest TSS removal of 64.64% with final TSS value of 64 mg/L while papaya seed coagulant has the lowest TSS removal of 50.28% with final value of 90 mg/L. The combination of papaya seed and orange peel coagulant able to remove 58.01% of TSS with final value of 76 mg/L. In addition, all three coagulant able to reduce TSS value less than 100 mg/L which considered as the acceptable condition for sewage discharge of Environmental Quality (Sewage) Regulations (Standard B).

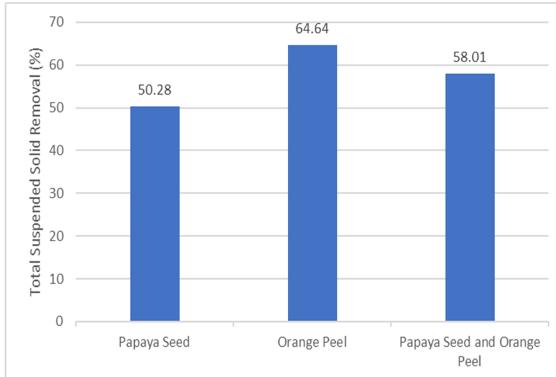

FIGURE 10. TSS removal at optimum coagulant dosage, pH and mixing speed of papaya seed, orange peel and the combination of papaya seed and orange peel coagulant

Chemical Oxygen Demand (COD)

The initial COD of the greywater sample was 476 mg/L. COD removal at optimum coagulant dosage, pH and mixing speed using papaya seed, orange peel and the combination of papaya seed and orange peel coagulant is shown in Figure 11 Papaya seed coagulant show the highest COD removal of 61.34% with the final COD value of 184 mg/L while orange peel coagulant showed the lowest COD removal of 54.62% with final COD value of 216 mg/L. The combination of papaya seed and orange peel coagulant removed 60.08% of COD with 195 mg/L as final value which is slightly near to papaya seed coagulant. Based on the acceptable conditions of sewage discharge of Environmental Quality (Sewage) Regulations (Standard B), papaya seed and the combination of papaya seed and orange peel coagulant able to reduce COD value less than 200 mg/L. However, orange peel coagulant with final concentration of 216 mg/L was unable to reduce COD value less than 200 mg/L. The coagulation-flocculation process could be attributed to the presence of macromolecules such as carbohydrates, protein, and lipids, with protein being the primary component. The TSS and COD parameters of greywater were improved after treatment by the combination of natural coagulants, papaya seed, and orange peel. Moreover, natural polymers have multiple charged functional groups like hydroxyl (-OH), carboxyl (-COOH), and amine (-NH_2) within their polysaccharide chain, which may be associated with the interaction between the polymer and dissolved particles during natural coagulation.

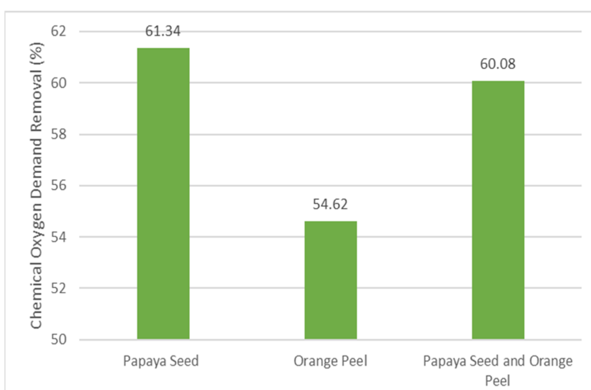

FIGURE 11. COD removal at optimum coagulant dosage, pH and mixing speed of papaya seed, orange peel and the combination of papaya seed and orange peel coagulant

CONCLUSIONS

There are several options available for cleaning greywater samples, including papaya seeds, orange peel, and a coagulant made from both. While the rough surfaces of orange peel powder create high porosity and surface areas, SEM analysis revealed that papaya seed powder has agglomerated particles with non-uniform pore morphology and size. Both papaya seed powder and orange peel powder contain functional groups that can neutralize water contaminants, thus aiding in the coagulation-flocculation process. At a concentration of 50 mg/L and pH 7.5, the use of papaya seed coagulant resulted in the removal of 86.3% of turbidity, 50.3% of total suspended solids (TSS), and 61.4% of chemical oxygen demand (COD). Both fast mixing (170 RPM for 25 minutes) and moderate mixing (60 RPM for 20 minutes) were effective in reducing turbidity. Orange peel coagulant was effective in eliminating 80.5% of turbidity, 64.6% of TSS, and 54.6% of COD when utilized at a pH of 7.5, a dosage of 10 mg/L, a rapid mixing speed of 100 RPM for 5 minutes, and a slow mixing speed of 40 RPM for 5 minutes. The best results were obtained with a combination of papaya seed and orange peel coagulant at a ratio dosage of 70:30, pH 7.5, fast mixing speed of 170 RPM for 25 minutes, and slow mixing speed of 60 RPM for 20 minutes, resulting in the removal of 85.8% of turbidity, 58.0% of TSS, and 60.1% of COD.

The combination of both coagulants shows significant removal owing to the existing of certain functional groups that play roles in coagulation process. In conclusion, papaya seed coagulant had the highest turbidity removal compared to orange peel and the combination of papaya seed and orange peel coagulant. However, orange peel showed significant tss removal than other coagulant. All three coagulants able to reduce tss value less than 100 mg/l which considered as the acceptable condition for sewage discharge of environmental quality (sewage) regulations (standard b). Based on the acceptable conditions of sewage discharge of environmental quality (sewage) regulations (standard b), papaya seed and the combination of papaya seed and orange peel coagulant able to reduce cod value less than 200 mg/l. However, orange peel coagulant with final concentration of 216 mg/l was unable to reduce cod value less than 200 mg/l.

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