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Combined Effects of Banana Peels and Pith as Dual Natural Plant-Based Coagulants for Turbidity Removal

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ABSTRACT

Plant-based coagulants are foreseen for their coagulation ability in par with chemical coagulants owing to their biodegradability, availability, and effectiveness, while chemical coagulants are known to pose a threat to the environment due to generation of toxic sludge. This study investigates the performance of combined banana peels and banana pith as dual natural plant-based coagulants for turbidity removal. Banana peel powder solution (BPPS), banana pith juice (BPJ), and the combination of BPPS and BPJ were prepared via several steps, including drying, grinding, sieving, mixing, and crushing. The three types of coagulant produced were tested in a series of jar test experiments under the effects of different dosages, pH, and mixing speeds. The scanning electron microscopy images of banana pith show the formation of multicellular fibers, while irregular shapes and micro-rough crate-like pores are formed in banana peels, which contribute to the adherence of contaminants. Jar test results revealed the highest turbidity removal of 97.3, 98.0, and 99.1% was attained by using 60, 80, and 20:20 mg/L of banana peel powder solution, banana pith juice, and a combination of banana peel powder solution and banana pith juice coagulant dosage, respectively, with an optimum pH of 2, at a rapid mixing rate of 180 rpm (5 minutes) and a slow mixing of 60 rpm (20 minutes). Significantly, this removal was attributed to the presence of proteins and polysaccharides in both banana peels and pith. The findings suggested that banana peels and pith are applicable as natural plant-based coagulants for turbidity removal in the water treatment process and a potential substitute for conventional coagulants. In addition, the combined banana peels and banana pith coagulant prove to be more efficient than the existing banana peels and banana pith coagulant.

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1. Introduction

Water serves as the greatest gift of nature for the endurance of ecological system and human beings. The rapid growth of population and accelerated pace of industrialization increased the demand of freshwater [1]. The anthropogenic activities lead to water quality deterioration that subsequently pose threats to human health [2]. The water from precipitation that flows into different water bodies such as rivers, lakes, ponds and reservoirs contain dissolved and suspended materials which included organic and inorganic matter, organisms from industrial effluents, domestic, agricultural and land erosion [3,4]. Surface water often carries colloidal and suspended particles that affect the water quality [5]. Suspended particles increase turbidity as it significantly worsens aquatic lives by absorbing light. Nutrient increase the growth of unwanted phytoplankton where it also absorbs light, leading to waterborne disease outbreaks, impacts recreation and tourism [6]. Various conventional water treatment process has been used to treat river water in reducing turbidity to the permissible level. Nevertheless, the challenge usually associated with the overuse of chemical coagulants such as alum which contributed to the generation of abundant sludge. Although alum display high coagulation performance and cost-effective treatment, but it is not biodegradable. The residuals could remain in the water and/or sludge after treatment which lead to water contamination and risk human health [7]. In addition, the usage of alum and poly aluminium chloride in water treatment process have been linked to cause Alzheimer's disease [8]. Therefore, to overcome problems related to hazardous sludge in water treatment, natural plant-based coagulants offer various advantages in comparison to chemical agents or other traditional treatment in terms of low toxicity, biodegradability, and low residual sludge production [9,10].

Coagulation and flocculation are physicochemical processes that commonly utilize in water treatment to reduce turbidity and microorganisms in the water [11]. Coagulation is the process of reagent addition to destabilize colloidal particles by charge neutralization allowing particles to agglomerate and separate from liquid media while flocculation promotes agglomeration and helps particles to settle down [12]. Coagulation is an important step in the water treatment process owing to its simple operation, cheap, and high efficiency in removing suspended particles [13]. During coagulation, the negatively charged suspended particles agglomerate with positively charged coagulants due to adsorption of ions and ionization of surface groups. The mechanisms of coagulation include by charge neutralization, sweep, and bridge formation [14].

Nowadays, there are many types of effective natural coagulants that has been developed from the plants based, animal based, and microbe-based coagulant [15-17]. Examples of animal-based coagulant include chitosan, alginate, and chitin while microbe-based coagulant such as Xanthan gum, *Aspergillus sp.*, *Enterobacter*, *Streptomonas*, and *Bacillus licheniformis* [15,17]. Plant-based coagulant is made from plant such as okra, red bean, sugar cane, corn, *Cactus latifera*, *Moringa oleifera*, and cassava peels [8,9,11,18,19]. The advantages of using natural coagulants in place of chemical coagulants including safe for human health, biodegradable sludge products, easy implementations, low-cost, and high materials availability [20].

Banana is a plant that is typically consumed as fresh fruit and popular across the world especially Southeast Asia [21]. Peels and pith of banana is a non-food waste material that can easily obtained and are the most discarded fruit wastes that can be found in Malaysia [22]. The total yield of banana production in Malaysian showed an upward trend of increase from 2007 to 2012 which results in 806 337.6 tonnes of banana biomass discarded in 2012 [23]. This agricultural by product contribute to the abundant of waste disposed daily in municipal landfills and cause environmental problem [24]. Rather than disposed these peels-piths to the landfills, alternative solution to use it as coagulants for water treatment is beneficial.

Banana peels and pith coagulant are low cost as the materials are made from waste and by-products of banana plant. It is non-toxic and biodegradable compare to chemical coagulant therefore environmentally friendly and sustainable for water treatment process. Both banana peels and pith has been used as natural coagulant to remove turbidity, suspended solids (SS), and some heavy metals. Banana peels able to remove turbidity of grey water up to 66% while banana pith juice display high chemical oxygen demand (COD), SS, and turbidity removal efficiencies of 80.1, 88.6, and 98.5%, respectively. The Inulin content in the pith juice was the active coagulant agent with high bonding capacity to form bridges of flocs [25]. Maurya and Daverey [26] reveals that banana peel powder coagulant reduced total suspended solids and COD of municipal wastewater up to 45.5 and 66.7%, respectively owing to the presence of functional groups such as carboxylic acid, hydroxyl and aliphatic amines. Fu *et al.,* [27] demonstrates that banana peel coagulant able to remove turbidity and color by 85.2 and 76.5%, respectively. Nonetheless, the use of natural coagulants by combination peels-pith from banana has not been investigated before which emphasizes the novelty of the present work. Likewise, coagulation-flocculation attributed to these coagulants is essential as an alternative to the conventional chemical coagulants. Hence, this study aims to remove turbidity in river water sample using banana peel and banana pith as natural coagulant.

2. Methodology

2.1 Water Sampling

The water sample for this study was collected from Muar River, Johor, Malaysia at sampling point located at main Jetty of Pekan Panchor situated at coordinates 2.1730° N and 102.7106° E as shown in Figure 1. Water samples was collected manually at point where the turbidity is high using high-density polyethylene (HDPE) bottles (10L per sample). The collected water sample were stored in a cool box and immediately transferred to the laboratory chiller prior to use. The collected water sample was characterized for turbidity, pH, chemical oxygen demand (COD), dissolved oxygen (DO), biochemical oxygen demand (BOD), total suspended solid (TSS) and ammonia nitrogen (AN) [28]. Table 1 show the physical and chemical analyses of water sample along with the National water quality standards (NWQS) for class I water in Malaysia [29]. The results shows that the turbidity, BOD, and TSS is higher than the standard value while pH is acidic and lower than the acceptable limit.

Fig. 1. Location of river water sampling

Table 1
Physical and chemical analyses of the water sample

Parameter	Unit	Value	NWQS
Turbidity	NTU	40.9±0.071	5
pH	-	5.64±0.028	6.5-8.5
COD	(mg/L)	6.00±0.36	10
BOD	(mg/L)	1.45±0.007	1
DO	(mg/L)	9.05±0.021	7
TSS	(mg/L)	34.0±0.21	25
AN	(mg/L)	0.02±0.007	0.1

2.2 Preparation of Banana Peels and Pith Coagulants

Banana peels and piths as shown in Figure 2 and Figure 3 were collected from Kripeks Md Shah & Asiyah Sdn. Bhd. factory at Senggarang, Batu Pahat, Johor, Malaysia. The peels and pith were washed thoroughly and rinsed with tap water followed by distilled water to remove any dirt. The washed peels were oven dried at 80°C for 24 hours as shown in Figure 2 (b). The dried peels were grinded into a powder followed by passing through 0.4 mm sieve. The obtained banana peels powder as in Figure 2 (c) of 0.5 g were stirred for one hour in 100 mL of distilled water to obtain homogeneous and well mixing of the banana peels in the water. Then, the mixture was filtered by using 0.45 µm Whatman filter paper to discard the soaked peel powders and obtain the solution called banana peels powder solution (BPPS) as shown in Figure 2 (d).

(a) (b) (c) (d)

Fig. 2. (a) banana peel (b) oven dried banana peels (c) banana peels powder (d) BPPS

For the preparation of banana piths juice, the pith was removed from the stem and separated from the foliage. Then, the pith was chopped to small pieces as shown in Figure 3 (b), about 100 g of the pith was mix and crush using a blender to obtain the banana pith juice (BPJ) as shown in Figure 3 (c). The coagulation experiment using BPJ recommended to conduct on the same day the BPJ produced to avoid any fermentation. For SEM characterization, chopped banana piths were oven dried at 180°C for 24 hours and grinded into fine powder form.

(a) (b) (c)

Fig. 3. (a) Banana pith (b) Chopped banana piths and (c) BPJ

2.3 Jar Test Experiment

The jar test apparatus was used to evaluate the performance of BPPS and BPJ as coagulant on water samples from Muar River at room temperature (26°C). The river water sample of 500 mL was transferred to six clean beakers of 1 L and placed in the jar test apparatus with one of the first beaker was labelled as blank sample (control). As to determine the effect of coagulant dosage, the pH of water sample was kept at its original state, the BPPS and BPJ coagulant dosage was varied between 20, 40, 60, 80, and 100 mg/L. Contrarily, to determine the effect of pH, BPPS and BPJ (different jar test experiment) coagulant was added at the same dosages and the water sample was varied (pH 2, 4, 7, 9, 12) using 1 M HCL and NaOH. The test was started with 5 min of rapid mixing at 100 rpm followed by 20 min of slow mixing speed at 25 rpm and 30 min of settling.

Another jar test experiment for the effect of mixing speed (with optimum dosage and pH) was subjected to three effects of rapid mixing and slow mixing: (5 min of rapid mixing at 100, 140,180 rpm) followed by (20 min of slow mixing at 25, 40, 60 rpm) and 30 min of settling. During settling, 50 mL of the water sample were withdrawn using pipette without agitating the sediment flocs. All these steps were also repeated by using BPPS mix with BPJ of different volume ratio (10:10, 20:20, 30:30: 40:40 and 50:50). The turbidity and pH of the samples were measured. The percentage of turbidity removal was calculated by using Eq. (1):

$$\text{Turbidity removal (\%)} = [\mathrm{T}^0 - \mathrm{T})/\mathrm{T}_0]100 \tag{1}$$

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

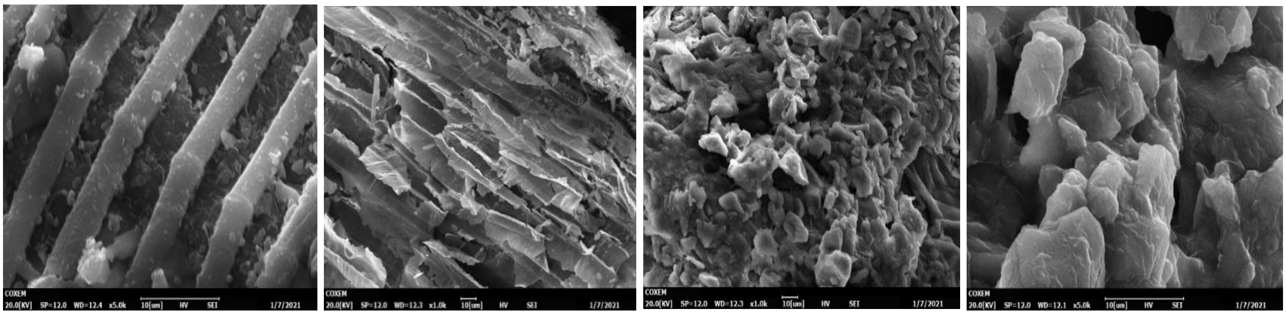
2.4 SEM-EDX and FTIR Analysis of Banana Peels and Pith

The surface morphology and elemental composition of banana peels and pith coagulants before the coagulation process were characterized using Scanning Electron Microscopy (SEM) and Energy Dispersive X-ray (EDX). In this study, a COXEM SEM-EDX (EM-30AX) (Korea) machine was used. The SEM was operated at magnification and at accelerating voltage of 5 kV. The samples were inserted into the SEM machine in a gold coated for the scanning process as described by Farie *et al.,* [30]. On the other hand, the EDX microanalysis is a technique of elemental analysis associated to electron microscopy based on the generation of characteristic that reveals the presence of elements present in the specimen.

The functional groups for BPPS and BPJ was analyzed by using Fourier Transform Infrared Spectroscopy (FTIR). The analyses were recorded on a Perkin Elmer spectrum 100 (USA) FTIR spectrometer. The spectrum was operated between the wavelength region of 600 and 3000 cm^{-1}.

3. Results
3.1 Banana Pith and Peels Characteristics

Figure 4 show the surface morphology for the banana pith and peels powder. Based on Figure 4, the results of SEM micrographs showed that the banana pith has multicellular fibres and fibrils as well as scaly-cellular surface structures. Each fibre displays bundle structures and lumens in the cross section of raw fibre in which each bundle consists of several fibrils. A microporous structure was observed for the image of banana peel at resolution of 1200x that was taken with a particle size of 10 µm (Figure 4 (c) and (d)). The morphology of banana peel showed the coagulant having irregular shapes and porous surface. The irregular shapes and surface exhibited micro-rough texture with crate-like pores that promotes better adherence of contaminants [31].



(a) (b) (c) (d)

Fig. 4 Surface morphology of (a)(b) banana pith and (c)(d) banana peels

Table 2 show the result of EDX analysis in which the majority elements found in banana pith and peels powder were oxygen, carbon, and potassium. Other elements such as chloride and aluminium inside the powder were also observed. The existence of elements was similar to the banana peels and pith as reported by Memon *et al.,* [32] and Alwi *et al.,* [33], respectively.

Table 2
Composition of banana pith and peels coagulants

Element	Weight (%)	
	Banana Pith	Banana Peels
Oxygen	57.23	42.59
Carbon	28.80	36.06
Potassium	9.73	18.48
Chloride	0.77	1.45
Aluminium	0.13	0.25

The removal of pollutant by natural coagulant is a complex phenomenon involving on the degree of copolymerization and ionization on the amount of substituted group within the polymer structure and the functional group [34]. Turbidity removal on using these binding mechanism and elements is attributed to the presence of functional group such as hydroxyl, carboxyl, and amines. To these senses, it is necessary to identify functional group in coagulants [35]. The presence of functional groups in BPPS and BPJ that responsible for turbidity removal is shown in Figure 5. The FTIR range of 600-3000 cm^{-1} reported transmittance peaks at various frequencies presence at different functional groups for both coagulants.

The FTIR spectrum strong peak due to O-H stretching at the frequency of 3262 cm^{-1} and 3255 cm^{-1}. Similar functional groups were observed on the banana peels by Kamsonlian *et al.,* [36] for O-H and C-O functional groups. It is also indicated the existence of free hydroxyl group of polymeric compounds such as lignin or pectin that contain the functional groups of alcohols, phenols, and carboxylic acids. The stretching bands around 1645 cm^{-1} in BPPS and BPJ is ascribed to asymmetric stretching of carboxylic COO- double bond of deprotonated carboxylate functional groups [31]. The deprotonated carboxylates become negatively charged thus increases the availability of binding sites for positively charged compounds [34]. The wavenumber of 579 cm^{-1} and 525 cm^{-1} in BPPS and BPJ, respectively, were contributed by C-I stretching, aliphatic iodo compounds [36]. The presence of few peaks between 1600 and 950 cm^{-1} could attribute to ester, polysaccharide, and proteins in the coagulant [37].

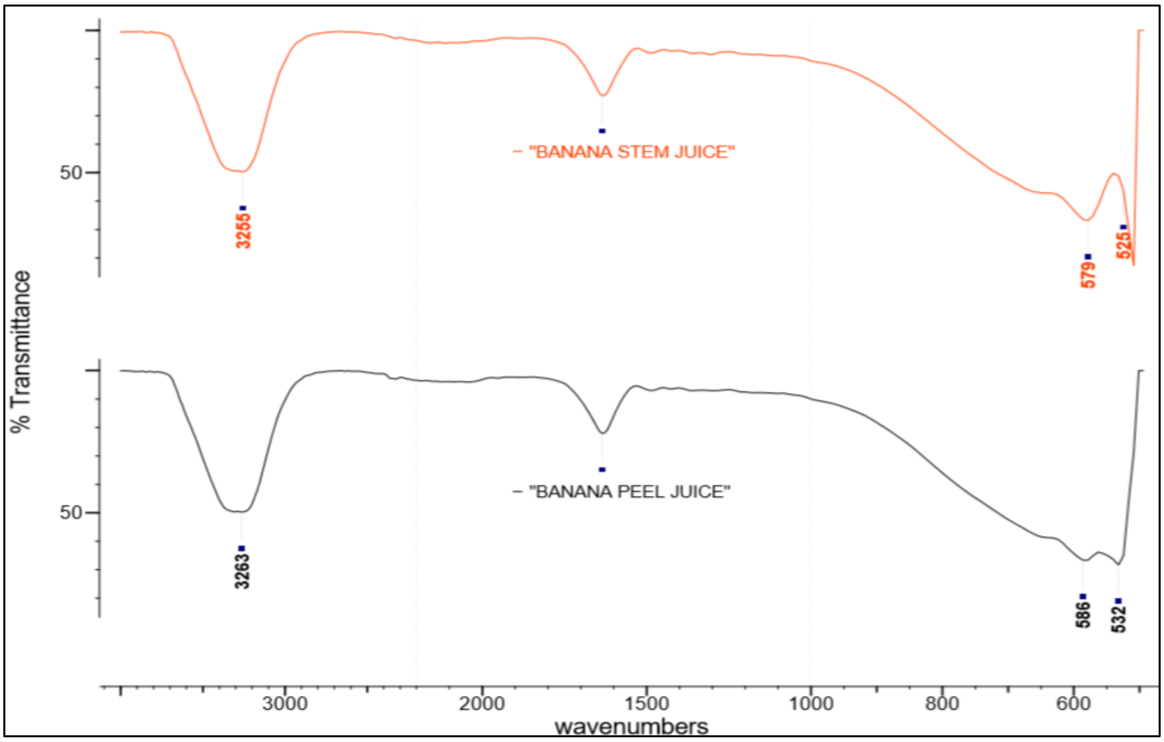
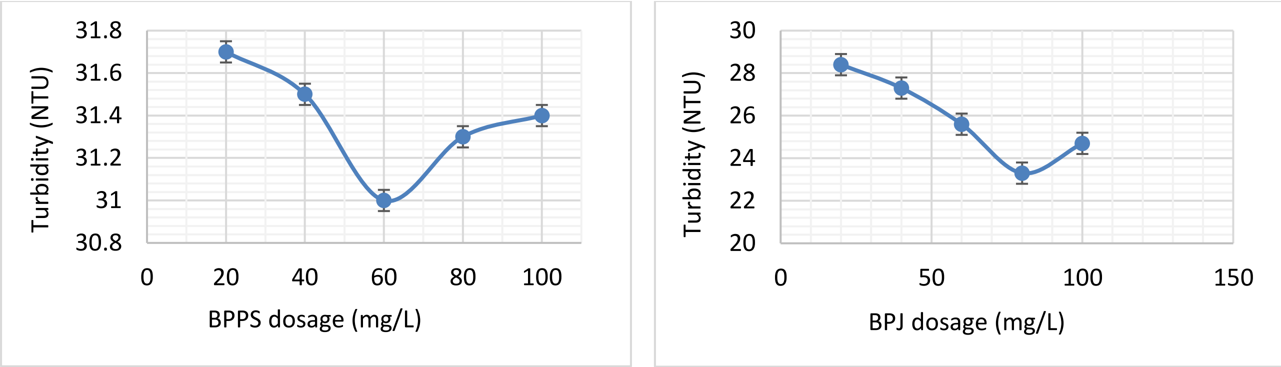


Fig. 5. FTIR spectra of BPJ and BPPS

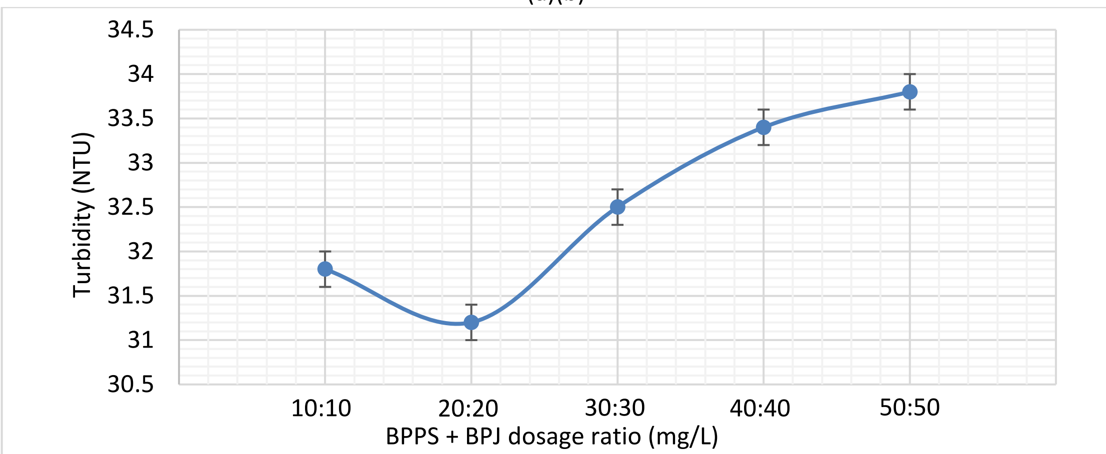
3.2 Factors Affecting the Coagulation Process

3.2.1 Effect of coagulant dosage

The turbidity removal from river water by BPPS, BPJ and BPPS+BPJ coagulants at different doses are presented in Figure 6. Coagulant dosage ranging from 20 mg/L to 100 mg/L were used without altering the pH of the raw water sample. Such condition was to measure the capability of the coagulant to work at natural pH state of river water. The natural pH value of the water samples was in between 5.53 – 5.65 which was moderate acidic stage. The optimum coagulant dosage for BPPS, BPJ and BPPS+BPJ was observed at 60, 80, and 20:20 mg/L, with remaining turbidity of 31, 23.3, and 31.2 NTU, respectively. Only little fine flocs formed and settled at the bottom of the beaker yet, more visible particles can be seen suspended in water sample after settled for 20 minutes. The effect of coagulant dosage with no pH adjustment in the BPPS, BPJ and BPPS+BPJ samples have very little improvement in turbidity removal with reduction of 24.2, 54.6, and 20.6%, respectively at optimum dosage. Low turbidity removal efficiency might be due to high coagulant dosage used, re-stabilization of colloidal particles and/or potential occurrence of phenomena that prevent the formation of inter-particle bridges [38]. However, the fact that residual turbidity did not decrease further at higher BPPS and BPJ concentrations was not expected. This may be attributed to high charge density of the coagulant whereas smaller dosages are adequate for disruption of suspended particulates and larger one’s cause interferences [39]. The increased of BPPS+BPJ coagulant dose had increased the turbidity. This is because the mix of BPPS+BPJ coagulant at higher dosage leads to charge reversal where particles start to restabilizing. Therefore, high dosage of BPPS+BPJ coagulant than the optimum dose (20:20) results in increased turbidity [40].



(a)(b)



(c)

Fig. 6. Effect of (a) BPPS (b) BPJ (c) BPPS+BPJ coagulant dosage on turbidity removal

3.2.2 Effect of pH

The effects of pH show turbidity reduction in water samples to be greatest at pH 2 and 4 (Figure 7). The significant effectiveness of BPPS with the experiment dosage of 60 mg/L, BPJ (80 mg/L) and BPPS+BPJ (20:20) mg/L under the effect of pH showed the obvious floc formations and the water appeared very clear. The result revealed that the optimum pH for achieving high efficiency reduction of turbidity by BPPS was (93.4%; pH 2) (92.8%; pH 4), BPJ (91.7%; pH 2) (90.5; pH 4) as well as BPPS+BPJ (92.3%; pH 2). The reduction of turbidity observed here are similarly to a study reported by Kakoi *et al.,* [34] where the turbidity reduction was 98.56% at a pH value of 4 by banana pith coagulant. According to Modenes *et al.,* [41] the significant effectiveness of banana pith at acidic pH may be associated by the protonation of the functional groups; amino and carboxyl where it resulting in high positive charge that employ firm electrostatic forces over negatively charged coagulated matter. Study shows acidic condition intensifies the coagulating agent in the natural coagulant, resulting in greater coagulation activity and turbidity removal [42]. Coagulant destabilizes the negatively charged colloidal particles present in the water by neutralizing the charges that cause repulsive forces between its colloids [43].

On the other hand, the effectiveness started to drop rapidly when the pH is increased to neutral and moderate alkaline pH except for BPPS coagulant. This is in agreement with the result obtained by Baatache *et al.*, [44]. Depending on the solution pH, the decrease of turbidity removal could be due to different mechanisms either by charge neutralization, polymerization bridging effect, or electrostatic patch. These mechanisms tend to reduce particles surface charge and destabilization of electrical repulsion forces between colloid particles. This subsequently hinder the binding of particles to form larger flocs [45].

The water looked opaque and less flocs after the settling process due to the pH increment in BPJ and BPPS+BPJ coagulant samples. However, as the alkaline pH medium increases, the turbidity removal increases as well. This is supported by Alwi *et al.,* [33] who also found that the turbidity removal increases as the pH level increased from 8 to 10. This could be due to the charges produced by inulin from banana stem juice that act as natural polymer which help in bridging and entrapping the microfloc to form larger floc. Nonetheless, acidic medium showed the highest turbidity removal where pH 2 was chosen as optimum pH for all the coagulants.

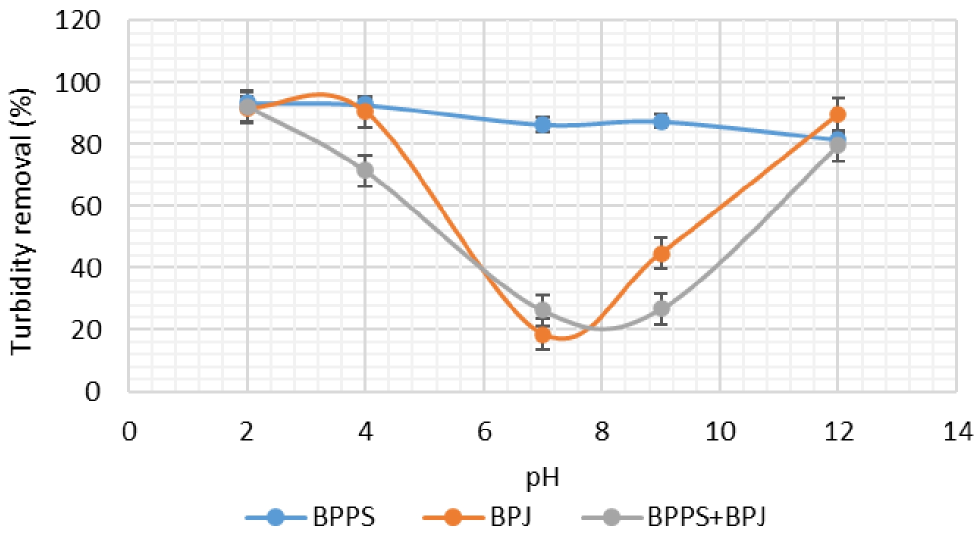
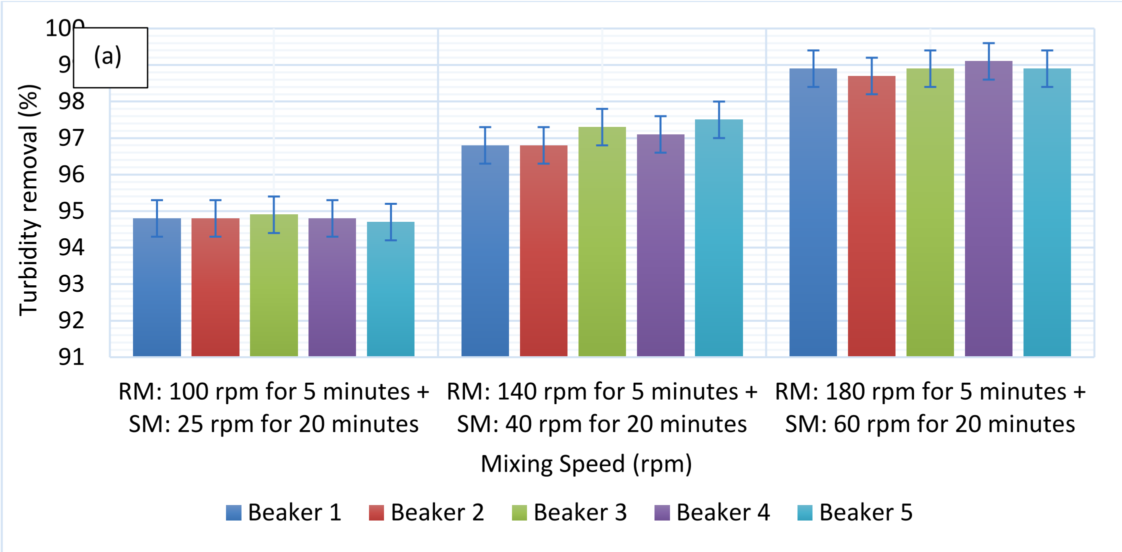


Fig. 7. Turbidity removal (%) vs. pH

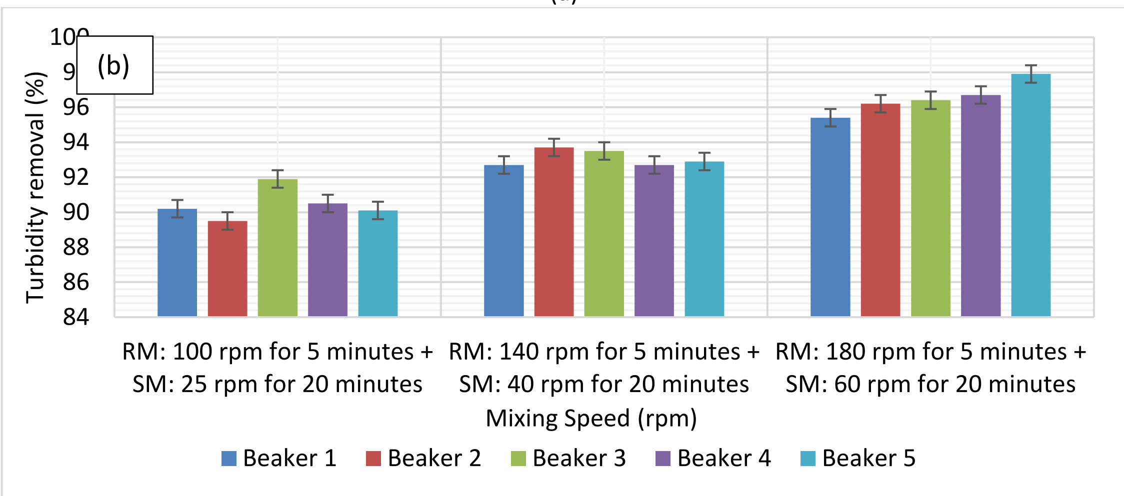
3.2.3 Effect of Mixing Speed

One of the important factors in achieving higher coagulant efficiency during coagulation-flocculation process is mixing speed [46]. Continuous experiments for all coagulants were performed under optimum dosage and pH to investigate the effects of mixing speed conditions on turbidity removal. Figure 8 shows that 5 min of rapid mixing at 180 rpm and 20 min of slow mixing at 60 rpm attained the greatest turbidity removal. BPPPS, BPJ and BPPS+BPJ coagulants able to remove 98.8, 97.9, and 99.1% of turbidity, respectively. In general, the removal efficiency increased with increasing mixing speed.

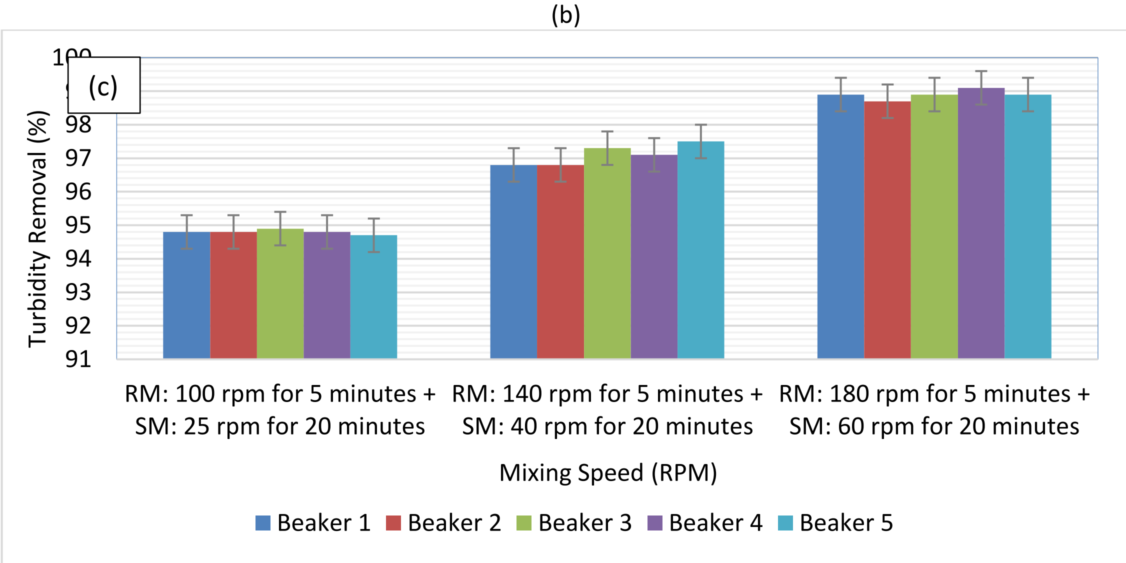
As previously found by Ernest et al., (2017) [39], flocs growth are not significantly influences by a lower mixing speed. In terms of floc settlement, slow mixing of 60 rpm formed larger floc and settled more uniformly compared to slow mixing of 25 rpm and 40 rpm. In this study, slow rpm was not to be considered on the floc formation properties. This finding is consistent that the repulsive forces tend to stabilize the suspension and prevent particle agglomeration [37]. In addition, coagulating agents such as proteins and polysaccharide in banana peels and pith facilitate the coagulation of colloids through the adsorption and bridging mechanisms [44].



(a)



(b)



(c)

Fig. 8. Effect of mixing speed on turbidity removal by (a) BPPS (b) BPJ (c) BPPS+BPJ coagulant

4. Conclusions

Turbidity removal from river water using banana peel and pith as natural coagulant demonstrate promising results for the application in water treatment. High turbidity removal up to 99% was achieved by using banana peels and pith as dual coagulant (BPPS +BPJ) at 20:20 mg/L dosage, pH 2, rapid mixing at 180 rpm (5 min), and slow mixing at 60 rpm (20 min). This significant removal of turbidity was attributed by the presence of coagulating agents (proteins and polysaccharide) that contributed to the coagulation process. The banana peels and pith could be used as potential natural plant-based coagulant for the removal of turbidity. Factors such as dosage, pH and mixing rate play important role in the coagulation-flocculation process. This research can be claimed as a low cost, environmentally friendly, and sustainable coagulants in water treatment process.

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