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Comparison of coagulation, adsorption, ultrafiltration, and hybrid process efficiencies on the kaolin–humic water treatment

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ABSTRACT

Turbidity (i.e., kaolin) and natural organic matter (i.e., humic acid, HA) are the major impurities that affect the surface water quality. To achieve high standard clean water quality, initially three different treatments were employed including coagulation, adsorption (using natural plant-base mangosteen pericarp, MP, as adsorbent), and ultrafiltration (UF) membrane process. Coagulation result shows higher pH contributed to the higher turbidity removal and humic acid removal. Meanwhile pH10 could hasten the sedimentation time to achieve more than 97% and 93% of turbidity and humic acid removal, respectively. Without the coagulant in the water treatment, the sole MP adsorbent was not effective. For ultrafiltration membrane, it exhibited an excellent treated water permeate with high quality (99.9% turbidity and 99% humic acid removal), but the relative flux sharply dropped within the first 5 min of operation due to fouling. Hence, the hybrid treatments (coagulation–adsorption and coagulation–UF) were further investigated to overcome the limitation of the single process. It was found that coagulation–adsorption was the best with high removals of turbidity and humic acid of above 98%. Also, a minimal chemical coagulant was used and lesser energy was needed for process flow compared to those of coagulation–UF to move towards green sustainable development.

Keywords: Water treatment; Coagulation; Adsorption; Ultrafiltration; Hybrid treatment

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1. Introduction

Water is an essential element for all living organisms in the world. This important resource would be facing shortage problems in the future due to the increasing water demand. On average, Peninsular Malaysia has consumed about 11 billion m^3 water and it is estimated to be about 18 billion m^3in 2050 [1] . This could intensify the risk of water shortage by the industrialisation point and non-point sources pollutant flowing into the Malaysia inland water. It was reported that half of the total of 1,055 river monitoring stations in Malaysia had been polluted [2]. Meanwhile, the other half needs extensive water treatment for safe water supply. The ubiquitous occurrence of natural organic matter (NOM) in the surface water as well as humic acid (HA) in water resource directly affect the quality of drinking water. Disinfection of surface water with the presence of HA may produce disinfection byproducts (DBPs) which would cause health problems [3]. Besides dissolved organic matter, the high turbidity due to high suspended solid content may also cause health problems. The presence of suspended solid in water bodies acts as a shield that would protect the microorganisms and viruses against disinfection [4], consequently promotes outbreaks of waterborne disease [5]. To meet the requirement of National Water Service Commission where drinking water must achieve a high quality standard and safe to consumer, it is necessary to impose several water treatment processes. Coagulation is one of the conventional water treatment processes used to treat water with high turbidity and high HA content. Higher turbidity and HA content in

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water resource means higher dose of coagulant required for their removal while the optimal alum dosage in the coagulation is normally in the range of 150–1000 mg/L [6]. However, the residue in the treated water would affect human health which potentially causes Alzheimer's disease [7]. Besides coagulation, adsorption can also be applied to remove impurities in water especially the colloidal particles. Activated carbon adsorbent is normally used in adsorption, however the high cost of this type of adsorbent becomes an issue [8]. Recently, with the growing interest on green awareness worldwide, the development of adsorbent using plants is more practical to match the industrial practices [9,10]. Considering sustainable development, a high quality adsorbent with low cost and high energy saving is required. Mangosteen is one of the tropical fruits producing a large amount of wastes where for every 10 kg of mangosteen harvested, about 6 kg of mangosteen pericarp (MP) is generated as waste [11]. The use of MP waste in coagulation process is considered new and further investigation is required. Carcinogenic risk related to traditional coagulants can be reduced since MP is toxic free and has anticancer property [12,13] and the safety of drinking water can be guaranteed. With the advances in water treatment technology, ultrafiltration (UF) membrane has been widely used to meet the water supply standard. However, HA, suspended solid, or colloidal particulate matters are known as major membrane foulants contributing to fouling and become an obstacle in the membrane technology [14]. Membrane pretreatment is necessary to reduce the high turbidity and organic matter present in water.

Alternatively, the hybrid system (i.e., coagulation–adsorption and coagulation–UF) could be proposed to overcome the limitation of each water treatment method as discussed previously. Prior to the further action taken to investigate the effectiveness of the hybrid system in terms of removal efficiency, the capability of the individual water treatment process is also necessary to be investigated. In line with this, the objective of this paper is to study the performance of coagulation, adsorption, and membrane treatment process and the hybrid system on the removal of high turbidity kaolin and HA–kaolin water.

2. Materials and method

This section discusses a series of experiments conducted along this research project. This research is divided into three parts as shown in Fig. 1. The main tasks for the first part were the preparation of mangosteen pericarp adsor-

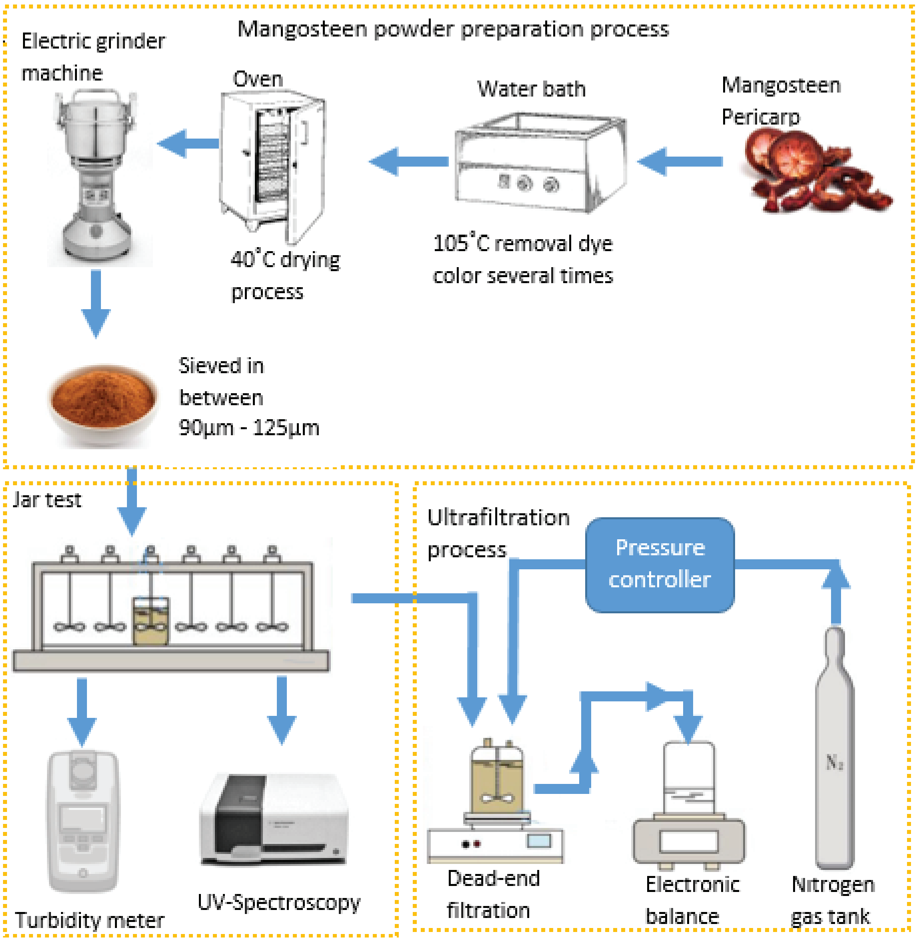


Fig. 1. Overall research flow chart.

bent and synthesis test water. The second part involved the jar test for coagulation and adsorption methods and the last part involved UF treatment.

2.1. Preparation of adsorbent from mangosteen pericarp

The mangosteen pericarps were collected from a wet market in Sungai Lembing, Kuantan. The samples were washed using distilled water to remove the impurities and then dried in an oven at 40°C for 7 d. Finally, it was ground to fine particles using an electric grinder machine (Sigma, IKA@ A11). The mangosteen pericarp particles were then sieved in the particle size range of 90–125 µm and boiled in water for 30 min to remove the colour of the pericarps [15]. The washed mangosteen pericarp powder was dried in an oven at 40°C for 7 d until completely dried. Finally, the fine mangosteen pericarp powder was kept in a tight container prior to use.

2.2. Synthetic test water

Two sets of synthetic water were prepared in this study which is kaolin test water and HA–kaolin test water. Kaolin was used because it can simulate inorganic suspended solids presence in the water [16]. The kaolin stock solution was prepared by adding 10 g of kaolin (Sigma-Aldrich, CAS no: 1332-58-7) in 1 L deionised water (DI) at constant stirring rate of 20 rpm for 1 h and then kept at room temperature for at least 24 h to allow complete hydration of the kaolin [17]. The HA stock solution was prepared by adding 1 g of HA (Sigma-Aldrich, CAS no: 1415-93-6) together with 0.4 g of NaOH (Merck Millipore, CAS no: 1310-73-2) in 1 L of DI water for easier dilution [18].

The kaolin turbidity was adjusted to 125 ± 25 NTU by adding DI into the kaolin stock solution until 1 L solution was achieved. The pH of the prepared turbid water was fixed at pH 7. The HA–kaolin test water was prepared by adding 1 g of HA and 0.4 g of NaOH in a beaker. Then, it was dissolved in 1 Lof DI under continuous stirring for 30 min [19]. Then, 10 mL of HA stock solution was added to the turbid water to form the turbid humic solution and the pH was adjusted to pH 7 as shown in Table 1.

2.3. Jar test experiment

Jar test (Velp, Model JLT6) was used for coagulation and adsorption to determine the efficiency of turbidity and humic acid removal. In the coagulation process, alum was employed as a coagulant while in the adsorption process, the mangosteen pericarp was used as the adsorbent.

Table 1
Characteristics of test water

Parameter	Kaolin test water	HA–Kaolin test water
pH	7	7
Turbidity, NTU	125±25	125±25
Absorbance at 254 nm (UV_{254})	–	0.3±0.025

2.3.1. Coagulation

Coagulation was carried out by controlling the pH solution at different pH values of 4, 6, 8, and 10 with rapid mixing at 125 rpm for 1 min and followed by slow mixing at 40 rpm for 29 min and then 30 min of quiescent settling time. The desired amount of alum was added with rapid mixing to conduct the coagulation process. The alum dosage used in determining the optimum dosage was set in the range of 100–1000 mg/L [6]. To move forward to the green and sustainable development, the minimum alum dosage was selected to ensure the aluminium salt does not overload in the treated water.

2.3.2. Adsorption

Adsorption was conducted in the same way of the coagulation at a constant pH 7, however, no coagulants were added. In this adsorption experiment, the mangosteen pericarp as an adsorbent was added at the same mixing speed and time as previously mentioned in Section 2.3.1.

2.3.3. Coagulation–Adsorption Process

For the coagulation/adsorption process, the pH values were set at 4, 6, 8, and 10. The determined alum and mangosteen pericarp dosages were added with the same mixing speed and time to maximise the contact between the mangosteen pericarp and synthetic test water. The dosage of the mangosteen pericarp used in this study was between 50 and 300 mg/L [20,21]. After 30 min of sedimentation time, the solution was collected at about 2 cm depth from the surface water to be analysed.

2.4. Ultrafiltration (UF) process

Normally, the colloid and humic acid removal by ultrafiltration (UF) is highly practiced because of its higher removal efficiency to entrap the suspended particles and colloidal materials to meet the drinking water standard. The commercial UF membrane in this research study was purchased from Amfor Inc., China, with a molecular weight cut-off (MWCO) of 50 kDa. The ultrafiltration experiments were conducted using a dead-end stirred cell ultrafiltration unit (Amicon, model 8200) with a maximum volume of 300 mL. Initially, the membrane was compacted at a constant pressure of 150 ± 5 kPa for 30 min to ensure that the solution is able to fully occupy the membrane pores [22] and hence a steady flow can be achieved. Prior to the experiments, a membrane with an effective area of 14.6 cm^2 was cut in a circular shape and was soaked in DI for overnight to eliminate the preservation layer on the membrane surface [23]. The membrane filtration was conducted in the following two steps:

Step 1: The membrane deionised water flux (DWF) as shown in Eq. (1) was determined by measuring the volume, V (L) of the collected DI at a constant pressure of 150 ± 5 kPa for 30 min.

$$DWF = \frac{V}{A \times t} \tag{1}$$

where A is the effective membrane area (m^2) and t is the time (h) taken to collect the volume.

Step 2: Step 1 was repeated using the test water as the feed solution. To reduce the concentration polarisation effect, the stirring speed of 500 rpm was applied. Then the permeate solution test water flux (TF) as shown in Eq. (2) was determined by measuring the permeate volume collected at a constant pressure of 150 ± 5 kPa for 30 min.

$$TF = \frac{V}{A \times t} \tag{2}$$

The permeate was weighed using an electronic balance and connected to a computer for data logging and the permeate mass was recorded for every 10 s. The recorded mass changes were converted to volume to calculate the flux using water density.

Then the ratio between the test solution flux (TF) to deionised water flux (DWF) which is expressed by the relative flux (RF) index was used to investigate the degree of fouling using Eq. (3).

$$RF = \frac{TF}{DWF} \tag{3}$$

2.5. Coagulation–UF process

Coagulation–UF was carried out at a constant pH 7 by performing coagulation process first and followed by UF. The same procedures were applied for the coagulation (Section 2.3.1) and the treated water from the coagulation was fed into the dead-end ultrafiltration membrane unit and the process was conducted as described in Section 2.4.

2.6. Water characteristic measurement

The water turbidity and humic acid content were measured using a turbidity meter (Hach, model 2100P) and UV–visible spectrophotometer (Hitachi, model U-1800), respectively. The humic acid content was determined at 254 nm wavelength and humic acid removal was calculated using Eq. (4):

$$\%\ UV_{254} removal = \left(1 - \frac{A_f}{A_o}\right) x100\% \tag{4}$$

where A_0 and A_f are the absorbance values (Abs) for initial and after the treatment, respectively.

Turbidity removal was measured using Eq. (5) where T_0 and T_f are the initial and final water turbidity values in NTU unit, respectively.

$$\%\ Turbidity\ removal = \left(1 - \frac{T_f}{T_o}\right) x100\% \tag{5}$$

3. Results and discussion

3.1. Coagulation

The optimum alum dosage used in the coagulation experiment should be determined first. Fig. 2 shows the effect of alum dosage on kaolin test water and Fig. 3 shows the effect of alum dosage on HA–kaolin test water. For kaolin test water, a dosage of less than 0.05 g/L did not satisfy the impurity removal. Beyond that, the removal efficiency increased sharply until the dosage of 0.2 g/L and gradually increased after 0.2 g/L of alum dosage. However, a significant difference was observed in the HA–kaolin test water when the alum was added until 0.1 g/L and a stable removal was achieved starting from 0.2 g/L of alum.

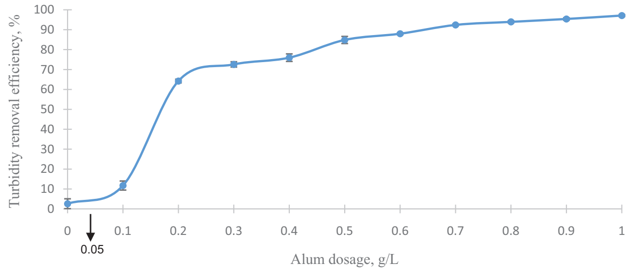


Fig. 2. Removal efficiency for the kaolin test water against alum dosage.

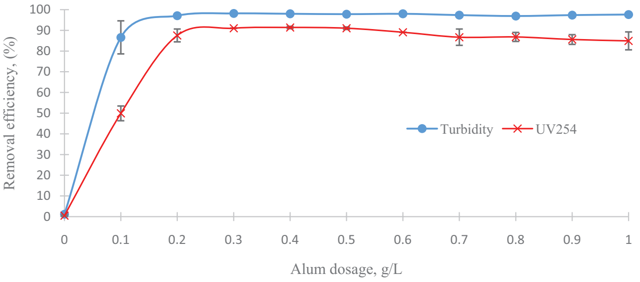


Fig. 3. Removal efficiency for the HA–kaolin test water against alum dosage.

This is due to the presence of humic acid formed by the adsorption on the surface of Al(OH)$_3$ to form the floc faster [24] and led to the aggregation of the colloidal particles by bridging between the humic acid and colloidal particles. The presence of the strong charged functional group in humic acid prompted the charge neutralisation, entrapment, and particle bridging. It is supported by Hsiung et al. [25] that the presence of humic acid offers a stronger adsorption force to agglomerate. The kaolin test water needs a higher alum dosage for neutralisation and destabilisation to complete. For the following experiment, 0.2 g/L of alum was selected because a stable had been achieved. However, pH plays a significant effect on the coagulation process [26]. To ensure the effective coagulation condition could be achieved, various pH values on the coagulation effect were investigated. Fig. 4 shows the turbidity removal efficiencies under the influence of pH and Table 2 shows the residual turbidity values after the coagulation process.

The removal efficiency increased as the pH solution was increased. The sedimentation time decreased to produce higher turbidity removal efficiencies. Whereas, at the low pH value, the sedimentation took an additional 30 min compared to those at the higher pH values. This indicates that higher pH value is able to shorten the sedimentation time of clarifying the turbidity. In addition, the higher pH in the solution increases the charge density, in which the neg-

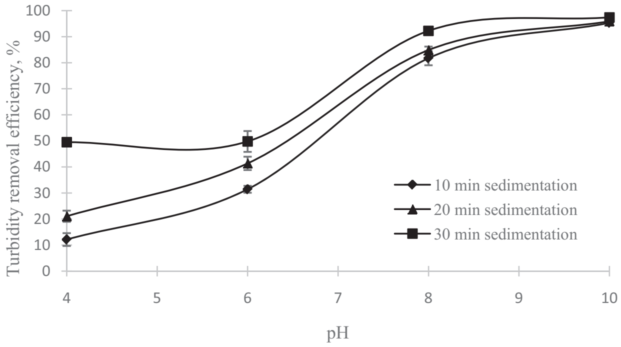


Fig. 4. Turbidity for the kaolin test water removal efficiency at various pH values and sedimentation times.

Table 2
Turbidity for kaolin test water at various pH values and sedimentation times

pH	Sedimentation time, t = 10 min	Sedimentation time, t = 20 min	Sedimentation time, t = 30 min
	Turbidity, NTU	Turbidity, NTU	Turbidity, NTU
4	111.0	99.6	67.1
6	87.3	74.6	64.0
8	23.1	19.2	9.9
10	6.0	5.3	3.2

ative charge carrier particle approaches near the colloid by linkage to the coagulant to form denser colloids of the floc formed. This enables the colloids to coalesce and aggregate strong attractive force between the colloidal and floc [27].

It is recorded that the maximal turbidity removal at pH 10 was up to 97% after 30 min of sedimentation. In the first 10 min of sedimentation, 95% of turbidity removal was achieved until 6 NTU. There was a slight difference in the residue turbidity between the first 10 and 30 min sedimentation. This is due to the neutralisation and stabilisation in the early stage and reaches a stable point after 10 min of aggregation process. In addition, the coagulation did not affect the suspended particles at low pH value. The aggregation was slow and ineffective where on 51% turbidity removal was achieved in 30 min.

Fig. 5 indicates the pH effect on the HA–kaolin test water and the effect is more significant than that on the kaolin test water. Humic acid acts in bridging the intermolecular aggregation that supplements the intramolecular interaction to one particle to another in the solution [28] and it is sufficient to sweep the floc to remove turbidity and UV_{254} in the solution. The presence of humic acid in the low pH solution slows down the removal efficiency since the carboxyl groups present in the humic acid make it difficult to be hydrolysed because of the high concentration of H^+ [29]. As shown in Table 3, the UV_{254} at pH 4 was higher where the turbidity and UV_{254} increased with the increasing pH values. At higher pH values (6–10), the negatively charged functional groups on humic acid with the positively charged acceptor site on the aluminium hydroxide surface would enhance the coagulation process [30].

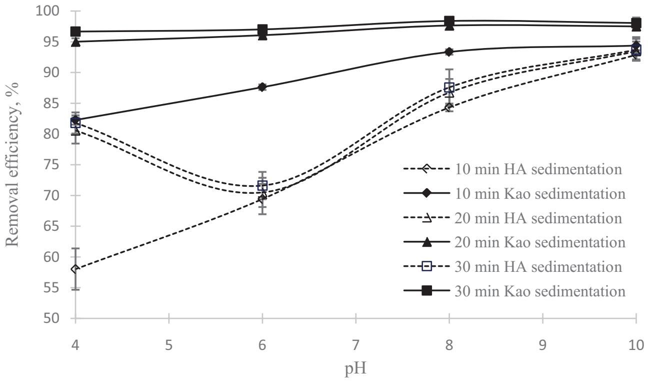


Fig. 5. Turbidity and UV_{254} for HA–kaolin test water removal efficiencies at various pH values and sedimentation times (continuous line is turbidity removal and dashed line is UV_{254} removal).

The residual turbidity and UV_{254}values at low pH differ in the first 10 and 30 min of sedimentation times. In contrast, at higher pH values, the turbidity and UV_{254}were almost the same after 20 min of sedimentation time which indicates a maximum turbidity and UV_{254} removal by 0.2 g/L of alum had been achieved and no further significant removal was observed beyond that.

3.2. Adsorption using Mangosteen Pericarp adsorbent

Table 4 shows that the turbidity and UV_{254}increase as the mangosteen dosage increased. This indicates the solely mangosteen adsorbent used was not able to improve the removal, whereas it contributed some amount of impurities hence the increased turbidity and UV_{254}. The fine particles of the mangosteen adsorbent cannot coalesce with the suspended and colloidal particles to aggregate. At the same time, the mangosteen particles were suspended in the water.

This may be due to the anthocyanin compound pigment agent in the mangosteen pericarp that had been disrupted by the dynamic stirring in the jar test and dissolved into the test water. Likewise, a study from Santos et al. [31] revealed that the external force can lead to the increase of solubilisation of the solute into the solvent. Furthermore, without the initially aggregated agent, the mangosteen pericarp particles cannot perform its bridging linkage function to one another in the solution.

3.3. Membrane ultrafiltration

Fig. 6 shows the RF for both test waters. The results indicate that the membrane was quickly fouled within the first 6 min. For the HA–kaolin test water, the flux decreased rapidly within first 3 min, because humic acid increased the compaction of the foulant onto the membrane surface together with the kaolin colloid. Teow et al. [32] stated that the humic acid aggregates on top of the cake layer (kaolin foulant layer) retarded the membrane flux. The denser membrane foulant on the membrane surface resists the membrane filtration. That is the reason the membrane filtration flux for HA–kaolin test water was worse than the

Table 3
Turbidity and UV_{254} for HA–kaolin test water at various pH values and sedimentation times

pH	Sedimentation time, t = 10 min		Sedimentation time, t = 20 min		Sedimentation time, t = 30 min	
	Turbidity	UV_{254}	Turbidity	UV_{254}	Turbidity	UV_{254}
4	7.3	0.122	6.4	0.056	4.3	0.053
6	1.9	0.098	1.9	0.096	1.8	0.096
8	2.0	0.049	1.7	0.049	1.7	0.053
10	1.2	0.025	1.1	0.027	1.2	0.026

Table 4
Treated water characteristics after the adsorption treatment

Mangosteen pericarp dosage, g	Kaolin test water		HA–kaolin test water	
	Turbidity, NTU	Absorbance, UV_{254}	Turbidity, NTU	Absorbance, UV_{254}
0.05	222	–	212	0.337
0.10	231	–	233	0.335
0.15	253	–	238	0.340
0.20	285	–	275	0.338
0.25	289	–	292	0.345
0.30	304	–	299	0.351

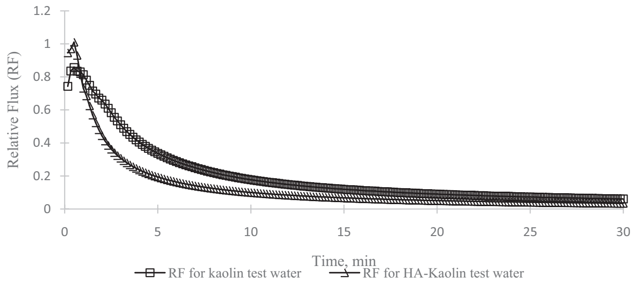


Fig. 6. Relative flux for the kaolin and HA–kaolin test waters.

kaolin test water flux. Although the flux declined very fast, the permeate quality was still able to reach 0.02 NTU for both kaolin and HA–kaolin test waters, and 0.003 of UV_{254} absorbance. Whereas, the volumes of permeate were 12.5 and 7 mL for kaolin and HA–kaolin, respectively. Further analysis should be conducted to find out the most effective and sustainable way for water treatment.

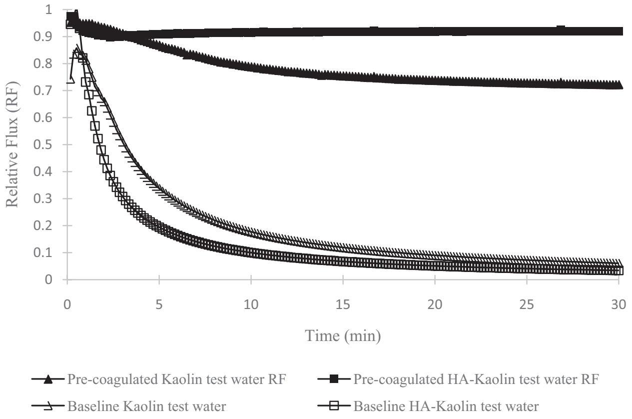


Fig .7. RF for kaolin and HA–kaolin test waters.

Table 5
Treated water characteristics after coagulation–UF

Test water	Coagulation effluent		Permeate	
	Turbidity, NTU	Absorbance, UV_{254}	Turbidity, NTU	Absorbance, UV_{254}
Kaolin	16.4	–	0.02	–
HA–kaolin	1.78	0.074	0.01	0.003

3.4. Coagulation–UF performance

Precoagulation on the membrane filtration was conducted and the results were compared with those of the sole ultrafiltration in Fig. 7. The optimised alum dosage of 0.2 g/L was added into 1 L of test water as a coagulant in the coagulation–UF experiment. Table 5 shows the characteristics of the test water after the process. The kaolin test water treated by ultrafiltration removed 99.8% of turbidity and the RF increased from 0.06 to 0.72 which is 12 times higher compared to that of the sole ultrafiltration within 30 min of the membrane filtration. With the presence of a small quantity of humic acid in the solution, the turbidity removal increased close to 100% and 99% of UV_{254} removal can be achieved. Meanwhile, RF increased from 0.03 to 0.92 or 30 times higher its flux performance.

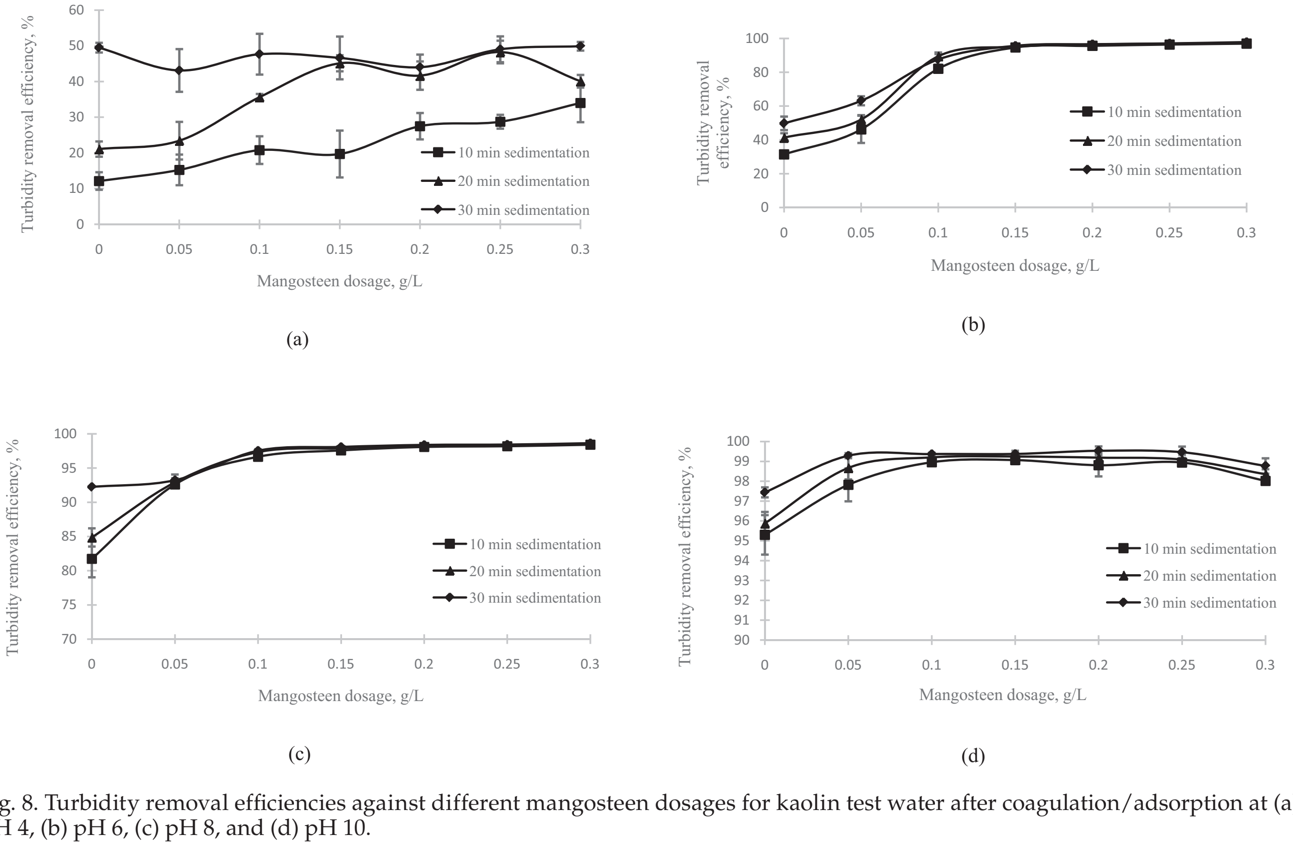
The better UF performance in the test of kaolin-HA with pre-coagulation compared to pre-coagulation kaolin only could be attributed to the better coagulation performance (lower turbidity) as shown in Table 5. However, with the presence of HA in pre-coagulated kaolin-HA water, it was expected that HA may contribute to a major fouling compared to pre-coagulated kaolin test water (without HA) but this did not happen in this current study. Even though many studies proved that HA is an important membrane fouling component, there are a few reports on the effect of membrane hydrophilicity on HA fouling behaviour [33]. In addition, the presence of scanty humic acid in the solution can impel to increase the aggregation. The presence of the humic acid in the solution offers a stronger adsorption force to agglomerate [25]. The residue humic acid in the treated water increased the hydrophilicity of the membrane in the ultrafiltration process [34]. Likewise, the presence of humic acid increased the membrane filtration [35]. Consequently, loosening the void between the membrane surface and foulants enables the solution to penetrate. A research from ZEO Health Ltd. [36] showed that the humic acid acted as a dilator to increase the wall membrane permeability. The treated water amounts from the sole membrane filtration process of the kaolin and HA–kaolin test water were 12 and 7 mL, respectively, which increased to 148 and 189 mL, respectively, after underwent the coagulation/membrane filtration.

3.5. Coagulation–Adsorption performance

Fig. 8 shows the turbidity removal efficiencies for kaolin test water at various mangosteen dosages, sedimentation times, and pH values. The coagulation/adsorption process can harvest around 950 mL of treated water after filtered by filter paper to remove the residual adsorbent on the water surfaces.

From Fig. 8, with the increasing of the pH values, the turbidity removal efficiency increased as well. However, beyond 0.1 g/L of the mangosteen dosage, the removal efficiency achieved 87% and became stable. The optimum dosage recorded was between 0.15 and 0.25 g/L. This is contributed by the enmeshment where the mangosteen pericarp adsorbent acts as the condenser nuclei of the precipitates or becomes enmeshed as the precipitates settle. Fig. 9 shows the removal efficiencies in HA–kaolin test water and the turbidity and UV_{254} recorded a higher removal efficiency of 95% and above compared to those in the kaolin test water. Furthermore, with the presence of humic acid in the coagulation/adsorption hybrid process, it did not only improve the removal efficiency but also hastened the time taken. The sedimentation time of 20 min can achieve the similar treated water quality, which is equivalent to the water quality treated at the 30 min of sedimentation. However, the higher pH value resulted in the higher UV_{254} removal. As discussed earlier, the presence of humic acid in the solution offers a stronger adsorption force to agglomerate because humic acid has higher sorption density at higher pH value [37]. The presence of the mangosteen pericarp aids to increase the nuclei precipitator between humic acid and mangosteen pericarp and offers a large enmeshment of the colloidal particles in the solution.

The mangosteen pericarp in these experiments acts as a nucleation precipitator to the growth of the floc in denser



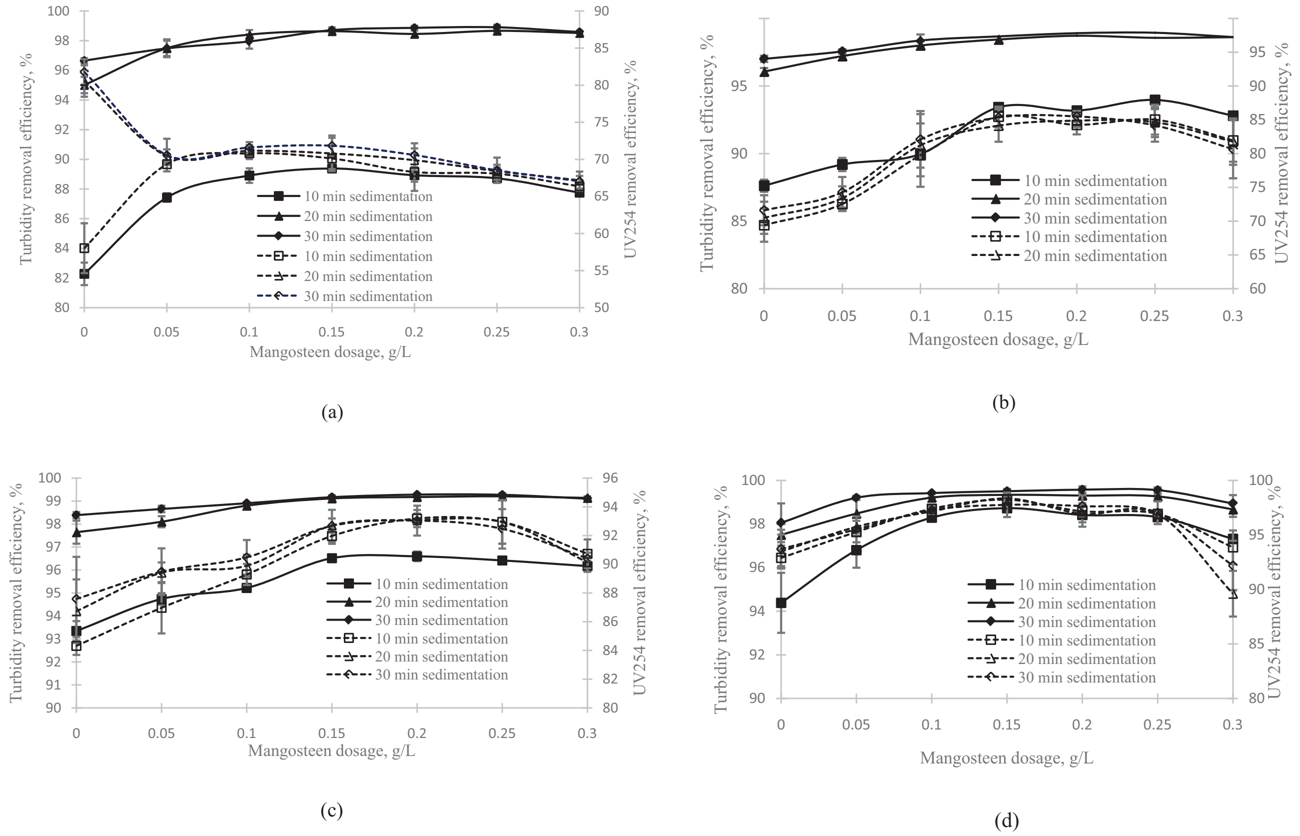
(a)

(b)

(c)

(d)

Fig. 8. Turbidity removal efficiencies against different mangosteen dosages for kaolin test water after coagulation/adsorption at (a) pH 4, (b) pH 6, (c) pH 8, and (d) pH 10.



(a)

(b)

(c)

(d)

Fig. 9. Turbidity and UV_{254} removal efficiencies against mangosteen dosages for HA–kaolin test water after coagulation/adsorption at (a) pH 4, (b) pH 6, (c) pH 8, and (d) pH 10 (solid line is turbidity removal and dashed line is UV_{254} removal).

mass to hasten the sedimentation. This is in agreement with a previous study that reported on the long linear side carbon chain that adsorbs more suspension colloidal particles and promotes the bridging effectively [37]. However, the addition of adsorbent in the coagulation process may not effective for turbidity removal but it is worth for UV_{254} removal process because a minor amount of mangosteen pericarp adsorbent can increase the UV_{254} removal efficiency especially at pH solution (pH 6 and pH 8) close to neutral pH as shown in Figs. 9b and 9c.

3.6. Fouled membrane

Fig. 10 shows the FTIR spectra for the purpose of analysing the foulants on the membrane surface. All of the membranes show the same wavelength of the basic characteristics of PES at 1147 cm^{-1} for the symmetrical vibration of the sulfone group (O=S=O), 1235 cm^{-1} for the C–O–C bond of PES, 1482 cm^{-1} for the C–N stretching vibration, 1573 cm^{-1} corresponds to the $-CH_2CH_2O-$ group in the main polymeric chain [38–40]. The other significant transmittance intensity represents the membrane composition. However, the transmittance intensity was observed to decrease after the filtration process. This lead to the clogging of some pores of the membrane because the foulants could have lessened the penetration of light, thus resulting in a lower transmittance intensity [41]. The transmittance intensity can be minimised with the coagulated test water to produce a similar peak as the virgin membrane. Thus, it signifies that the coagulated test water can minimise the pore clogging.

The transmittance intensity for the membrane filtration in the coagulated water observed has higher light transmittance through the membranes compared to the test water without coagulation. This is because of the foulants that formed a dense layer on the membrane (without coagulation) has reduced the light penetration.

3.7. Mangosteen Pericarp adsorbent characteristics analysis

Fig. 11 shows the surface morphology of the mangosteen pericarp at 20000x and 80000× magnifications. The presence of the rough mangosteen particles with cracked surface morphology as shown in Fig. 10a is the surface contact areas which increased with the solution. Moreover, the solution dispersion across the rough mangosteen particles increased the friction force between the suspended particles and mangosteen pericarp [42] which impelled the precipitate nucleation but with the condition that the initial coagulant bridging must form. With the presence of alum coagulant as previously studied [43], MP interstices increase its surface contact areas and roughness. This interstice entrapped the impurity from the solution which was then removed. The presence of cracks plays a major role in biosorption [44]. Unfortunately, without alum coagulant it caused colloidal particular to suspend in the water.

Fig. 12 shows the FTIR spectra for MP. There are active groups present which included adsorption bands at 2111, 1600, 1434, and 1018 cm^{-1}. These bands are quite similar to other natural substances used in adsorption process [45]. It is supported by previous researchers who find that the spectra between 1600 and 1400 cm^{-1} increased the adsorption of the flocs to aggregate [13]. The compounds present in the MP particles with a long-linear side chain aid to increase the neutralisation and promote the removal efficiency.

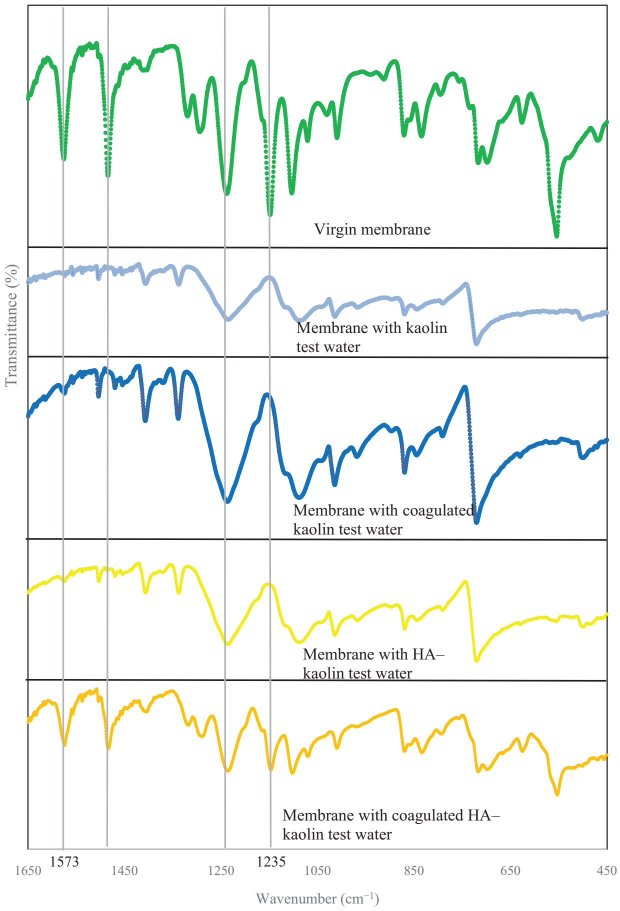


Fig. 10. FTIR spectra of virgin membrane and used membranes.

3.8. Comparison of overall performance

Five different water treatment experiments were conducted and the best criteria were recorded in Table 6. The removal efficiency was evaluated according to the turbidity and UV$_{254}$ removal efficiencies. To keep the research fit with the intention of reducing the chemicals used, the experiments for ultrafiltration and coagulation–UF were set at pH 7 and gave reasonable and acceptable removal efficiencies with not more than 5% differences.

From the results, the highest turbidity and UV$_{254}$ removal efficiencies of 99% were recorded from ultrafiltration and coagulation–UF. However, considering green and sustainable development by reducing the chemical used, the coagulation–UF process would be the best selection because it can reduce the coagulant dosage from 1.0 to 0.2 g/L.

4. Conclusion

The single treatment process of adsorption did not efficiently remove the turbidity and HA. The coagulation and UF process alone were efficiently remove the turbidity and HA, however the higher alum dosage required for

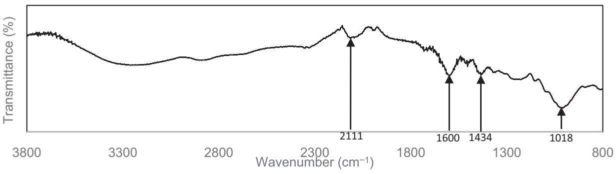
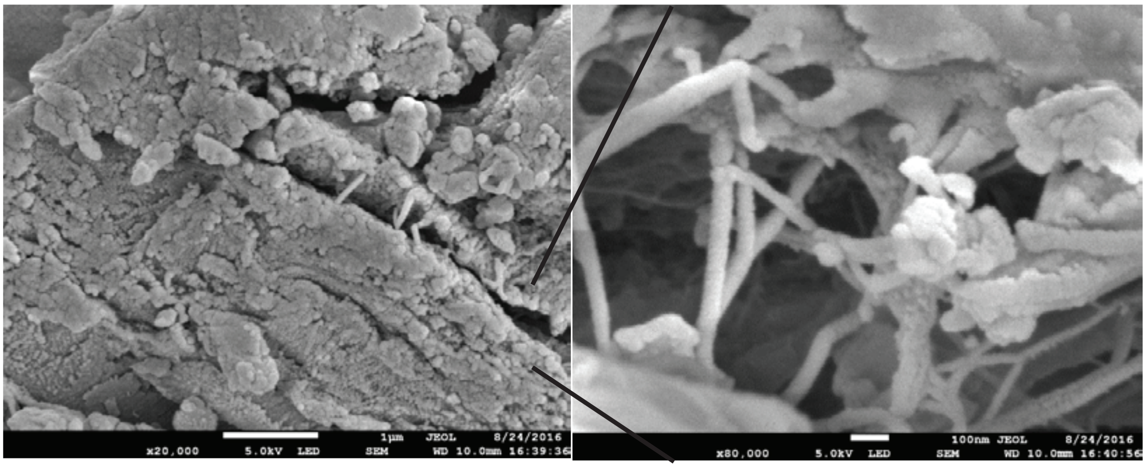


Fig. 12. FTIR spectra of MP.



(a) (b)

Fig. 11. FE-SEM micrographs of the surface of the mangosteen pericarp at (a) 20000× and (b) 80000× magnifications.

Table 6
Removal efficiency criteria

Parameters methods		Turbidity removal efficiency, %	UV_{254} removal efficiency, %	Relative Flux, RF
Coagulation	Kao	97.4 (pH 10)	–	–
	HA–Kao	98.5 (pH 10)	91.3 (pH 10)	–
Ultrafiltration	Kao	99.9 (pH 7)	–	0.06
	HA–Kao	99.9 (pH 7)	99.0 (pH 7)	0.03
Adsorption	Kao	The higher the adsorbent, the higher the turbidity removal		
	HA–Kao	The higher the adsorbent, the higher the UV_{254}		
Coagulation-Ultrafiltration	Kao	99.9 (pH 7)	–	0.72
	HA–Kao	99.9 (pH 7)	99.0 (pH 7)	0.92
Coagulation-Adsorption	Kao	98.3 (pH 8 & 0.2 g/L MP)	–	–
	HA–Kao	99.2 (pH 8 & 0.2 g/L MP)	93.0 (pH 8 & 0.2g/L MP)	–

*Kao is kaolin test water, HA–Kao is humic acid–kaolin test water, coagulant used was 0.2 g/L alum, ultrafiltration was set at 150 ± 5 kPa.

coagulation and severe fouling of UF membrane become a problem. The hybrid processes coagulation–UF and coagulation–adsorption produced a high-quality treated water which would meet the basic drinking water standard. The coagulation–UF enabled to remove 99.9% of turbidity and HA where coagulation–adsorption at higher pH produced a better treated water quality. The coagulation–adsorption process at above pH 8 with 0.2 g/L of mangosteen pericarp adsorbent removed up to 99% of turbidity and UV_{254}. In overall, the coagulation–adsorption hybrid process was considered the best in terms of less energy used with higher water production compared with those of the coagulation–UF process.

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