Citrus fruit peel waste as a source of natural coagulant for water turbidity removal

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Abstract. This research aimed to determine the suitability of citrus fruit peel waste namely as \textit{Citrus Aurantiifolia} (key lime) and \textit{Citrus Microcarpa} (kasturi lime) to act as a natural coagulant for water turbidity removal. The performance of these two coagulants was assessed in terms of turbidity removal using jar test experiments for synthetic low turbid water. The results indicated that the optimum dosage and turbidity removal for \textit{Citrus Microcarpa} and \textit{Citrus Aurantiifolia} was found at 30 mg/l with 75.6\% turbidity removal and 60 mg/l with 74\% turbidity removal, respectively. \textit{Citrus Aurantiifolia} showed higher removal efficiency as compared to \textit{Citrus Microcarpa}. The study demonstrated that both citrus fruit peel waste has the potential to be used as a substitute for chemical based coagulant for a future alternative in water treatment.

1. Introduction
Turbidity is one of the main problems in the water treatment process. It measures the cloudiness and haziness of water caused by the suspended and colloidal particles such as organic, inorganic and biological contaminants\cite{1}. Raw surface water and wastewater are an example of water type of which has the problem due to turbidity. Coagulation-flocculation is a part of the process in water treatment, which widely used due to its effectiveness include destabilization of colloids, removal of organic and inorganic, and pathogen microorganism\cite{2}\cite{3}\cite{4}. Commonly in conventional water treatment process chemical based coagulant such as aluminum sulfate is widely used to remove the turbidity, natural organic matter, and color \cite{4}\cite{5}. Inorganic coagulant such as aluminum sulfate, ferric chloride, calcium carbonate and the synthetic organic polymer (polyaluminum chloride (PACI) polyethylene imine) are an example of coagulants that are being used in water treatment \cite{6}. Some studies have reported that aluminum sulfate and polyaluminium chloride used in coagulation process may lead to Alzheimer’s disease caused by its remaining residue and can develop serious damage to human health such as abnormal tissue in or on brain tissue (brain lesions) \cite{7}\cite{8}\cite{9}. Whereas the application of synthetic polymers has strong carcinogenic effects and neurotoxic \cite{10}. Many studies have been done by previous researchers on the development of natural coagulant of which can be extracted from plant tissues, agriculture, and fruit waste to remove turbidity in water and wastewater treatment \cite{11}\cite{12}\cite{13}.
The main benefits of utilizing natural plant-based coagulants as water treatment material are the availability of the substances, practical in terms of cost, highly biodegradable, safe to human health and environmental friendly [14]. It also produces less voluminous and non-toxicity sludge[15]. Choy et al mentioned that study reported fruit waste is used in treating synthetic turbid water, raw surface water and wastewater. Citrus Microcarpa is known as ‘kasturi lime’ or ‘limau Kasturi’ in Malaysia and Citrus Aurantiifolia, is known as a ‘Key Lime’ or ‘Limau Nipis’. It is believed to have originated in northern India and adjoining parts of Myanmar, or in northern Malaysia. This type of fruit is widely used in Malaysia, mainly as juice for drinks, foods flavor, medicinal application and the peels are thrown away [16]. The benefits of citrus fruit have mainly been attributed to the presence of bioactive compounds, such as phenolics (flavanone glycosides, hydroxycinnamic acids), ascorbic, citric acids, and carotenoids [17][16]. This study aims to determine the suitability of Citrus Microcarpa peels and Citrus Aurantiifolia peels as natural coagulants. This suitability of this fruit will be determined by the optimum dosage to remove the turbidity in water. Thus the implementation of this source as natural coagulant can greatly contribute to reducing the amount of fruit peel waste to be discarded to the landfill.

2. Material and Methods

2.1. Sample Collection
Key lime peels and Kasturi lime peels were collected from several markets around Permatang Pauh, Pulau Pinang and stalls that serve fruit drinks which located near Jalan Perdana (in front of Casa Prima Apartment). The samples then were processed by separating their peels to be used as a natural coagulant. After washing them, the peels were dried for 2 to 3 days under the bright sun to reduce their moisture content. Then the semi-dried peels were air-dried into ventilation oven for about 2 hours at the temperature of 105°C until the moisture content was removed about 95% to improve their ability to break into a fine powder when grinding, since the presence of moisture inside the lime peels would reduce their capability to break apart into fine pieces.

2.2. Preparation of Stock Solution
The ground peels were sieved into a fine powder to obtain the smaller solids with the diameter of less than 1.25 mm, then mixed with distilled water at the ratio of 1 g: 30 ml [18]. The mixture then was stirred for 20 minutes to make sure that the peels solution was well-mixed with the distilled water. Then, the solution was allowed to rest for 30 minutes so that the remaining solids could settle [19]. The mixture solution was filtered using 110 mm filter paper to remove tiny residue inside the mixture solution and to maintain its active components (crude extracts) [20]. The supernatant from the filtration process was used as a natural coagulant.

2.3. Preparation of Synthetic Turbid Water
In this study, synthetic low turbid water was prepared by adding kaolin powder to distilled water for all jar test experiments. Synthetic turbid water was used instead of using raw water samples from a nearby river in order to obtain desirable turbidity ranges. The initial turbidity of the turbid water was set to low turbid (20 NTU – 40 NTU). The synthetic water was prepared by dissolving 2.8 g of kaolin powder in 7 L of distilled water. Then, the kaolin suspension was left to be stirred mechanically for 30 minutes so that the uniform dispersion of kaolin powder could be achieved. This suspension was used as a stock solution for the preparation of water samples.
2.4. Jar Test Experiment
Coagulation and flocculation were carried out using a conventional jar test apparatus. Jar test apparatus consists of six glass beakers and six powered stirrers that function to mix the water in the beakers. Jar testing is proven to be one of the most efficient methods and these procedures are capable to evaluate the minimum or ideal coagulant dose required to achieve certain water quality objectives[3][21][22][7].

Jar Test was carried out as a batch test, with various dosages of coagulants for each of sample material in order to determine the ideal dosage of coagulants for the highest turbidity removal. 500 ml of low turbid water (20 NTU – 40 NTU) were filled into the six beakers. The coagulants were added into the beakers with corresponding to a dosage of 0 (control), 5, 10, 15, 20 and 25 mg/L (set 1), 0 (Control), 10, 20, 30, 40 and 50 mg/L (set 2), 0 (control), 20, 40, 60, 80 and 100 mg/L (set 3). The pH for all sets was adjusted to a constant value of 6.5 using 0.5 M sulphuric acid (H2SO4) and 0.5 M sodium hydroxide (NaOH). The mixture was subjected to 1 minute of rapid mixing at 80 rpm followed by 15 minutes of slow mixing at 30 rpm and 20 minutes of settling. Finally, the sample of supernatant was withdrawn using a pipette in order to determine the turbidity of the sample. In the experiment, the study was conducted by varying dosage of coagulant in order to obtain the ideal coagulant dosage for highest turbidity removal.

2.5. Laboratory testing (Turbidity Removal)
The laboratory testing was conducted to find the initial turbidity and final turbidity of water tested. Turbidity was measured using turbidity meter (2100Q Portable Turbidimeter) and it was expressed in Nephelometric Turbidity Units (NTU).

\[ \text{Turbidity Removal Efficiency (\%) = } \frac{T_i - T_f}{T_i} \times 100 \] (1)

Where \(T_i\) = Initial turbidity
\(T_f\) = Final turbidity

3. Results and Discussion
Table 1 shows a comparative study of turbidity reduction efficiency of coagulants using Lime and Kasturi lime peel extracts corresponding to the various dosage of coagulant. Results indicates that the peel extract demonstrated good performance in improving the quality of water in terms of turbidity removal. Optimal turbidity reduction efficiency with various sets dosage of coagulant (0,5,10,15,20,15) (set 1) mg/L, (0,10,20,30,40,50) (set 2) mg/L and (0,20,40,60,80,100) (set 3) mg/L are: for Citrus microcarpa (kasturi lime) peel extract : 70%, 75.6%, 73.7% respectively; for Citrus aurantiifolia (lime) peel extract: 70.2%, 72.5%, 74% respectively. Citrus Microcarpa peel extract showed optimum dosage for set 1, set 2 and set are 15 mg/L, 30 mg/L and 60 mg/L respectively and using Citrus Aurantiifolia peel extract optimum dosage for set 1, set 2 and set 3 are 15 mg/L, 40 mg/L and 60 mg/L respectively. Seghosime et al reported using the Citrus Aurantiifolia seed instead of peel to treat the low turbid water achieved 64% percentage removal [3]. But in this study, it shows using the Citrus Aurantiifolia peels extract it can reach about 70.2 % removal efficiency.

The fruit peel waste contains bioactive compounds, such as phenolics (flavanone glycosides, hydroxycinnamic acids), ascorbic, citric acids, and carotenoids that would influence the reduction of turbidity removal [17][16]. Asrafuzzaman et al have established that once turbidity reduction efficiency is reached above 70 % (such as those obtained in this study) with natural coagulants, WHO acceptable limits can be obtained through filtration [21]. Ismail et al have reported using dragon fruit peel waste (dragon fruit white fleshed) obtained the optimum dosage of coagulant and turbidity removal is 90 mg/l at efficiency 67 %, respectively [13]. The turbidity reduction using mango duncan and key lime were at 74% and 66.4% respectively [3]. It is also supported from those results were obtained through experimenting moringa oleifera concentration as natural coagulant [23], which reveal turbidity removal 83.2% was achieved at 50 NTU (low turbid water).
Table 1. A comparative study of turbidity reduction efficiency of different coagulants.

| Coagulants     | Set | Dose used mg/L | Initial Turbidity (low turbid water) (20-40)NTU | Final Turbidity NTU | % of turbidity removal |
|----------------|-----|----------------|-----------------------------------------------|--------------------|------------------------|
| *Citrus*       |     |                |                                               |                    |                        |
| *Microcarpa*   |     |                |                                               |                    |                        |
| (kasturi lime) | 1   | 0              | 27.90                                         | 9.95               | 64.3                   |
|                |     | 5              |                                               | 9.88               | 64.6                   |
|                |     | 10             |                                               | 9.48               | 66.0                   |
|                |     | 15             |                                               | 8.38               | 70.0                   |
|                |     | 20             |                                               | 10.27              | 63.2                   |
|                |     | 25             |                                               | 10.05              | 64.0                   |
|                | 2   | 0              | 29.80                                         | 9.99               | 66.5                   |
|                |     | 10             |                                               | 7.46               | 75.0                   |
|                |     | 20             |                                               | 8.50               | 71.5                   |
|                |     | 30             |                                               | 7.27               | 75.6                   |
|                |     | 40             |                                               | 9.43               | 68.4                   |
|                |     | 50             |                                               | 8.68               | 70.9                   |
|                | 3   | 0              | 20.10                                         | 5.83               | 71.0                   |
|                |     | 20             |                                               | 6.16               | 69.4                   |
|                |     | 40             |                                               | 5.76               | 71.3                   |
|                |     | 60             |                                               | 5.28               | 73.7                   |
|                |     | 80             |                                               | 5.84               | 70.9                   |
|                |     | 100            |                                               | 6.12               | 69.6                   |
| *Citrus*       |     |                |                                               |                    |                        |
| *Aurantifolia* |     |                |                                               |                    |                        |
| (lime)         | 1   | 0              | 21.80                                         | 9.08               | 58.3                   |
|                |     | 5              |                                               | 7.98               | 63.4                   |
|                |     | 10             |                                               | 8.41               | 61.4                   |
|                |     | 15             |                                               | 6.50               | 70.2                   |
|                |     | 20             |                                               | 7.50               | 65.6                   |
|                |     | 25             |                                               | 6.76               | 69.0                   |
|                | 2   | 0              | 25.50                                         | 9.94               | 61.0                   |
|                |     | 10             |                                               | 7.78               | 69.5                   |
|                |     | 20             |                                               | 8.36               | 67.2                   |
|                |     | 30             |                                               | 8.50               | 66.7                   |
|                |     | 40             |                                               | 7.00               | 72.5                   |
|                |     | 50             |                                               | 7.12               | 72.1                   |
|                | 3   | 0              | 20.10                                         | 9.15               | 54.5                   |
|                |     | 20             |                                               | 6.00               | 70.1                   |
|                |     | 40             |                                               | 7.31               | 63.6                   |
|                |     | 60             |                                               | 5.22               | 74.0                   |
|                |     | 80             |                                               | 7.15               | 64.4                   |
|                |     | 100            |                                               | 7.76               | 61.4                   |
3.1. Effect of Initial Turbidity on Coagulation Performance

Figure 1(a) shows a graph of Kasturi lime peel dosage against turbidity reduction (NTU) for the three-initial turbidity 27.9 NTU, 29.8 NTU, and 20.1 NTU which categorized as low turbid water. Results indicate that the turbidity reduction for initial turbidity of 27.9 NTU, 29.8 NTU, and 20.1 NTU are 8.38 NTU, 7.27 NTU, and 5.28 NTU respectively. Based on the previous study done by [3] using the same dosage of coagulant (0,5,10,15,20,25) ml the turbidity reduction for low turbid water (50 NTU) was 13 NTU and 16 NTU using Duncan Mango Seed and Citrus Aurantiifolia Seed respectively. It can be proved that Citrus Microcarpa (Kasturi lime) has the potential for improving the quality of low turbid water.

Figure 1 (b) shows a graph of lime peel dosage against turbidity reduction (NTU) for the three-initial turbidity 21.8 NTU, 25.5 NTU, and 20.1 NTU which categorized as low turbid water. Results indicated that the turbidity reduction for initial turbidity of 21.8 NTU, 25.5 NTU, and 20.1 NTU are 6.50 NTU, 7 NTU, and 5.22 NTU respectively. The same pattern of turbidity reduction between Kasturi lime and lime can be seen in this research study. Citrus Microcarpa (Kasturi Lime) shows higher removal efficiency as compared to the Citrus Aurantiifolia due to higher turbidity reduction for the same types of low turbid water.

This result is in support with those are reported on the previous study on turbidity removal using natural coagulants [21].Greater turbidity reduction was found at optimum dosage of 25ml (for both Duncan mango and Key lime seed extracts) and the corresponding turbidity reduction is 12 and 13.3 NTU respectively.

![Graph](image1)

![Graph](image2)

**Figure 1.** Kasturi lime extract dosage against turbidity reduction (NTU) for the low turbid water

4. Conclusions

This study was aimed to determine the suitability of citrus fruit peel waste namely as Citrus Aurantiifolia (key lime) and Citrus Microcarpa (Kasturi lime) to act as a natural coagulant for water turbidity removal by evaluating the best coagulant dosage as well as the percentage of turbidity removal in the water sample. The results indicated that the best coagulant dosage and turbidity removal for Citrus Microcarpa and Citrus Aurantiifolia was found at 30 mg/l with 75.6% efficiency and 60 mg/l with 74 % efficiency respectively. Citrus Aurantiifolia showed greater removal efficiency as compared to Citrus Microcarpa . The study demonstrated that the citrus fruit peel waste had the potential to use as a substitute for chemical based coagulant for a future alternative in water treatment.
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