



Potential Use of Fruit Seeds and Plant Leaves as Coagulation Agent in Water Treatment

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Abstract

The treatment of turbid water by coagulation-flocculation was found to be the most common and cost-effective method. Over the years, chemical coagulants have been widely applied to enhance the coagulation process. However, the utilisation of chemical coagulants exhibits several drawbacks, including generation of voluminous sludge and being non-biodegradable as well as toxic compounds to aquatic life. Due to global concerns over the harmful effects, the application of natural coagulants is a promising solution. Therefore, this study was conducted to investigate the potential of plant-based natural coagulants to replace chemical coagulants for water treatment. Fruit seeds (*Carica papaya*, *Nephelium mutabile*, and *Euphoria malaiense* seeds) and plant leaves (*Pandanus*, *Centella asiatica*, and *Cymbopogon citratus* leaves) were selected as natural coagulants in this study. A series of jar tests were performed using raw water from a water treatment plant. The effects of pH and coagulant dosage were evaluated based on the turbidity removal and coagulation activity. Of all the examined plants, the *Carica papaya* seeds appeared to be the best coagulant for water treatment. An optimum dose of 130 mg/L of this coagulant resulted in a 95.5% turbidity removal and 94% coagulation activity, at optimum pH 7.5. Overall, this study reveals the potential use of fruit seeds and plant leaves as coagulation agents in the water treatment process.

Keywords: Natural Coagulants, Turbidity, Coagulation Activity, *Carica papaya* seeds, Water treatment

1 Introduction

Water is essential to all living organisms as it is the most fundamental nutrient to life. Safe and readily available water is crucial for human health to avoid various diseases due to the presence of harmful bacteria or chemicals in water. However, lack of access to clean water has become one of the world's biggest problems, especially in developing countries. According to Vaithyanathan et.al (1), India is suffering from water crisis which mainly due to improper wastewater and solid waste management, where the population only able to access 4% of usable water sources. Thus, effective water treatment is required to remove undesirable compounds and contaminants so that the water is safe to use for different purposes. A common way to treat turbid water is by using coagulation. Coagulation is a physical-chemical process for water treatment which is typically applied prior to the sedimentation and filtration processes. The application of coagulants in the coagulation-flocculation process is necessary as they are able to remove high suspended solids and colloidal particles in the water, thus enhancing the quality of treated water. The common chemical-based coagulants used in water and wastewater treatment are aluminium sulphate, ferric sulphate and iron (III) chloride (2-4). Karbassi and Pazoki (5) noted that addition of PAC could remove 99% of turbidity in petrochemical wastewater. Despite their efficiency, these chemical coagulants suffer from several drawbacks such as the

generation of large volumes of sludge, are non-biodegradable, and cause detrimental effects to human health and aquatic life (2, 6). The generation of voluminous sludge which is non-biodegradable has become a major problem when chemical coagulants are used in water and wastewater treatment. Therefore, it is essential to replace chemical coagulants with a more sustainable approach to counteract the drawbacks.

Recently, investigations on the usage of natural coagulants in water and wastewater treatment have caught the attention of many researchers. Natural coagulants exhibit many advantages when compared to the application of chemical coagulants in many aspects. They are highly biodegradable (4), non-corrosive (7), generate a lower amount of sludge (8), environmentally friendly, and cheap (7). Moreover, they are less likely to produce treated water with high pH level. Several studies have demonstrated that natural coagulants extracted from plants, animals and microorganisms such as chitosan, *Moringa oleifera*, *Quercus robur* acorn, *Cicer arietinum*, banana pith, *Jatropha curcas* seeds, rice starch, and dragon fruit foliage have excellent efficiency in removing high turbidity in water and wastewater treatment (9-16).

Jatropha curcas seeds, dragon fruit foliage and *Sterculia foetida* seeds achieved high turbidity removals of 99%, 99.2%, and 97%, respectively (9, 15, 17). The application of *Cicer arietinum* showed high turbidity and suspended solid removals of 86% and 87%, respectively (14). Meanwhile, the addition of orange peel as a natural coagulant was able to remove 96% of

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turbidity (18). Furthermore, the addition of natural coagulants extracted from nature can reduce costs and produce high quality treated water. The application of natural coagulants could increase the formation of flocs, thus improving the removal of suspended particles in water during the coagulation process. Although natural coagulants have many advantages, their application as coagulation agents has not been entirely explored in water treatment.

In this study, selected natural coagulants extracted from the seeds and leaves of plants: *Carica papaya* seeds, *Nephelium mutabile* seeds, *Euphoria malaiense*, *Pandanus* leaves, *Centella asiatica* leaves, and *Cymbopogon citratus* leaves have been utilised for water treatment purposes. Unnisa and Bi (19) studied the application of *Carica papaya* seeds as a natural coagulant to remove turbidity and the results showed that *Carica papaya* were able to remove 100% of water turbidity. However, *Nephelium mutabile* seeds, *Euphoria malaiense* seeds, *Pandanus* leaves, *Centella asiatica* leaves and *Cymbopogon citratus* leaves have not been explored as much. Thus, this study aims to investigate the coagulation potentials of these natural coagulants compared to chemical coagulants for the treatment of turbid water.

2 Materials and Method

2.1 Materials

The water samples were collected from an inlet point at the Sultan Ismail Water Treatment Plant (WTP), located near Universiti Teknologi Malaysia, Johor. Fruit seeds and plant leaves: *Carica papaya* seeds, *Nephelium mutabile* seeds, *Euphoria malaiense* seeds, *Pandanus* leaves, *Centella asiatica* leaves and *Cymbopogon citratus* leaves were collected from the surrounding neighbourhood and local market.

2.2 Preparation of natural coagulants

After collecting the materials, the whole seeds of *Carica papaya*, *Nephelium mutabile* and *Euphoria malaiense* were dried in an oven at a temperature between 103°C to 105°C for 24 hours. After drying, they were ground into a fine powder using a grinder. Meanwhile, the fresh *Pandanus*, *Centella asiatica* and *Cymbopogon citratus* leaves were first washed using tap water, chopped and oven dried for 24 hours at a temperature between 103°C to 105°C. The dried leaves then were crushed into a fine powder using a mortar pestle. The powder obtained from both fruit seeds and plant leaves were sieved with a mesh size of 0.4mm and mixed with three different solutions, including deionised water, sodium hydroxide, NaOH and sodium chloride, NaCl. A 10 mg sample of powder from the fruit seeds and plant leaves were weighed and suspended in 200 mL of each solution. The solution was vigorously mixed for 10 min using a mechanical stirrer to extract the coagulation active component, and then filtered. The filtered solutions, called crude extracts, were kept in a refrigerator at 4°C. The crude extracts then can be used as a coagulant for water treatment without any further preparation.

2.3 Coagulation Test

This study used the jar test to determine the coagulation activity of each type of seeds and leaves. Raw turbid water (500 mL) was filled into six beakers and 5 mL of the natural coagulants were added to each beaker. The beaker was rapidly mixed at 200 rpm for 1 minute before slow mixing was applied for about 15 minutes at 60 rpm. The purpose of rapid mixing was to properly disperse the coagulant, while a slow speed mixing was used to

promote particle collision in the water and further facilitate the particle aggregation to form flocs (20). After slow mixing, the flocs were then left to settle for 30 min. Residual turbidity was measured as - T_{fc} . The same jar test was carried out without coagulants and the residual turbidity was measured as - T_{fb} . Coagulation activity was calculated as:

$$\text{Coagulation activity (\%)}: [(T_{fb} - T_{fc}) / T_{fb}] \times 100 \quad (1)$$

2.4 Analytical Methods

A pH meter and nephelometer were used to measure the pH and turbidity of water samples. To determine the optimum condition, the pH of water sample was kept constant at 7.5, while natural coagulants varied between 10 mg/L to 130 mg/L, in order to obtain the optimum dosage. The pH value was varied between 2 to 12 with a constant value of optimised dosage, to determine the effects of pH on treating water. All tests in this study were conducted according to the standard methods of water and wastewater treatment (21).

3 Result and Discussions

3.1 Physical Properties of Natural Coagulants

Table 1 shows the physical properties of the fruit seed and plant leaf-based natural coagulants. *Carica papaya* seeds had the highest initial weight of 333g, while the *Cymbopogon citratus* leaves had the lowest initial weight of 74g as shown in Table 1. After undergo the drying and grinding process, the highest yield weights were shown by the *Nephelium mutabile* seeds with 240 g, followed by 201 g for the *Euphoria malaiense* seeds, 55 g for the *Carica papaya* seeds, 14 g for the *Cymbopogon citratus* leaves, 13 g for the *Pandanus* leaves, and 9 g for the *Centella asiatica* leaves.

Among the natural coagulants, the *Nephelium mutabile* seeds had the lowest moisture content of 20.5 % and this contributed to a greater yield. In contrast, the *Centella asiatica* leaves had the highest moisture content at 89.5 % and resulted in the lowest yield. Table 1 clearly shows that that moisture content plays an important role in determining the yield. Thus, it can be concluded that the *Nephelium mutabile* seeds as natural coagulants are efficient in terms of yield properties.

Table 1: Physical Properties of Natural Coagulants

Materials	Initial Weight (g)	Dry Weight (g)	Yield (%)	Moisture Content (%)
<i>Carica papaya</i> seeds	333	55	16.5	83.5
<i>Nephelium mutabile</i> seeds	302	240	79.5	20.5
<i>Euphoria malaiense</i> Euphoria malaiense seeds	328	201	61.3	38.7
<i>Pandanus</i> leaves	77	13	16.9	81.0
<i>Centella asiatica</i> leaves	86	9	10.5	89.5
<i>Cymbopogon citratus</i> leaves	74	14	18.9	83.1

3.2 Effects of Solvents on Natural Coagulants

Deionised water, sodium chloride (NaCl) and sodium hydroxide (NaOH) were tested in the study to obtain the best solvent to extract natural coagulants. The best solvent was chosen based on the highest removal performance achieved by the natural

coagulants. Fig.1(a) and Fig.1(b) show the performance of natural coagulants in the deionised water. Based on Figure 1(a), the *Nephelium mutabile* seeds had the highest turbidity removal of 31.5% in deionised water compared to the *Carica papaya* seeds (31.3%), *Euphoria malaiense* seeds (30.1%), *Pandanus* leaves (11.6%), *Centella asiatica* leaves (9.9%), and *Cymbopogon citratus* leaves (9.4%). Figure 1(b) also shows that the *Nephelium mutabile* seeds had the highest coagulation activity among the natural coagulants at 10.5%. The coagulation activity for the *Carica papaya* seeds, *Euphoria malaiense* seeds, *Pandanus* leaves, *Centella asiatica* leaves and *Cymbopogon citratus* leaves were 10%, 8.7%, 6.8%, 4.6%, and 4%, respectively. However, based on the results displayed in both figures, all the natural coagulants show a low performance in turbidity removal and very low coagulation activity when deionised water was used as the solvent. Therefore, it can be concluded that deionised water is not suitable to be used as a solvent due to its low capability to extract selected fruit seeds and plant leaves compared to NaOH and NaCl.

Figure 2(a) and Figure 2(b) show the performance of natural

coagulants when sodium chloride, NaCl was added as the solvent. The NaCl solution achieved optimum concentration at 2.0 M as shown in Figure 2(a). At the concentration of 2.0 M, the *Pandanus* leaves had the highest turbidity removal of 59.2%. This was followed by the *Cymbopogon citratus* leaves, *Centella asiatica* leaves, *Nephelium mutabile* seeds, *Carica papaya* seeds and *Euphoria malaiense* seeds with removal rates of 59.1%, 55.9%, 34.6%, 31.8%, and 28.6%, respectively. For coagulation activity, based on Figure 2(b), the *Pandanus* leaves showed the greatest activity at 43.1%, followed by the *Cymbopogon citratus* leaves, *Centella asiatica* leaves, *Nephelium mutabile* seeds, *Carica papaya* seeds, and *Euphoria malaiense* seeds with coagulation activities of 40%, 38.6%, 20.8%, 18.5%, and 18.5%, respectively. From the results, it is clearly seen that all the natural coagulants were unable to achieve high removal performance and coagulation activity in the NaCl solution. This means that the NaCl solution is also not suitable to be used as a solvent in water treatment. Figure 3(a) and Figure 3(b) show the performance of natural coagulants in a sodium hydroxide, NaOH solution.

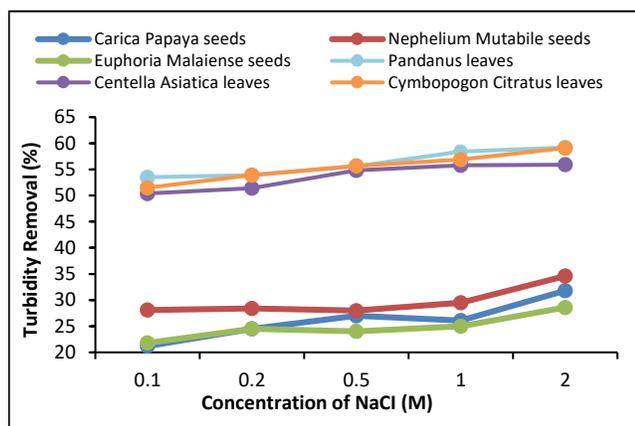


Figure 1: (a) Turbidity removal and (b) Coagulation activity of natural coagulants using deionised water

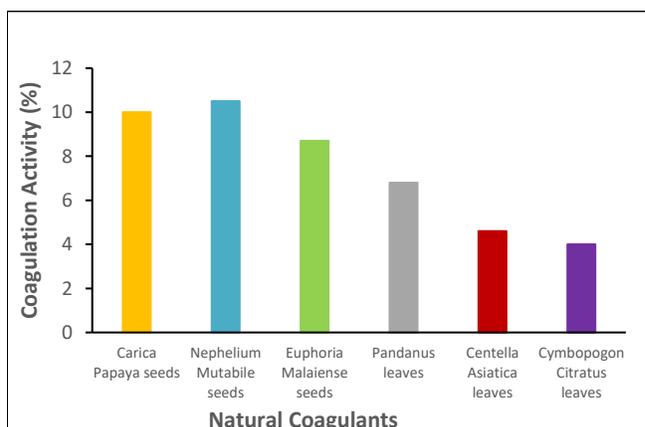
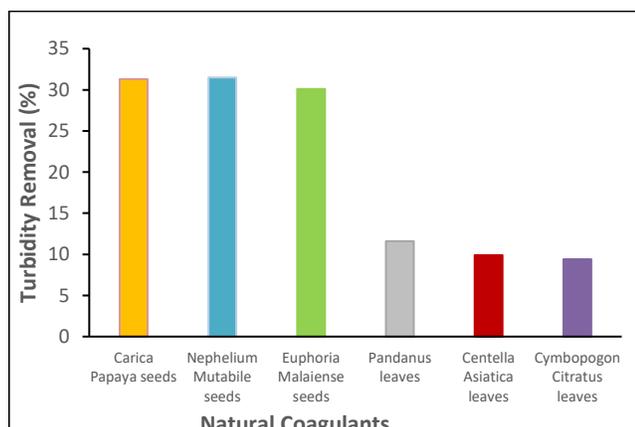
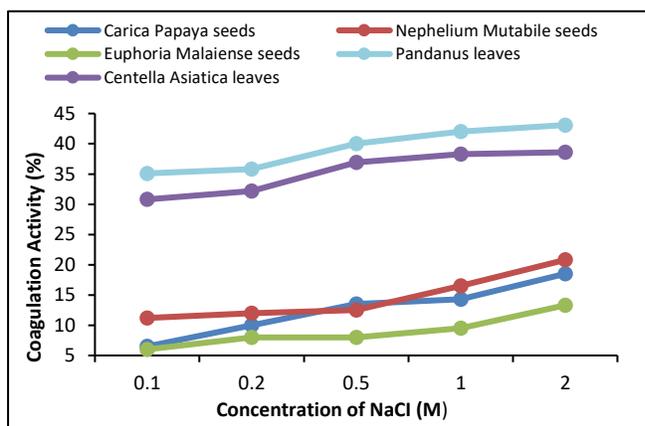


Figure 2: (a) Turbidity Removal and (b) Coagulation Activity of natural coagulants using NaCl solution

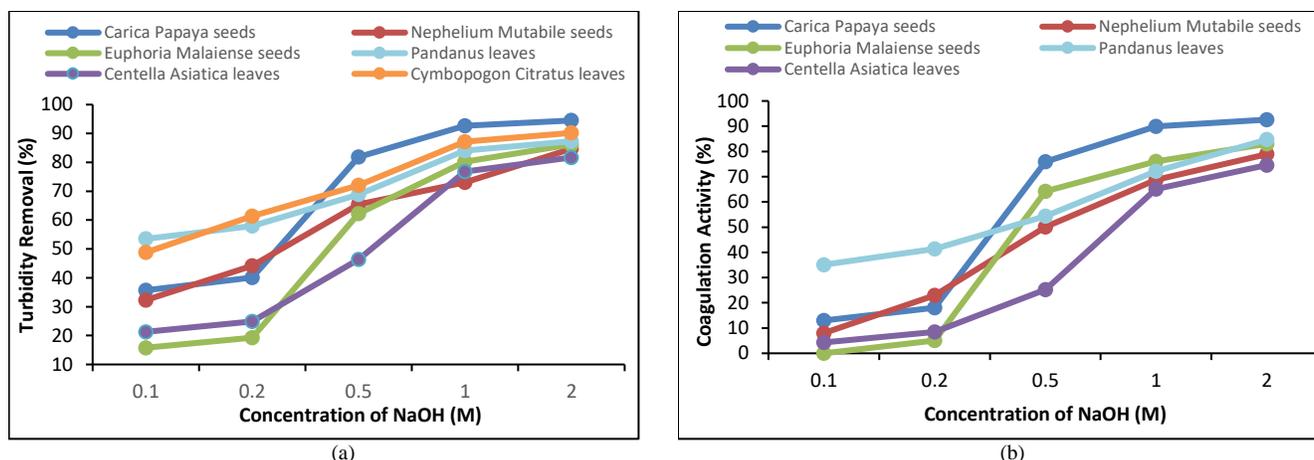


Figure 3: (a) Turbidity Removal and (b) Coagulation Activity of natural coagulants using NaOH solution

The optimum concentration obtained by the NaOH solution was similar to NaCl at 2.0 M. Based on Figure 3(a), among all the natural coagulants, the *Carica papaya* seeds had the highest turbidity removal of 94.5%. Meanwhile, the turbidity removal achieved by the *Cymbopogon citratus* leaves, *Pandanus* leaves, *Euphoria malaiense* seeds, *Nephelium mutabile* seeds, and *Centella asiatica* leaves were 90.2%, 87.3%, 86.2%, 84.7%, and 81.7%, respectively. The results in Figure 3(a) show similar trends with Figure 3(b), where the *Carica papaya* seeds also had the highest coagulation activity of 92.6%, followed by the *Cymbopogon citratus* leaves, *Pandanus* leaves, *Euphoria malaiense* seeds, *Nephelium mutabile* seeds, and *Centella asiatica* leaves with coagulation activities of 88.9%, 84.7%, 83%, 78.9%, and 74.6%, respectively. All the natural coagulants in the NaOH solution show high performance when compared to the other solutions of NaCl and deionised water. This is due to the high efficiency of the NaOH solution in extracting more coagulant agents from these plant-based natural coagulants, which resulted in a high coagulation activity, thus leading to a higher turbidity removal. Therefore, it can be concluded that the NaOH solution is the best solvent required to extract natural coagulants in water treatment.

3.4 Effect of Dosage on Natural Coagulants

Coagulation efficiency can be determined by considering several factors, including coagulation dosage (22). To achieve excellent results in water treatment, it is important to determine the optimum conditions during the coagulation process. This is because a high coagulation dosage may inhibit the high removal performance of coagulants in water treatment (19). Figure 4(a) and Figure 4(b) show the performance of each natural coagulant, under various dosages. The pH was kept constant at 7.5.

Based on Figure 4(a), the 130mg/L dosage was selected as the optimum coagulant dosage as it showed the highest turbidity removal. At the optimum dosage, *Carica papaya* seeds had the highest turbidity removal of 95.5%, while the *Cymbopogon citratus* leaves, *Euphoria malaiense* seeds, *Nephelium mutabile* seeds, *Pandanus* leaves and *Centella asiatica* leaves had turbidity removals of 93.4%, 93%, 92%, 90.5%, and 90.2%, respectively. It is clearly shown in Figure 4(a) that the coagulant dosage significantly affects the turbidity removal efficiencies. The removal performance for all the natural coagulants increase with

an increment in the dosage. In contrast, it is observed that a high coagulant dosage has a slightly adverse effect towards removal performance. In the case of the *Euphoria malaiense* seeds, the removal performance decreased from 91.7% to 90.4% as the dosage increased from 70 to 90 mg/L. The performance of natural coagulants shown in Figure 4(b) are associated with the performance shown in Figure 4(a). It is clearly seen that the *Carica papaya* seeds has a higher coagulation activity of 95.4% while the *Centella asiatica* leaves has the lowest coagulation activity of 89%.

3.5 Effect of pH on Natural Coagulants

The pH is another important factor for coagulation efficiency. During the coagulation process, the surface charge of coagulants may be affected by the pH (22). Coagulants with a low surface charge might cause the slow growth of flocs particle, thus leading to low removal performance in water treatment. Therefore, it is crucial to conduct jar tests to determine the optimum pH in treating turbid water. Figure 5(a) shows the effect of varying pH on turbidity removal by the natural coagulants while Figure 5(b) shows the pattern of coagulation activity occurring during the experimental work at various pH values. The pH values varied from 2, 4, 7.5, 10, to 12, at constant dosage (130 mg/L). Based on the figures, the best pH value to remove turbidity was at pH 7.5, while the lowest percentage removal of turbidity was at pH 12. All the natural coagulants showed removal trends where the percentage of removal increased from pH 2 and rapidly decreased after pH 7.5. Based on Figure 5(a), at optimum pH 7.5, the *Carica papaya* seeds contributed the highest turbidity removal of 95.6%, followed by the *Nephelium mutabile* seeds, *Cymbopogon citratus* leaves, *Euphoria malaiense* seeds, *Pandanus* leaves and *Centella asiatica* leaves with removal rates of 94.1%, 93%, 91.3%, 89.2%, and 87%, respectively. Figure 5(b) demonstrates that the coagulation activities of all natural coagulants showed positive effects when the pH value is increased. It is noticeable that most coagulation occurred at pH 7.5, compared to other pH value. The highest coagulation activity in Figure 5(b) is also shown by the *Carica papaya* seeds with a value of 96.7%. In contrast, the *Centella asiatica* leaves had the lowest coagulation activity of 86.1%. Of all the pH values investigated, pH 7.5 was the optimum for all the natural coagulants as both results showed the highest turbidity removal and coagulation activity

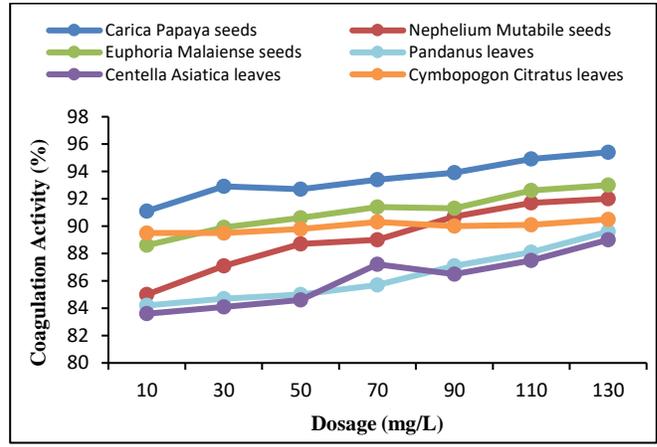
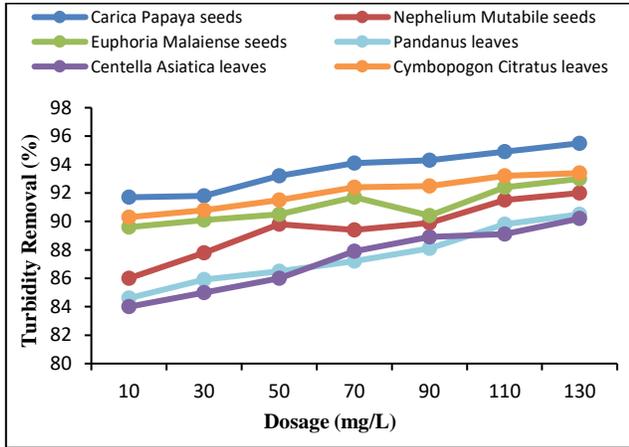


Figure 4: Effect of various coagulant dosage on (a) Turbidity Removal and (b) Coagulation Activity of different natural coagulants

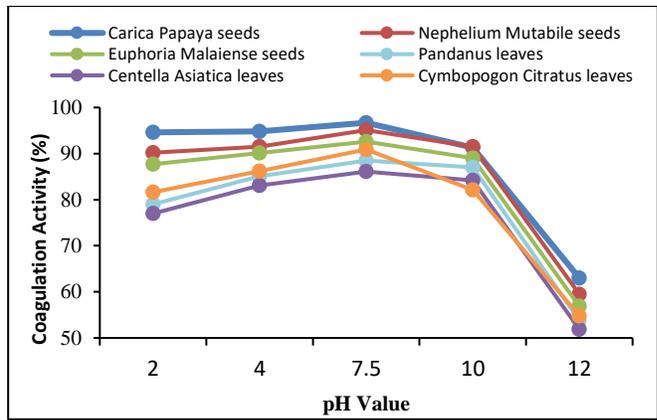
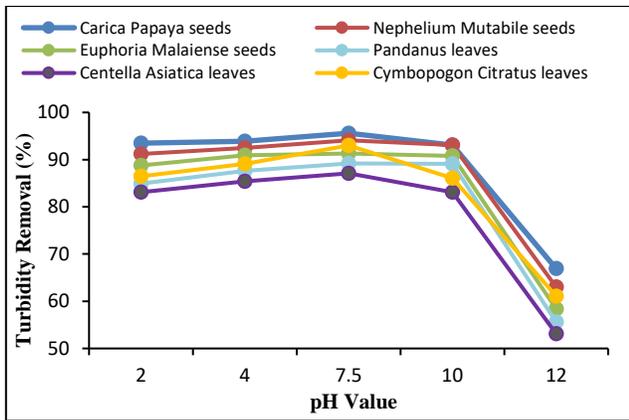


Figure 5: Effect of various pH on (a) Turbidity Removal and (b) Coagulation Activity of different natural coagulants

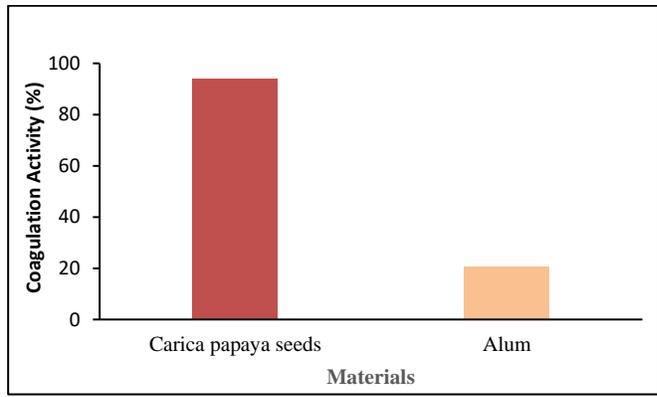
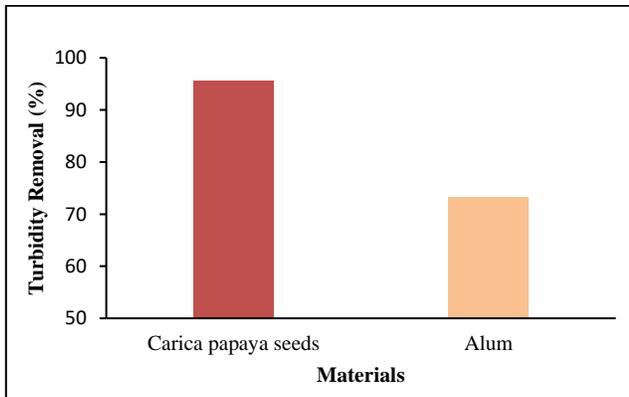


Figure 6: Comparison performance of (a) Turbidity Removal and (b) Coagulation Activity between *Carica papaya* seeds and Alum

3.6 Potential Use of Natural Coagulant in Water Treatment

This study further compared the performance of a natural coagulants with a chemical coagulant. The potential use of natural coagulants in water treatment can be determined through this comparison. In the present study, the *Carica papaya* seeds was selected as the best coagulant as it produced the highest turbidity

removal and coagulation activity. Figure 6(a) and Figure 6(b) show the performance comparison between natural coagulant (*Carica papaya* seeds) and chemical coagulant (alum). The test was carried out at optimum conditions (coagulation dosage of 130 mg/L and pH 7.5). The results showed that the natural coagulant acts as a better coagulant agent compared to alum.

Based on Figure 6(a), the *Carica papaya* seeds managed to remove almost 95.5% of turbidity, compared to alum which could only remove water turbidity by up to 73%. Based on Figure 3(b), the coagulation activity of alum was only 20.8%, lower than the *Carica papaya* seeds which exhibited a high coagulation activity of 94%. Thus, based on the results, it can be proven that *Carica papaya* seeds (natural coagulant) has a tremendous potential to treat polluted water and is superior to alum (chemical coagulant).

4 Conclusions

In terms of yield properties, the *Nephelium mutabile* seeds are sufficient to be used as natural coagulants. This is due to the *Nephelium mutabile* seeds having the lowest moisture content of 20.5% which resulted in a greater yield. All the natural coagulants react the best in a sodium hydroxide solution (NaOH) compared to deionised water and sodium chloride solution (NaCl). The NaOH solution achieved the optimum concentration at 2 M which contributed to higher turbidity removal and coagulation activity in the *Carica papaya* seeds at 94.5% and 92.6%, respectively. At optimum condition (130 mg/L dosage and pH 7.5), the *Carica papaya* seeds appeared to be the best coagulant with the highest turbidity removal of 95.6% and coagulation activity of 94%. This study also proved that natural coagulants have a higher efficiency in treating turbid water compared to chemical coagulants. At optimum condition, the *Carica papaya* seeds contributed to excellent turbidity removal of 95.6% and coagulation activity of 94%, while, alum only achieved 73% and 20.8% of turbidity removal and coagulation activity, respectively. Overall, fruit seed and plant leaf-based natural coagulants have a high potential and could be commercialised as coagulants to replace chemical coagulants in water treatment.

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Ethical issue

Authors are aware of all the publication ethics and adhere to the publishing requirement. The manuscript is an original work and not under consideration for publication elsewhere and has not been published elsewhere in the same form, in English or in any other language. The manuscript does not involve any studies related to human participants or animals performed by any of the authors.

Competing interests

The authors declare that there is no conflict of interest that could have appeared to influence the work reported in the paper.

Authors' contribution

All authors have contributed in writing, gathering experimental data and analysis. All the authors have worked together to complete the research and mutually agree to submit the paper for publication.

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