ABSTRACT

Coagulation process using low dosage of plant-based coagulant to remove high chemical oxygen demand (COD) and turbidity is important for water treatment. This study presents the treatment of pond water using plant-based pineapple leaf coagulant to achieve high COD and turbidity removal. The coagulation was performed using a jar test experiment of pond water at different pH followed by different dosages of pineapple leaf coagulant. It was found that the highest COD and turbidity removal ranged between 94.1 – 94.6 % and 88.3 – 88.4 % at pH 8 respectively, using low dosage (50 mg L\(^{-1}\)) of pineapple leaf coagulant. The final COD and turbidity values ranged between 7.3 – 8.0 mg L\(^{-1}\) and 17.7 – 17.8 formazin turbidity unit (FTU) respectively, which are lower compared with results from other studies that used high dosage coagulants. Moreover, the final pH, COD, turbidity, dissolved oxygen (DO), and total suspended solid (TSS) values of the treated pond water were below the standard limits set by the National Water Quality Standards for Malaysia (NWQSM) class IIB, which represents water bodies suitable for recreational use with body contact (DOE, 2016). Therefore, it is expected that the newly-formulated waste utilisation of pineapple leaf coagulant can reduce the usage of chemical coagulants and can further be used for different types of water.
Contribution/ Originality: This study contributes to the newly-formulated environmental friendly coagulant from pineapple leaf. The findings of this study are beneficial for the water industry to achieve optimum removal of pH, COD, turbidity, DO, and TSS values of treated water using this coagulant to ensure full compliance with the standards and preserving the environment.

1. INTRODUCTION

World population is experiencing a continuous growth and is estimated to be approximately 7.4 billion in 2016 \([1]\). Due to the overgrown population, wastewater production will also increase significantly and it will be channeled either directly or indirectly into water sources such as seas, rivers, lakes, or ponds \([2, 3]\). Among all water sources available, ponds and lakes are expected to receive the worst impacts because of their size and static water properties \([4]\). Due to their small size, ponds require better water treatment by aerobic, anaerobic, or coagulation method as they contain organic and inorganic matters \([5]\).

Coagulation treatment has been widely studied as an alternative method to treat polluted water \([6]\). Coagulation treatment is a turbidity removal process in water by combining small particles into flocs using chemical or natural coagulants. However, chemical coagulants have long been associated with high procurement costs as well as having detrimental effects on human health and environment \([7]\). Natural coagulants have been found to replace chemical coagulants due to their abundance and safeness. Many studies have been carried out using plant-based coagulants such as dragon fruits, banana stems, cacti, acorns and chestnuts, papaya peels, *Jatropha curcas* seeds and *Moringa oleifera* \([6-12]\). This technology is not only low-cost and simple, but also has several advantages such as low initial cost, simplicity of design, ease of operation, insensitivity to toxic substances, and complete removal of pollutants even from dilute solutions \([13]\). The effectiveness of these coagulants depends on a few parameters such as pH and dosage in order to achieve high COD and turbidity removal efficiency. Dosage is an essential factor affecting the adsorption process as it shows the adsorption capacity of the coagulants \([13]\). For example, lead-contaminated water requires 100 mg L\(^{-1}\) of papaya peels to obtain the highest turbidity removal \([8, 14]\).

Pineapple leaf is one of the alternative plant-based coagulants for water treatment. Pineapple, which is also known as *Ananas comosus* and belongs to the *Bromeliaceae* sp., is a perennial plant that ranked third on the list of the most important tropical fruits in the world. Pineapple leaf, which consists of cellulose, hemicellulose and lignin, can provide good thermal and noise insulation \([13, 15]\). Pineapple leaf showed tremendous pollutant removal of heavy metal \([16]\). The decolorization of crystal violet (CV) dyes from textile effluents using pineapple leaf powder (PLP) based on absorption capabilities has also been investigated \([17]\). However, studies on treatment of polluted pond water to achieve high COD and turbidity removal with low dosage of coagulants are still limited. Moreover, the final treatment of polluted pond water to achieve the standard discharge limit set by the authorities has not been carried out. Thus, this study proposes the treatment of pond water using low dosage of plant-based pineapple leaf coagulant to achieve high COD and turbidity removal to meet the standard discharge limit set by authorities for pond water quality which suitable for recreational use.

2. MATERIALS AND METHODS

2.1. Sample Preparation

Samples of raw pond water were obtained from a pond in Kota Samarahan area, Sarawak, Malaysia. The water was collected at 3 different sampling points i.e. side, middle, and bottom of the pond. All samples were preserved and stored at temperature less than 4 °C prior to use. This step was important in order to prevent any biodegradation due to microbial activity which may affect the result of the experiment. pH, total suspended solid (TSS), chemical oxygen demand (COD), turbidity, dissolved oxygen (DO) and biological oxygen demand (BOD) of
the samples were characterised and analysed based on Standard Methods for the Examination of Water and Wastewater [18].

2.2. Preparation of Pineapple Leaf Powder

Pineapple leaves were collected from a farmyard Padawan area, Kuching, Sarawak. The leaves were rinsed with tap water to remove dust, dirt, soil, and any unwanted particles [18]. They were dried in an oven at temperatures ranging between 50 and 80 °C until the moisture content decreased to below 10 %. Dried leaves were chopped into smaller pieces approximately 1 – 2 cm. The cut leaves were then ground into powder form where the sizes ranged between 1 and 2 mm. Then, the pineapple leaf powder was sieved to separate the larger particles from the powder to ensure that the coagulant readily coagulates with the pollutants. This preparation was conducted according to Idris, et al. [7].

The coagulation experiments were performed using a jar test based on the method performed by Idris, et al. [7]. 500 mL of pond water was poured into each of 6 beakers. The stirrer was switched on and operated at the ‘flash mix’ speed of approximately 120 rpm for 1 min after the addition of the coagulant. The stirrer speed was reduced to slow mixing (30 rpm) for 20 min and the paddles were withdrawn for a sedimentation process of 30 min. The supernatants were analyzed for pH, COD, and turbidity removal. The effect of pH was studied in the range of 2 – 10 and the effect of pineapple leaf dosage was tested in the range of 15 – 150 mg L⁻¹. The experiments were repeated up to 3 times to ensure reproducibility of the result and the adjustment of pH for the pond water samples was controlled by addition of H₂SO₄ and NaOH.

2.3. Analytical Method

The COD of the raw and treated pond water were determined by HACH COD (DR 2000) analyzer in accordance with HACH method (HACH, 1997). The TSS was determined based on Idris, et al. [7]. Turbidity measurement was carried out using the HANNA Model (HI 93703) microprocessor turbidity meter with measurement in FTU. The DO content was determined by using DO meter (Eutech, DO 700). The Biological Oxygen Demand (BOD) and Total Organic Compound (TOC) content were determined based on Standard Methods for the Examination of Water and Wastewater [18].

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Raw pond water</th>
<th>Tasik Biru lake water in Sarawak²</th>
<th>NWQSM class IIB²</th>
</tr>
</thead>
<tbody>
<tr>
<td>pH</td>
<td>7.21</td>
<td>7.76</td>
<td>6 – 9</td>
</tr>
<tr>
<td>TSS (mg L⁻¹)</td>
<td>96.67</td>
<td>1.40</td>
<td>50</td>
</tr>
<tr>
<td>COD (mg L⁻¹)</td>
<td>136</td>
<td>8.22</td>
<td>25</td>
</tr>
<tr>
<td>Turbidity (FTU)</td>
<td>152</td>
<td>-</td>
<td>50</td>
</tr>
<tr>
<td>DO (mg L⁻¹)</td>
<td>6.10</td>
<td>5.45</td>
<td>5 – 7</td>
</tr>
<tr>
<td>BOD (mg L⁻¹)</td>
<td>8</td>
<td>1.42</td>
<td>3</td>
</tr>
</tbody>
</table>

Note: ¹NREB [19], ²DOE [20].

3. RESULTS AND DISCUSSIONS

Table 1 shows the characteristics of raw pond water in Kota Samarahan area, Sarawak prior to the jar test coagulation experiment in comparison to Tasik Biru lake water in Sarawak. The lake water quality was used as a benchmark to compare the quality of pond water. It was obvious that the pond water located in Kota Samarahan area, Sarawak was slightly polluted compared to Tasik Biru lake water and NWQSM class IIB [20]. Although the pH and DO values of the pond water were almost similar to Tasik Biru lake water and within the standards limit of NWQSM, the values of COD, TSS, BOD, and turbidity were still high, which indicated that it would require further treatment.
3.1. Effect of Pineapple Leaf Coagulant on COD and Turbidity Removal at Different pH

The pH of pond water was determined by conducting the experiment with the addition of a predetermined fixed dosage (25 mg L⁻¹). The dosage value was selected based on a preliminary experiment on dosage effectiveness of pineapple leaf coagulant. COD and turbidity were monitored in order to determine the appropriate pH of pond water based on run 1 and run 2 to ensure reproducibility of the results.

Figure 1(a) and (b) show the effect of pineapple leaf on removal of COD and turbidity of pond water at different pH and fixed dosage (25 mg L⁻¹). It showed that the surface charge of pineapple leaf is strongly related to the pH of the solution. The pineapple leaf mainly consists of polar functional groups such as hydroxyl (-OH) and carbonyl (-C=O & -NH₂) groups that might be involved in coagulation process [13]. The highest percentage of COD removal Figure 1.a was found to range between 55.2 and 63.7 % and turbidity removal Figure 1.b was found to be more than 90.0 %, both at pH 2. At a pH lower than 4, anionic coagulant is coil up cationic pond water just like nonionic coagulant due to the amide groups (found in pineapple leaf) being replaced by the more inert carboxylate groups (at low pH), which reduced the number of hydrogen bonding sites available in the coagulant (pineapple leaf) [21]. This result is almost similar with Shak and Wu [22], who obtained the highest removal using Cassia obtusifolia seed gum coagulant at pH 3. This is a quite similar observation with Abidin, et al. [9], where the highest turbidity removal percentage was more than 99.0 % at pH 3 by using Jatropha curcas seeds.

Meanwhile, the lowest percentage of COD removal obtained ranged between 15.6 and 23.3 % at pH 6. This was due to the slight activity of anionic coagulant (pineapple leaf) on cationic pond water. Nevertheless, at pH 7 and 8 (alkaline based), the removal of COD and turbidity were found to increase where the highest COD removal was found to range between 57.8 and 59.0 %, and turbidity removal was found to be more than 88.0 %, both at pH 8. As pH increases, the carboxylate groups of anionic coagulant (pineapple leaf) are ionized Pillai [21], creating a strong electrostatic attraction between the cationic pond water (positively charged) and the pineapple leaf powder surface (negatively charged). This study also showed that the treatment of pond water using pineapple leaf performed better in both acidic and alkaline conditions. This study was in agreement with Neupane, et al. [17], where the maximum adsorption of textile effluents using pineapple leaf powder (PLP) on adsorbents was found at pH 8. Thus, for further dosage analysis in this study, pH 8 was selected since pH 2 (acidic condition) required additional H₂SO₄ treatment, which further increased the cost of treatment.
3.2. Effect of Different Dosages of Pineapple Leaf on COD and Turbidity Removal

The dosage of pineapple leaf was determined by conducting the experiment at a constant pH (pH 8). Figure 2 (a) and (b) show the effect of pineapple leaf on removal of COD and turbidity of pond water respectively at different dosages. The highest COD and turbidity removal ranged between 94.1 – 94.6 and 88.3 – 88.4 % respectively, using 50 mg L⁻¹ dosage of pineapple leaf coagulant. The final COD and turbidity values ranged between 7.33 – 8.00 mg L⁻¹ and 17.66 – 17.81 FTU respectively. The removal percentage decreased as dosage increased (>100 mg L⁻¹) due to an increase the organic load in water, thus resulting in the possibility for undesired and increased microbial activity. Subsequently, it contributes to the increase of COD in an aqueous solution [7]. The treatment of polluted pond water in Kota Samarahan area, Sarawak was successfully carried out using pineapple leaf. The final concentrations of COD and turbidity values were lower compared to the regulatory standards, making the pineapple leaf suitable to be used as a plant-based coagulant for applications in wastewater treatment.
3.3. Comparison Studies

Table 2 shows the comparison of plant-based coagulant dosage treatment using pineapple leaf with other studies. The removal percentage of COD and turbidity obtained in this study ranged between 94.1 – 94.6 and 88.3 – 88.4 %, which are considered high as compared to Kakoi, et al. [23] and Choy, et al. [24], both in acidic solution. As compared to Zhang, et al. [12], in alkaline solution, the COD and turbidity percentage removal in this study were also higher, although a similar dosage was used. Generally, the dosage of pineapple leaf used in this study was relatively low (50 mg L⁻¹) as compared to other studies which produced lower COD and turbidity removal. Moreover, the final pH, COD, turbidity, DO, and TSS values of the treated pond water Table 3 were below the standard limits set by the NWQSM class IIB [20]. Nevertheless, the final value of BOD was found to increase from 8 to 18 mg L⁻¹ due to the organic nature of pineapple leaf coagulant, which can increase microbial activity.

Table 2. Comparison of plant-based coagulant dosage treatment using pineapple leaf with other studies.

<table>
<thead>
<tr>
<th>Types of wastewater</th>
<th>Coagulants</th>
<th>Optimum dosage</th>
<th>Optimum pH</th>
<th>COD Removal (%)</th>
<th>Turbidity Removal (%)</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>Synthetic turbid water</td>
<td>Rice starch</td>
<td>120 mg L⁻¹</td>
<td>4</td>
<td>-</td>
<td>50</td>
<td>Choy, et al. [24]</td>
</tr>
<tr>
<td>Sewage water</td>
<td>Cactus</td>
<td>50 mg L⁻¹</td>
<td>7</td>
<td>50 – 60</td>
<td>80 - 85</td>
<td>Zhang, et al. [12]</td>
</tr>
<tr>
<td>River water</td>
<td>Banana pith</td>
<td>0.1 kg m⁻³</td>
<td>4</td>
<td>54.37</td>
<td>98.6</td>
<td>Kakoi, et al. [23]</td>
</tr>
<tr>
<td>Textile effluent</td>
<td>Pineapple leaf</td>
<td>1 g L⁻¹</td>
<td>8</td>
<td>-</td>
<td>-</td>
<td>Neupane, et al. [17]</td>
</tr>
<tr>
<td>Pond water</td>
<td>Pineapple leaf</td>
<td>50 mg L⁻¹</td>
<td>8</td>
<td>94.12 – 94.61</td>
<td>88.28 – 88.38</td>
<td>This study</td>
</tr>
</tbody>
</table>

Table 3. Comparison of treated pond water in Kota Samarahan area, Sarawak with standard limits.

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Raw pond water</th>
<th>Treated pond water</th>
<th>NWQSM class IIB²</th>
</tr>
</thead>
<tbody>
<tr>
<td>pH</td>
<td>7.21</td>
<td>8.3</td>
<td>6 – 9</td>
</tr>
<tr>
<td>COD (mg L⁻¹)</td>
<td>136</td>
<td>7.3 – 8.0</td>
<td>25</td>
</tr>
<tr>
<td>Turbidity (FTU)</td>
<td>152</td>
<td>17.7 – 17.8</td>
<td>50</td>
</tr>
<tr>
<td>TSS (mg L⁻¹)</td>
<td>96.67</td>
<td>32</td>
<td>50</td>
</tr>
<tr>
<td>DO (mg L⁻¹)</td>
<td>6.1</td>
<td>7.3</td>
<td>5.00 – 7.00</td>
</tr>
<tr>
<td>BOD (mg L⁻¹)</td>
<td>8</td>
<td>18</td>
<td>3</td>
</tr>
</tbody>
</table>

Note:²DOE [20].
4. CONCLUSION

A study on the effectiveness of pineapple leaf as a plant-based coagulant on pond water was successfully carried out. It showed a significant COD and turbidity percentage removal which ranged between 94.1 – 94.6 and 88.3 – 88.4 % respectively at pH 8 using low dosage of pineapple leaf coagulant (50 mg L\(^{-1}\)). The final values of COD and turbidity were found to be in the range of 7.3 – 8.0 mg L\(^{-1}\) and 17.66 – 17.81 FTU respectively. The results found higher percentage of COD and turbidity removal compared to other studies using different plant-based coagulants with higher or similar dosages. Moreover, the final pH, COD, turbidity, DO, and TSS values of the treated pond water were below the standard limits set by the NWQSM class IIB, which represents water bodies suitable for recreational use with body contact \(^{[20]}\). The newly-studied plant-based coagulant from pineapple leaf biomass successfully reduced COD and turbidity of treated pond water to achieve water pond quality, and is expected to be suitable for treatment of other types of wastewater from secondary industrial plants.

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**Competing Interests:** The authors declare that they have no competing interests.

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