Utilization of Moringa Seed Powder (*Moringa Oleifera*) as a Natural Coagulant for Reducing Pollution Parameters in Tofu Wastewater

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Abstract. The wastewater generated from the tofu industry often contains elevated levels of COD and TSS pollutants. One effective method for treating this wastewater is through the coagulation-flocculation process. Moringa seeds, recognized as a biomass with natural coagulant properties, contain a bioactive compound known as 4αL-rhamnosylxy-benzyl-isothiocyanate, capable of adsorbing particles present in wastewater. The objective of this study is to assess the efficiency of reducing pollutant parameters (COD, TSS, and Turbidity) in wastewater from the tofu industry by utilizing Moringa seeds as a natural coagulant. The research was conducted using a jar test apparatus with coagulation stirring at 100 rpm and flocculation at 40 rpm for 12 minutes. The study involved varying coagulant doses (2, 3, 4, 5, and 6 g/L of tofu wastewater) and coagulation stirring times (1, 3, and 5 minutes). Based on the results obtained, the optimal stirring duration and dose, as well as the efficiency of reducing each pollutant parameter, were determined. For COD, the optimal conditions were found to be 3 minutes stirring time with a dose of 4 g/L, resulting in an efficiency of 64.88%. Similarly, for TSS parameters, the optimum conditions were achieved with 3 minutes of stirring time and a dose of 4 g/L, yielding an efficiency of 52.71%. Lastly, for turbidity, the most effective conditions were observed with 3 minutes of stirring time and a dose of 4 g/L, with an efficiency of 58.85%.

Keywords: Coagulation, Flocculation, Moringa Seeds, Natural Coagulant, Tofu Wastewater.

1. Introduction

The tofu industry is among those that generate wastewater as a by-product, which holds the potential for environmental pollution if not adequately treated. One of methods to treat tofu wastewater is by using a membrane bioreactor (MBR). This method is capable of reducing COD by up to 90%. However, it has the disadvantage of high procurement costs for membrane modules and operational expenses, rendering it impractical for implementation in small-scale industries such as the tofu industry [1]. Another effective method for treating tofu wastewater is the coagulation-flocculation process. This method has proven effectiveness in reducing Total Suspended Solids (TSS) by up to 81% using aluminum sulfate coagulant (Al₂(SO₄)₃) [2], and Chemical Oxygen Demand (COD) by up to 81.8% using Poly Aluminum Chloride (PAC) coagulant [3]. The coagulation-flocculation process requires a coagulant to bind pollutant particles present in the water [4]. Synthetic coagulants are commonly employed by industries [5]. However, synthetic coagulants may lowering the water pH and generating sludge that may still contain chemicals, also posing a danger if accumulated in the environment [6]. Natural coagulants offer several advantages which are easily biodegradable, produce minimal sludge, are environmentally safe upon accumulation, and exhibit low toxicity [5].

*Moringa oleifera* holds significant importance as an herbal plant due to its vast array of medicinal and non-medicinal benefits. It has been discovered that bioactive constituents are present in every part of the plant [7]. The leaf and roots are consumed, but the seeds are discharged [8]. Moringa seeds are among the natural ingredients with the potential to serve as a natural coagulant due to their content of the active substance
4αL-rhamnosyloxy-benzyl-isothiocyanate, which enables them to absorb particles in wastewater [9]. Previous research has shown that Moringa seeds can effectively reduce pollutant parameters, including COD by 69.58%, TSS by 89.73%, and turbidity by 89.42% [10].

The use of natural coagulants to eliminate contaminants in tofu wastewater was explored over years ago. However, a comprehensive understanding of the efficacy of Moringa seeds as a natural coagulant in treating tofu wastewater remains incomplete. Therefore, the specific aim of this study was to assess the effectiveness of Moringa seed as a coagulant for the removal of COD, TSS, and turbidity from tofu wastewater. Additionally, coagulant evaluation via Fourier transform infrared (FTIR) spectroscopy was conducted to analyze the protein content in Moringa seed coagulants before and after the coagulation process. This experimental investigation marks the investigation into utilizing Moringa seed extract for tofu wastewater treatment.

2. Method

2.1 Materials Preparations

Moringa seeds are initially separated from the fruit and subsequently cleaned. These cleaned seeds undergo a drying process in an oven at 105°C for approximately 24 hours. Once dried, the Moringa seeds are pounded using a mortar and pestle. Subsequently, the ground Moringa seeds are filtered through a 100 mesh sieve. The tofu wastewater was collected from the outlet of a tofu industry located at Karang Joang sub-district. The procedure for water sampling adhered to the standards specified in SNI 6968.59:2008, which outlines methods for sampling wastewater through grab sampling. The initial tofu wastewater quality, including COD, TSS, and turbidity, was analyzed using standard methods for examining water and wastewater.

The tofu wastewater typically exhibits an acidic pH, which can impede the efficiency of the coagulation-flocculation process. Therefore, it is essential to optimize the pH of tofu wastewater samples to achieve the most effective pH value for the coagulation-flocculation process. The pH variations tested ranged from 5 to 9, achieved by the addition of strong acid (HCl) and strong base (NaOH) to the tofu wastewater. At this stage, a dosage of 3 g/L was utilized, accompanied by a coagulation stirring speed of 100 rpm for 3 minutes and a flocculation stirring speed of 40 rpm for 12 minutes. Subsequently, the mixture underwent sedimentation process for 60 minutes.

2.2 Experimental Method

The primary research was conducted utilizing a jar test apparatus in a batch system. A coagulation stirring speed of 100 rpm and a flocculation stirring speed of 40 rpm were applied for 12 minutes, followed by sedimentation for 60 minutes. The coagulant dosage ranges from 2 – 6 g/L while the time of rapid mixing time ranges from 1 – 5 minutes.

Subsequent to the sedimentation process, tests will be conducted to analyze parameters such as COD, TSS, and turbidity using standard water and wastewater examination methods. The volume of wastewater utilized in the experiments was 500 ml. The removal efficiency was calculated using equation (1).

\[
\text{Removal efficiency(\%)} = \frac{c_e}{c_o} \times 100\% \quad (1)
\]

where \(c_o\) represents the initial concentration of COD, TSS, and turbidity, and \(c_e\) denotes the final concentration of COD, TSS, and turbidity after the coagulation-flocculation process.
3. Result and Discussion

3.1 Characterization of Tofu Wastewater

Table 1 shows the effluent of the tofu wastewater treatment plant. According to Table 1, it is clear that the quality of tofu wastewater does not adhere to the standards stipulated by Minister of Environment Regulation No. 5 of 2014.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Concentration</th>
<th>Standard</th>
</tr>
</thead>
<tbody>
<tr>
<td>COD</td>
<td>3123.9</td>
<td>150 mg/L*</td>
</tr>
<tr>
<td>TSS</td>
<td>1958 mg/L</td>
<td>200 mg/L*</td>
</tr>
<tr>
<td>Turbidity</td>
<td>410 NTU</td>
<td>-</td>
</tr>
</tbody>
</table>

*) Minister of Environment Regulation No. 5 of 2014.

The elevated level of COD is attributed to the utilization of organic materials such as soybeans in the tofu manufacturing process. Tofu wastewater contains organic constituents, including protein (40-60%), fat (8-12%), and carbohydrates (25-50%) [11]. Additionally, the TSS concentration measures at 1958 mg/L, with a turbidity concentration of 410 NTU. The heightened TSS concentration is attributed to colloids originating from residual organic materials remaining from tofu production. Turbidity in tofu wastewater arises from residual water remaining after tofu soaking and soybean peeling, which retains substantial starch content [12]. Based on the outcomes of the initial characteristic analysis, a treatment process is imperative to mitigate pollutant parameters in the wastewater generated by the tofu industry.

3.2 pH Optimalization

The determination of the optimal pH value is based on achieving the maximum allowable turbidity parameter. The outcomes of the pH optimization process are illustrated in Figure 1.

Coagulation-flocculation processes conducted outside the optimum pH range may result in the failure of floc formation [13]. During pH optimization, the initial turbidity value was measured at 401 NTU with a pH of 5.12, utilizing a coagulant dose of 3 g/L and a rapid stirring duration of 3 minutes. The findings from the pH optimization experiment revealed that the optimal pH for Moringa seed powder coagulant was 9, leading to a significant reduction in turbidity concentration by up to 56%. This reduction in turbidity concentration suggests that at a pH of 9, the proteins present in Moringa seeds act as acids, generating H⁺ ions, which subsequently destabilize colloids in the wastewater, facilitating floc formation [14]. Conversely, under acidic pH conditions, there is a rise in turbidity concentration. This phenomenon occurs because in acidic environments, proteins tend to acquire protons from the wastewater, resulting in the production of OH⁻ ions. However, these ions are unable to destabilize and bind colloids effectively due to their identical negative charge, thereby leading to increased turbidity [15].
3.3 Effect of Dosage and Rapid Mixing Time on COD

Figure 2 illustrates that the optimal COD removal efficiency is achieved at 3 minutes with a dosage of 5 g/L, resulting in a removal efficiency of 64.88%.

The effectiveness of COD reduction increases from 24% to 64% as the dosage escalates from 2 g/L to 5 g/L. This decline in COD concentration can be attributed to the uniform distribution of cationic protein content from Moringa seeds, enabling it to bind with organic compounds present in the wastewater [16]. However, at dosages ranging from 5 g/L to 6 g/L, the removal efficiency decreases to 37.49% due to surpassing the optimum dosage level. Excessive dosages lead to the restabilization of colloid particles, creating repulsive forces between them as they share similar positive charges, consequently reducing the percentage of removal allowances [17].

Different outcomes were observed at rapid stirring durations of 1 minute and 5 minutes, resulting in varied COD concentrations. At 1 minute, it’s evident that the COD concentration increases with the rise in coagulant dosage from 2 g/L to 6 g/L due to the dissolution of organic compounds present in the coagulants into the wastewater [18]. The rise in COD concentration is primarily attributed to the residual oil content in Moringa seeds, which hasn’t completely dissipated. Additionally, the substantial presence of Dissolved Organic Carbon (DOC) in peeled Moringa seeds significantly contributes to the elevation of COD concentrations [19].

Conversely, at a rapid stirring duration of 5 minutes, an escalation in COD removal efficiency is observed across dosages ranging from 2 g/L to 6 g/L. This is attributed to the ability of coagulants to destabilize organic compounds and facilitate floc formation [16].

3.4 Effect of Dosage and Rapid Mixing Time of TSS

Figure 3 shows that the optimal TSS concentration removal was achieved at a rapid stirring duration of 3 minutes and a dosage of 4 g/L, yielding a TSS concentration removal efficiency of up to 52.71%.

Within the dosage range of 2 g/L to 4 g/L, the TSS removal efficiency increased from 10% to 52%. This improvement is attributed to the proteins contained in Moringa seeds, which, when dissolved in water, generate H ions. These ions play a crucial role in destabilizing negative particles in wastewater, promoting their aggregation and subsequent formation of flocs (E. Ningsih et al., 2018). However, at dosages ranging from 4 g/L to 6 g/L, there was a
decrease in efficiency from 52% to 25%. This decline is because doses exceeding the optimum limit can cause restabilization of colloid particles.

At 1 minute and 5 minutes of rapid stirring, there was an increase in TSS concentration. Within the dosage range of 2 g/L to 4 g/L, an increase in dosage corresponded to an increase in removal efficiency. This phenomenon occurs because the dosage falls within the optimal range, allowing the coagulant to effectively destabilize colloids in the wastewater [20]. However, at dosages ranging from 4 g/L to 6 g/L, the TSS concentration increased. This was due to exceeding the optimal dosage level, resulting in decreased removal efficiency [21]. Uneven distribution of coagulants prevents the formation of flocs, leading to the added dose creating new suspended particles [22]. The small size of the coagulant (0.147 mm) also contributes to the increase in TSS concentration, as smaller coagulants are more difficult to precipitate [23].

At a rapid stirring duration of 5 minutes, the TSS concentration increased because the rapid stirring time exceeded the optimum limit. In this condition, the formed flocs are smaller in size, making them difficult to settle from the wastewater and resulting in an increase in TSS concentration [24]. The small floc size results from the floc breaking due to the excessively long rapid stirring time, making it challenging for the flocs to settle from the wastewater and causing an increase in TSS concentration.

### 3.5 Effect of Dosage and Rapid Mixing Time on Turbidity

Figure 4 shows the effect of dosage and rapid mixing time on turbidity.

The optimal fast stirring duration for achieving the highest turbidity concentration removal was determined to be 3 minutes, with an optimal coagulant dose of 4 g/L, capable of removing turbidity concentrations by up to 58.85%. The efficiency of turbidity concentration removal within the dosage range of 2 g/L to 4 g/L ranged from 20% to 58%. This is attributed to the cationic protein content present in Moringa seeds, which can form hydrogen bonds with suspended particles in wastewater, leading to the destabilization of colloidal particles. Subsequently, these particles attract each other to form flocs [25]. However, at doses ranging from 4 g/L to 6 g/L, the efficiency of concentration removal decreased to 39%-36%. This decline in efficiency occurs due to surpassing the optimum dosage level, where all colloid particles have been destabilized, and the coagulant can no longer bind to colloid particles to form flocs, resulting in an increase in turbidity concentration [26].

At rapid stirring durations of 1 and 5 minutes, the obtained removal efficiency was suboptimal. This is attributed to the rapid stirring time exceeding the optimum limit, resulting in a lack of increase in floc size. Instead, the floc breaks down into small particles that are difficult to settle [27].
Based on Figure 4, the optimal turbidity concentration removal was achieved with a fast stirring time of 3 minutes and a dose of 4 g/L, resulting in a removal efficiency of up to 58.85%. This is due to the protein content found in Moringa seeds, which, when dissolved in water, has the ability to generate H⁺ ions. However, exceeding the optimal time limit and dosage can lead to the restabilization of colloidal particles, as resulted in stirring time of 5 minutes. This restabilization prevents the formation of flocs, thereby reducing the efficiency of removal. In restabilization zones, there was observed a deterioration in residual turbidity [28]. The turbidity increased during the floc regrowth process, leading to a decrease in removal efficiency. As for 1-minute stirring time, the removal efficiency is not optimal because it has a mixing time that is too short, so that reducing can floc formation.

3.6 FTIR Analysis
The FTIR analysis of Moringa seed coagulant before and after the coagulation-flocculation process is illustrated in Figure 5.

![Figure 5. FTIR Analysis](image)

The peak representing the N-H functional group confirms the presence of an amine group, observed at a wavenumber of 2926.20 cm⁻¹. Subsequently, vibrational transitions occur, forming the C=O functional group, indicating the presence of a carboxyl group at a wavenumber of 1704.19 cm⁻¹. These functional groups signify the presence of active functional groups within Moringa seeds, which are inherent to organic compounds [29].

Following the coagulation process, changes in the peak cluster are observed in Moringa seeds, with the emergence of O-H (hydroxyl) at a wavenumber of 3288.97 cm⁻¹ and C=C (alkene) at a wavenumber of 1637.64 cm⁻¹. The formation of hydroxyl groups is attributed to the alkaline condition of the wastewater, leading to the formation of hydroxyl groups in natural coagulants after the coagulation process [30]. However, certain functional groups are lost after the coagulation process, namely the amine (N-H) and carboxyl (C=O) groups. This is due to the reaction between the OH⁻ ions on the colloid surface and the positively charged groups of the coagulant derived from the amine and carboxyl groups [31].

4. Conclusion
The optimum dosage and rapid stirring duration for each parameter are as follows: for COD, a rapid stirring duration of 3 minutes and a dosage of 4 g/L; for TSS, a stirring duration of 3 minutes and a dosage of 4 g/L; and for turbidity, an agitation duration of 3
minutes and a dosage of 4 g/L. Furthermore, the optimum removal efficiency achieved for each parameter is 64.88% for COD, 52.71% for TSS, and 58.85% for turbidity.

5. Author’s Declaration

Author contributions and responsibilities - The authors made major contributions to the conception and design of the study. The authors took responsibility for data analysis, interpretation and discussion of results. The authors read and approved the final manuscript.

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6. References


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