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The Effect of pH on the Effectiveness of Tamarind and Moringa Seeds Biocoagulants in Tofu Wastewater Treatment

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Abstract: The wastewater generated by the tofu industry has the potential to cause serious environmental pollution if it is not properly treated before being discharged into environment. This wastewater contains high levels of organic and inorganic materials that can reduce water quality and harm aquatic ecosystems. One of the effective and environmentally friendly treatment methods that can be applied is the coagulation–flocculation process using natural materials as biocoagulants. This study aims to evaluate the effectiveness of Moringa seeds (Moringa oleifera Lam) and Tamarind seeds (Tamarindus indica L.) as natural biocoagulants in reducing several water quality parameters, including Total Suspended Solids (TSS), Total Dissolved Solids (TDS), Dissolved Oxygen (DO), and Ammonia in tofu wastewater. The variations of pH used in this study were 4, 6, and 8, with a biocoagulant dosage of 4 g/L. The treatment was carried out through a coagulation–flocculation process consisting of rapid mixing at 200 rpm for 2 minutes, followed by slow mixing at 20 rpm for 20 minutes, and a sedimentation period of 50 minutes. The results indicated that the optimum reduction of TSS was achieved at pH 4 using Tamarind seed biocoagulant, with a reduction efficiency of 72%. Meanwhile, the optimum Ammonia removal was obtained at pH 6 using Moringa seed biocoagulant, achieving a 22% reduction. The DO level increased by 81% at pH 4 using Tamarind seeds. However, the TDS parameter did not yet meet the quality standards set by Government Regulation of the Republic of Indonesia Number 22 of 2021. Overall, these findings demonstrated that pH variation plays a crucial role in influencing the performance of natural biocoagulants as sustainable and eco-friendly alternatives for tofu wastewater treatment.

Keywords: biocoagulant, *Moringa oleifera*, *Tamarindus indica*, pH variation, tofu wastewater

1. INTRODUCTION

The tofu industry is one of the sectors that has experienced rapid growth in Indonesia. In its production process, this industry generates both solid and liquid waste. The solid waste, known as tofu dregs, is the residue primarily produced from the soybean filtration and coagulation process. Meanwhile, the liquid waste is generated from various stages of production such as washing, boiling, pressing, and molding. This liquid waste contains a high concentration of organic materials and has a low pH, ranging from 4 to 5 (Anggraini, 2014).

Moringa seeds contain a high level of protein with cationic polyelectrolyte properties due to their positive charge, making them effective in interacting with negatively charged colloidal particles and accelerating the formation of flocs and sedimentation (Hidayat, 2016). According to Wijaya et al. (2024), Moringa seeds contain oil (40%), fat (34.7%), protein (32.18%), flavonoids (1.26%), and tannins (4.5%). The significant charge difference between the coagulant and the colloidal particles accelerates the coagulation process. Tamarind seed extract also contains active compounds such as proteins, tannins, and metal ions (Mg^{2+}, Fe^{3+}) that function as natural polyelectrolytes. These compounds accelerate floc formation through bridging and charge neutralization mechanisms (Martina et al., 2018). The protein content in Tamarind seeds (15 – 20%) can carry either a positive or negative charge depending on the pH, while tannins (20.2%) act as the main coagulant. Other compounds such as starch, gum, and albuminoids support the coagulation process by forming particle aggregates. Its essential oils also help reduce unpleasant odors (Wardani and Tuhu, 2017).

The liquid waste generated from tofu production contains high levels of organic matter (40–60% protein, 25 – 50% carbohydrates, and 10% fat). These organic compounds are difficult to degrade biologically (Bangun et al., 2013). The analysis of untreated tofu wastewater showed a pH value of 3.57, TDS of 1,320 mg/L, ammonia of 29.84 mg/L, and TSS of 4,450 mg/L. The pH value of 3.57 is below the quality standard range, according to the Government Regulation number 22 of 2021, which is in the range 6 – 9. According to Asmadi and Suharno (2012), the ideal pH of wastewater should be close to neutral to support biological activity. Tofu wastewater also contains organic substances and gases, such as carbon dioxide (CO_2), ammonia (NH_3), dissolved oxygen (O_2), and hydrogen sulfide (H_2S) (Pagoray et al., 2021). During anaerobic degradation, acidic compounds are formed, leading to a drop in pH. Therefore, pH control is crucial to ensure the wastewater treatment process runs effectively and does not cause negative environmental impacts. The study conducted by Haslinah (2020) showed that the addition of Moringa seed biocoagulant at a dosage of 4000 mg/L achieved a COD removal efficiency of 86.4% in tofu wastewater. Based on the research by Sari (2017), Moringa seed coagulant can work optimally in reducing pollutants at a pH range of 6–8, while Yusuf et al. (2022) reported that the addition of 4000 mg/L of Tamarind seed biocoagulant was able to remove 82% of BOD, 82% of COD, 78% of TSS, and 84% of turbidity, with an optimum pH of 4 in tofu wastewater.

Coagulation-flocculation is a physico-chemical method used to remove suspended solids and colloids through particle destabilization (coagulation) and aggregation into larger, settleable flocs (Sarwono et al., 2017). Coagulation uses positive ions from the coagulant to neutralize negatively charged colloidal particles, such as organic matter and microorganisms in the wastewater (Setyawati et al., 2018). The process begins with rapid mixing to evenly distribute the coagulant and form microflocs (Ekoputri et al., 2023). Flocculation is the subsequent stage, where microflocs combine to form larger, denser flocs through slow mixing (Syahputra et al., 2022). The effectiveness of this process is influenced by the type and dosage of coagulant, pH, wastewater characteristics, mixing speed and duration, settling time, and temperature (Haslinah, 2020).

This study aims to determine the effect of pH variation on the biocoagulation ability of Moringa and Tamarind seeds in reducing TDS, Ammonia, DO, and TSS parameters, as well as to observe pH changes in tofu wastewater. The study was conducted using two variations: the type of biocoagulant, in which Moringa and Tamarind seeds were used as biocoagulants, and the pH variation, which included pH values of 4, 6, and 8. The results of this study were compared with the quality standards based on Government Regulation of the Republic of Indonesia Number 22 of 2021 concerning National Water Quality Standards. Results of this study would give insight to an optimal coagulant type and pH condition for environment-friendly tofu wastewater treatment.

2. MATERIALS AND METHODS

This research was conducted at one of the tofu processing industries located in Samarinda City. The tofu wastewater samples were taken from Tofu Processing Industry X, situated on Gg. Margo Mulyo, Lempake, North Samarinda District, East Kalimantan. This location was chosen because it is situated very close to the Lempake Dam. The time of sampling liquid waste from tofu was on March 13, 2025 from 12:50 to 13:00 pm with cloudy weather condition. The study involved two types of variables: independent and dependent variables. The independent variables are coagulant variation (Moringa Seed and Tamarind Seed) and pH variation (4, 6, and 8), while the dependent variables are the water quality parameters: pH, TDS, TSS, DO, and ammonia. The variations used in this study are presented in Table 1.

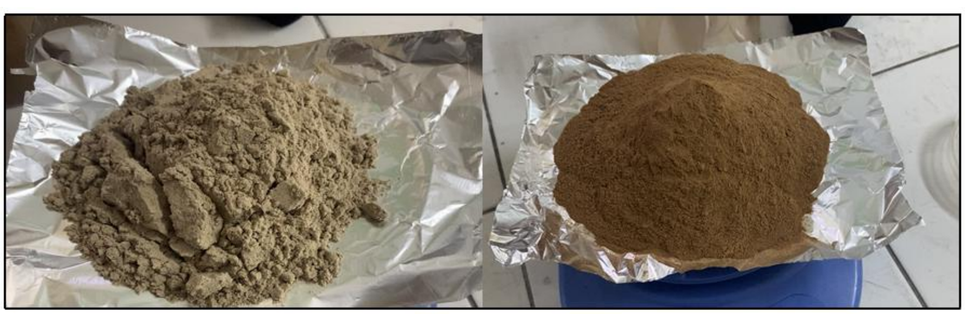
Tabel 1. Testing Variations

No.	Variation Code	Coagulant	pH Variation
1	K(1)	Moringa Seed	4
2	K(2)	Moringa Seed	6
3	K(3)	Moringa Seed	8
4	A(1)	Tamarind Seed	4
5	A(2)	Tamarind Seed	6
6	A(3)	Tamarind Seed	8

The preparation stage was carried out by preparing the necessary tools and materials. The equipments used in this study are jar test apparatus, sample bottles, blender, oven, beaker glass, 100 mesh sieve, analytical balance, pH meter, spray bottle, mortar and pestle, spatula, measuring cylinder, watch glass, glass funnel, desiccator, camera, and Imhoff cone. The materials used in this study were tofu wastewater, distilled water, Moringa seed. and Tamarind seed biocoagulants, stationery, label paper, and 1 M NaOH solution.

The sample collection of tofu wastewater used grab sampling method. The samples were being directly taken from the outlet channel of Tofu Industry X. The samples were collected in a clean container with a capacity of 50 liters, referring to the Indonesian National Standard (SNI) 6989.59:2008 concerning the Method for Sampling Wastewater.

The preparation of the biocoagulants started by peeling the Moringa and Tamarind seeds. Then, all seeds were grounded into a fine powder and sieved using a 100-mesh sieve. The sieved powder was dried in an oven at 105° C for 120 minutes until the moisture content reached approximately 10%, then stored in a closed container to prevent dust contamination (Harahap, 2023).



(a) **(b)**

Figure 1. (a) Moringa Seed Powder and (b) Tamarind Seed Powder

Figure 1 shows the result of processing seeds into coagulant powder. Figure 1(a). is Moringa seed coagulant and Figure 1(b). is Tamarind seed coagulant powder. The testing stage began with determining the initial pH value of the raw wastewater. A 1000 mL beaker glass containing 1000 mL of tofu wastewater sample was prepared, and the sample pH was adjusted to 4, 6, and 8 by adding a NaOH solution (Yusuf et al., 2020). The testing procedure started by preparing 1 liter of wastewater sample adjusted to pH levels of 4, 6, and 8. Then, 4 grams of Moringa seed and Tamarind seed biocoagulants were added to each sample. The mixture was stirred using a jar test apparatus by rapid mixing at 200 rpm for 2 minutes, followed by slow mixing at 20 rpm for 20 minutes. Afterward, the samples were allowed to settle for 50 minutes before testing the parameters of pH, TSS, TDS, and ammonia.

3. RESULTS AND DISCUSSION

3.1. Initial Characteristic of Tofu Wastewater

The results of the initial parameter analysis on the tofu industry liquid waste samples are showed in Table 2. Based on Table 2, all parameters exceeded the quality standards. These results indicate that treatment of the tofu wastewater is necessary before being discharged into environment.

Table 2. Analysis Results of Tofu Liquid Waste Samples Before Processing

Parameter	Unit	Quality Standards	Results	Analysis Method	Status
pH	-	6 – 9	3.57	SNI 6989.11:2019	not satisfied
TDS	mg/L	1000	1320	SNI 6989.27:2019	not satisfied
Amonia	mg/L	0.1	29.84	SNI 6989.2:2019	not satisfied
TSS	mg/L	40	4450	IKM/IPLab/7.2.5	not satisfied
DO	mg/L	> 6	0.31	IKA/IPLab/6.4.13	not satisfied

3.2. pH Analysis Result

Based on Figure 2, the initial pH of the wastewater affects the change in pH after treatment using biocoagulants from Tamarind seeds (A) and Moringa seeds (K). At an initial pH of 4, the final pH decreased to 3.8 (K1) and 3.9 (A2); at pH 6, the final pH of A2 decreased to 5.46, while K2 remained stable at 6.01; and at pH 8, a more significant pH decrease occurred, namely 5.27 (A3) and 5.34 (K3). The optimum pH value was obtained in treatment A2 (Tamarind seed biocoagulant at pH 6), as the final pH showed almost no change, remaining at 6.01.

This finding is consistent with the study by Herawati et al. (2017), which showed that pH 6 does not cause a significant pH decrease during the coagulation-flocculation process using Moringa seeds, as this condition is close to the isoelectric point of the protein in Moringa seeds. The isoelectric point is the condition at which the coagulant carries a neutral charge; if the solution's pH is above the isoelectric point, the coagulant will be negatively charged, whereas if the pH is below the isoelectric point, the coagulant will be positively charged. Moringa seeds have an isoelectric point of $\geq$10 (Herawati et al., 2017).

The pH decrease observed at initial pH 4 and 8 is caused by the activity of proteins and tannins in Moringa seeds, which act as positive charged macromolecules and function as positively charged polyelectrolytes that release charges into the solution, thereby lowering the pH (Hak et al., 2017).

Compared to Government Regulation No. 22 of 2021, with allowable pH range of 6–9, the variation that has value of pH that still meets the standard is the K2 variation. The K2 variation used Moringa seeds at pH 6.00 and produced final pH of 6.01.

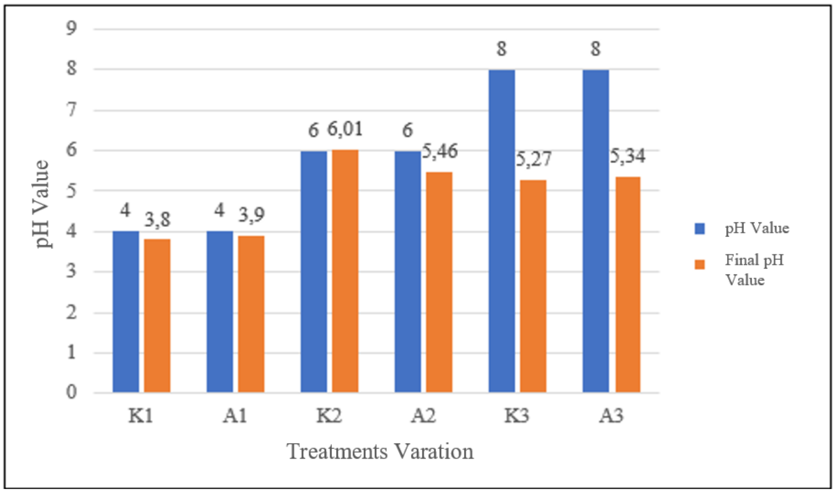


Figure 2. pH Analysis Results

3.3. TSS Analysis Result

Based on Figure 3, TSS test results at pH variations of 4, 6, and 8 show differences in the effectiveness of the two biocoagulants. At pH 4, A1 variation is the most optimal, with a final TSS of 1,25 mg/L and an efficiency of 71.9%. At pH 6, K2 variation is more effective (1,78 mg/L; 60%) compared to A2 (2,15 mg/L; 51.7%). At pH 8, K3 variation showed the best performance with a TSS of 1,76 mg/L and an efficiency of 60.4%, while A3 variation only achieved 2,14 mg/L and 52% efficiency. The ability of the organic coagulants from Tamarind and Moringa seeds to reduce TSS levels is attributed to their protein and tannin content (Figure 4). The organic coagulant from Tamarind seeds contains 15–20% protein and 20.2% tannin, while Moringa seeds contain 34.18% protein and 4.12% tannin. Compared to Indonesian Government Regulation No. 22 of 2021, the TSS values in all variations of pH and biocoagulant did not meet the water quality standards. The best reduction in TSS levels was obtained in the A1 variation, which used Tamarind seeds at pH 4, resulting in a final TSS concentration of 1,25 mg/L, while the maximum allowable limit according to the standard is 40 mg/L.

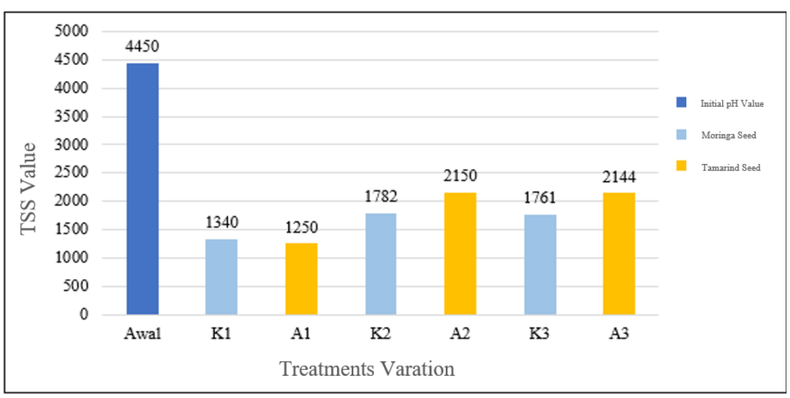


Figure 3. TSS Analysis Results

Figure 4 shows the reaction between tannins and proteins. In general, proteins contain amino (-NH_2) and carboxyl (-COOH) groups, while tannins are polyphenolic compounds with multiple reactive hydroxyl (-OH) groups capable of forming hydrogen bonds. When proteins in tofu wastewater come into contact with tannins, interactions occur between the -NH_2 groups of the proteins and the -OH groups of the tannins, forming protein–tannin complexes. These complexes can precipitate, thereby reducing organic compounds and lowering the TSS, BOD, and COD levels of the wastewater. Under acidic conditions around pH 4, tannins are more easily protonated, producing H^+ ions that strengthen the hydrogen bonds between the amino groups of proteins and the hydroxyl groups of tannins in the biocoagulant (Mawaddah, 2018).

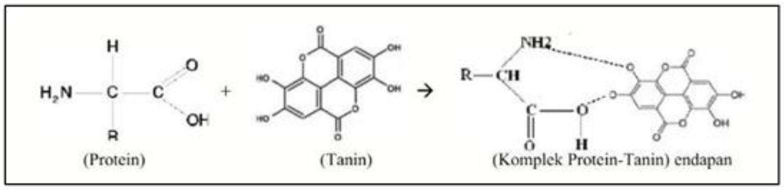


Figure 4. Binding of Tannins and Proteins (Mawaddah, 2018)

3.4. TDS Analysis Result

Based on Figure 5, the results of TDS testing at pH variations of 4, 6, and 8 show an increasing trend. Compared to Government Regulation No. 22 of 2021, TDS values in all variations of pH types did not meet the quality standards (maximum: 1000 mg/L). In treatments K1 and A1, similar values of 2340 mg/L were obtained, indicating the use of Moringa and Tamarind biocoagulants did not have a significant effect on reducing TDS in tofu wastewater. Higher increases were observed in treatments A2, K3, and A3, with TDS values ranging from 4580 to 5170 mg/L.

This increase in TDS values may be caused by the ionization of biocoagulant proteins under basic conditions, which releases dissolved minerals such as Ca^{2+}, K^+, and Mg^{2+} (Ndabirgengesere and Naraasiah, 1998). Additionally, an excessive coagulant dosage may also contribute to the rising value of TDS, as at concentrations exceeding the optimum dose, cationic ions from the biocoagulant are no longer effective in binding colloids. It can be identified by a leftover coagulant settling in a beaker glass, indicating it did not fully bind the suspended solids. Hermida et al. (2021) stated that overdosing can lead to the dominance of cations (from the biocoagulant) over impurity anions, thereby increasing turbidity and the concentration of dissolved substances in the water.

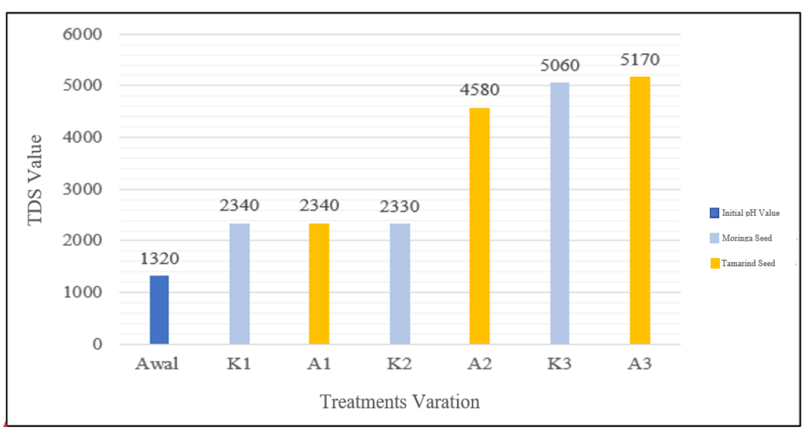


Figure 5. TDS Analysis Results

3.5. Ammonia Analysis Result

Based on Figure 6, the initial ammonia concentration in tofu wastewater was 29.84 mg/L. The most optimal treatment was observed in variation K2 (Moringa seeds at pH 6), while in K1 and K3, the ammonia concentration increased after treatment. Meanwhile, the use of Tamarind seeds at pH 4, 6, and 8 was not effective in reducing ammonia levels. According to Muryanto (2020), ammonia in solution exists as ammonium ions (NH_4^+) at low pH, whereas at alkaline pH it predominantly exists as free ammonia (NH_3). At pH 6, the protein content in Moringa seeds remains relatively stable and is close to its isoelectric point, allowing it to function optimally. This condition is supported by the presence of ammonia in the form of NH_4^+, which enables interactions through adsorption or ionic bonding mechanisms with the cationic proteins in Moringa seeds, resulting in a decrease in the ammonia parameter in the K2 variation.

Compared to Government Regulation No. 22 of 2021, ammonia values in all variations of pH and biocoagulant treatments still do not meet the applicable quality standards. The best reduction in ammonia concentration was obtained in the K2 variation, which used Moringa seeds at pH 6, resulting in a final concentration of 23.38 mg/L (maximum: 0.1 mg/L).

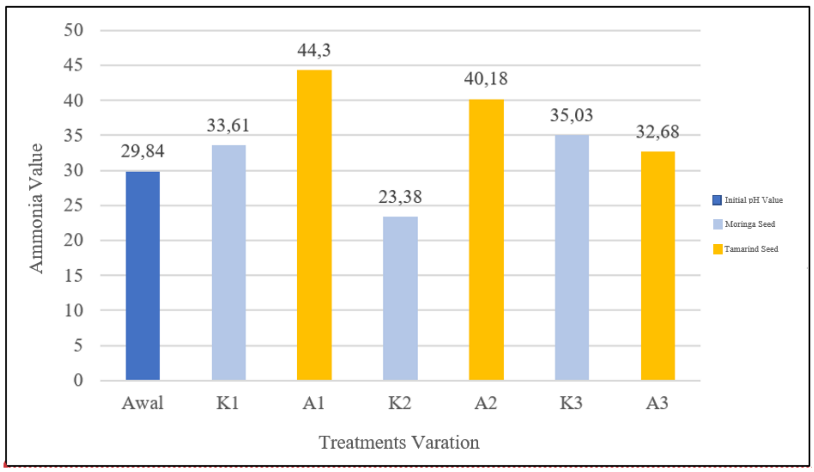


Figure 6. Ammonia Analysis Results

4. CONCLUSION

Based on the results of this study, pH variation has a significant effect on the effectiveness of the coagulation-flocculation process using biocoagulants from Moringa seeds and Tamarind seeds in reducing the quality parameters of tofu wastewater. The optimum pH was obtained at 4 with treatment A1 (Tamarind seeds), which produced the best reduction in TSS and TDS values of 1.25 mg/L and 2.34 mg/L, respectively, along with an increase in DO value up to 1.62 mg/L. The most effective reduction in ammonia concentration was achieved with treatment K2 (Moringa seeds) at pH 6, resulting in ammmonia value of 23.38 mg/L.

In general, Tamarind seeds showed higher effectiveness under acidic conditions, while Moringa seeds worked more optimal under near-neutral conditions. This is due to the release of H^+ ions from protein compounds in the biocoagulant at low pH, which helps neutralize the charge of colloidal particles, thus enhancing the floc sedimentation process. Therefore, pH adjustment prior to the coagulation-flocculation process is an important factor in improving pollutant removal efficiency in tofu wastewater so that the treated water can better approach environmental standards.

Compared to the Indonesian Government Regulation No. 22 of 2021 for a Class 1 water quality standards, all tested parameters did not meet the standards. Therefore, additional treatment stages are required after the coagulation-flocculation process to ensure that the treated water meets the applicable environmental quality standards.

Based on a comparison with chemical coagulants (PAC) as reported by Mursitaningrum et al. (2024), the use of PAC demonstrated better performance in reducing TSS levels, achieving a removal efficiency of 94% at a dosage of 4 g/L. In contrast, this study showed that using Tamarind seed biocoagulant at the same dosage resulted in a TSS reduction of only 72%. Nevertheless, the use of biocoagulants remains a viable option due to their relatively high efficiency and greater environmental friendliness, as the resulting residues are non-toxic and do not contribute additional harmful chemical substances to the environment.

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