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New Modification of Natural Coagulants and Flocculants from Papaya Seeds (*Carica Papaya L.*) and *Aloe Vera* Gel in The Removal of Remazol Yellow Dye

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Abstract

A new modification of natural coagulant and flocculant from papaya seeds (Carica papaya L.) and aloe vera gel has been developed in this study to remove Remazol Yellow dye. The research parameters studied included the initial concentration of the test solution, pH, and stirring speed during the coagulation process. The initial concentration of the test solution varied from 10 to 50 mg/L, the pH ranged from 1 to 6, and the stirring speed was 650 to 1050 rpm during the coagulation phase. The coagulation and flocculation results were characterized using an FTIR spectrophotometer. The results showed that the optimum conditions were an initial concentration of 30 mg/L at pH 2 and a stirring speed of 950 rpm, resulting in the highest removal effectiveness of 97.55%. The results of FTIR spectrophotometry characterization showed that the sulfonate group ($-SO_3^-$) in Remazol Yellow had been removed, indicating that the protonated groups ($-NH_3^+$ and $-COOH_2^+$) in papaya seeds and the anionic groups of the dye had electrostatic interactions. This study shows that the modification of papaya seeds and aloe vera gel as natural coagulants and flocculants shows sustainable potential for azo dye-based textile wastewater treatment.

Keywords: Aloe Vera, Papaya seeds, Coagulation-flocculation, Remazol yellow, Azo dye

1 Introduction

Particularly when it comes to the utilization of technology and raw resources in the manufacturing process, the textile industry is expanding quickly [1]. In 2019, Indonesia's textile sector produced 407.5 billion per month, or 4.89 trillion annually [2]. Through the batik business, South Sulawesi is now working to boost the local population's economy [3]. With regional motifs like South Sulawesi Batik, which uses Toraja carvings, lontara calligraphy, and stylized pinisi ships to represent local cultural identity, batik an Indonesian cultural asset recognized by UNESCO since 2009 [4] has grown into a high-value industry [5]. Originating from the Javanese terms "amba" (*to write*) and "nitik" (*dot*) [6], the creative process continues to use traditional methods, with wax serving as the principal medium [7].

The growth of the batik industry harms the environment [8]. Textile dyes are a significant source of non-biodegradable waste and one of the main ingredients used in the batik business [9]. Remazol yellow is one kind of dye used in South Sulawesi's batik business. One kind of azo dye that is frequently utilized in the textile sector is Remazol Yellow [10]. This dye is resistant to washing and has good color persistence [11]. Remazol Yellow uses a nucleophilic addition process to create covalent connections with fabric fibers [12]. The fabric fibers will emit hydrolyzed or unreacted dyes throughout the soaping and rinsing procedure [13]. However, Remazol is a fast-drying dye due to the covalent link it forms with fabric fibers [14].

Azo dyes are the principal colorants used in textile dyeing and printing because they contain primary aromatic amine groups [15]. In addition to being densely colored, the resulting garbage can harm the environment if improperly managed, particularly in water areas [16]. Because of their complicated structure and durability, many azo dyes are challenging to biodegrade [17].

The coagulation-flocculation process, which is chemical in nature, is one efficient way to treat waste [18]. Wastewater's colloidal particles become unstable during the coagulation stage [19], and then they join to create larger particles during the flocculation stage [20]. A substance known as a coagulant is required in the coagulation-flocculation process to aid in the sedimentation of particles [21]. During the sedimentation process, this coagulant is utilized to neutralize conditions or minimize small particles mixed in water [22].

When synthetic coagulants are used excessively in the environment, environmentally hazardous chemical sludge can accumulate [23]. The pH of the purified water is impacted, and synthetic coagulants are more costly than biocoagulants and environmentally harmful [24]. Biocoagulants are utilized as an alternative to synthetic coagulants to combat the pollution created by dye waste [25]. Because the natural compounds found in biocoagulants are more environmentally benign [26] and perform a crucial and efficient function in the dye removal process [27], the potential of natural resources is used [28]. Relatively inexpensive, widely available, biodegradable, and producing less sludge than synthetic coagulants are only a few benefits of biocoagulants [29].

Papaya seeds' high protein content makes them a natural coagulant. One advantage of using papaya seeds as a natural coagulant is that they are reasonably priced [30], reducing agricultural waste, and allowing for natural decomposition [31]. The use of papaya seeds as a color coagulant has not been widespread [29]. This work lowers the amount of remazol yellow in batik waste by employing the natural coagulant found in papaya seeds. Papaya seeds protein contains an active group called amine (NH_2), which at an acidic pH will be protonated to $-NH_3^+$ and function as a cationic polyelectrolyte [32]. *Aloe vera* flocculant is added to the most recent innovation, the coagulant made from papaya seeds. Similar to cactus plants, *aloe vera* contains mucilage or gel [33], which earlier studies have shown to filter water. *Aloe vera* naturally has a pH of 4-5[34], making it an acidic biocoagulant [35].

The novelty of this research in the use of papaya seeds a natural coagulant and Aloe vera as an environmentally friendly natural flocculant in the removal of yellow Remazol dye. The absorbance of the solution is measured using a UV-visible device and translated into the percentage of dye elimination. The coagulation-flocculation process's ideal parameters are established in order to remove dye from wastewater as quickly as possible.

2 Method

Fig. 1 shows an illustration of the overall method that consists of the coagulation and flocculation processes.

2.1 *Papaya seed coagulant preparation*

After being crushed and dried, papaya seeds are sieved.

2.2 *Preparation of Aloe vera flocculant*

The transparent inner layer (*parenchyma*) of the *aloe vera* is taken off. After centrifugation, the *aloe vera* is mashed, and the supernatant is gathered.

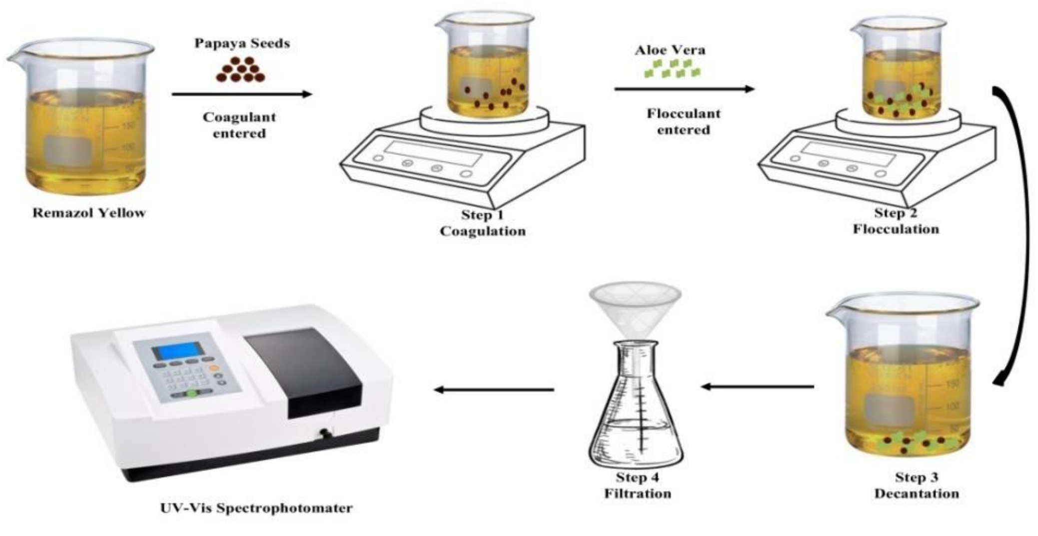

Figure 1 Coagulation-flocculation process illustration

2.3 The impact of the Remazol yellow test solution's concentration on the percentage of the dye that is removed

The initial concentration of the test solution varied from 10, 20, 30, 40, and 50 mg/L. Two milligrams of papaya seeds were then added to the beaker after the solution's pH had been brought down to 2. The Remazol yellow test solution with concentrations of 10, 20, 30, 40, and 50 mg/L was in the beaker. Two phases of stirring were used: five minutes of quick stirring at 950 rpm and thirty minutes of gradual stirring at 125 rpm while adding five grams of *aloe vera* (n = 3). A UV-Vis spectrophotometer was then used to determine absorbance at λ_{max} 417 nm.

2.4 The impact of pH on the percentage of Remazol yellow dye removed

The ideal dosage of 2 mg of papaya seeds was then added to a beaker containing a 30 mg/L Remazol yellow solution after the solution's pH was adjusted to variations 1, 2, 3, 4, 5, and 6 using 0.5 M NaOH to make the pH basic and 0.5 M HCl for the acidic pH. With the addition of 5 g of *aloe vera*, stirring was done in two stages: first, quickly for five minutes at 950 rpm, and then slowly for thirty minutes at 125 rpm (n = 3). A UV-Vis spectrophotometer was then used to determine absorbance at λ_{max} 417 nm.

2.5 The impact of coagulation speed on the percentage of Remazol yellow dye removed

A beaker containing 100 mL of Remazol yellow dye test solution with a concentration of 30 mg/L and adjusted to the ideal pH was filled with 2 mg of papaya seeds. The beaker was set on a stirrer, and the coagulation process was carried out by optimizing the stirring speeds of 650, 750, 850, 950, and 1050 rpm for 5 minutes, followed by slow stirring at a speed of 125 rpm for 30 minutes while adding 5 g of *aloe vera* (n = 3). Additionally, a UV-Vis spectrophotometer was used to evaluate absorbance at λ_{max} 417 nm.

2.6 Fourier Transform Infrared (FTIR) characterization

An FTIR instrument was used to characterize the Remazol yellow dye and the best findings that were obtained.

2.7 Calculation of the removal efficiency of azo dyes in the Remazol yellow test solution at various initial solution concentrations, pH and coagulation speed

Removal efficiency is the percentage decrease in azo dye content during the coagulation process, where C is the final concentration of azo dye in the Remazol red test solution following coagulation (mg/L) and C_0 is the initial concentration of azo dye measured without any coagulation process (mg/L).

$$\text{Removal Efficiency (\%)} = \frac{(Co-C)}{Co} \times 100\% \quad \text{(Equation 1)}$$

3 Result and Discussion

3.1 Effect of the initial concentration of the test solution on the percentage of Remazol yellow dye removal

The colloidal particles in the solution, which are at the perfect concentration to interact with the papaya seed coagulant mass during the coagulation and flocculation process, are impacted by the Remazol yellow dye test solution's optimal concentration. The neutralization of the colloidal particles' charge is essential to the coagulation process because it enables the dye and coagulant to clump together and form flocs. The elimination efficiency of the Remazol yellow dye was 61.36%, 66.67%, 97.55%, 91.58%, and 85.58% at the optimal concentration of 10, 20, 30, 40, and 50 mg/L of the test solution.

Fig. 2 shows the graph of the correlation between the percentage of Remazol yellow dye removal and the concentration of Remazol yellow solution. The experiment's results are listed in **Table 1**, where the removal efficiency of Remazol yellow dye rises in tandem with the test solution's concentration, reaching its maximum efficiency of 97.55% at a concentration of 30 mg/L. At a concentration of 40 mg/L, the effectiveness of removing Remazol yellow dye declines after reaching the ideal value of 30 mg/L. According to several studies, an excessive amount of Remazol yellow dye can decrease the effectiveness of removal.

Table 1 Effect of the initial concentration of the test solution on the percentage of Remazol yellow dye removal

Concentration of Test Solution	% Removal 1	% Removal 2	% Removal 3	Average	% Error	% Thoroughness
10 mg/L	61.33	61.64	61.11	61.36 ± 0.27	0.44%	99.56%
20 mg/L	66.44	66.90	66.67	66.67 ± 0.23	0.34%	99.66%
30 mg/L	**97.72**	**97.25**	**97.70**	**97.55 ± 0.26**	**0.27%**	**99.73%**
40 mg/L	91.96	91.20	91.58	91.58 ± 0.38	0.42%	99.58%
50 mg/L	85.11	85.99	85.63	85.58 ± 0.44	0.52%	99.48%

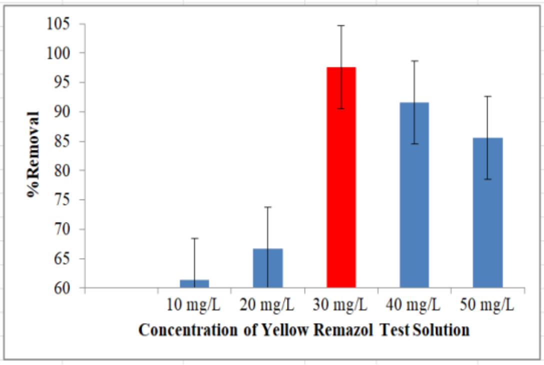

Figure 2 Effect of the initial concentration of the test solution on the percentage of Remazol yellow dye removal

The decrease in this study is believed to be caused by the accumulation of Remazol yellow dye molecules on the papaya seed coagulant's surface, which creates a repulsive force between the molecules with the same charge and eventually prevents more absorption of the dye. However, because there aren't enough Remazol yellow dye molecules to exploit all of the coagulant's active surfaces, the removal effectiveness is likewise limited at low concentrations of the dye. Thus, particle bridging or charge neutralization loses some of its effectiveness. The following experiment will determine the ideal concentration of the Remazol yellow dye test solution, which is 30 mg/L.

3.2 Effect of the initial pH of the test solution on the percentage of Remazol yellow dye removal

Fig. 3 shows a graph of the correlation between pH and the percentage decrease in Remazol yellow concentration utilizing natural flocculant from *aloe vera* and coagulant from papaya seeds. The highest removal percentage was at pH 2 (**Table 2**).

Most of the proteins found in papaya seeds are made up of amino acids. Amino acid groups will undergo protonation in acidic settings to become positively charged (-NH_3^+ and -$COOH_2^+$), and deprotonation in basic conditions to become –NH^- and –COO^-.

Table 2 Effect of the initial pH of the test solution on the percentage of Remazol yellow dye removal

pH	% Removal 1	% Removal 2	% Removal 3	Average	% Error	% Thoroughness
1	60.56	60.28	60.09	60.31 ± 0.24	0.39%	99.61%
2	**97.26**	**97.71**	**97.26**	**97.41 ± 0.26**	**0.26%**	**99.74%**
3	93.64	93.15	93.12	93.30 ± 0.29	0.31%	99.69%
4	85.25	85.65	85.12	85.34 ± 0.28	0.32%	99.68%
5	59.09	59.63	59.72	59.48 ± 0.34	0.57%	99.43%
6	40.38	40.19	40.93	40.50 ± 0.39	0.95%	99.05%

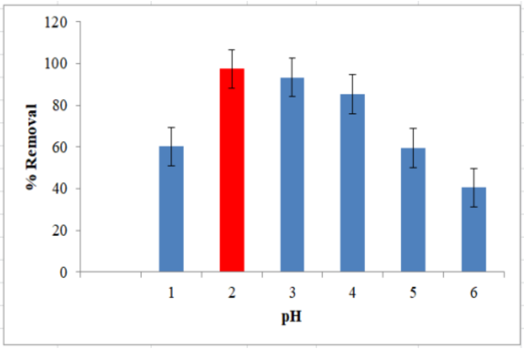

Figure 3 Effect of the initial pH of the test solution on the percentage of Remazol yellow dye removal

This behavior is closely related to the protonation state of functional groups present in papaya seed proteins. In the acidic pH range of 2–6, the amine groups are primarily protonated to form $–NH_3^+$, which serves as the dominant positively charged site responsible for the electrostatic attraction with the $–SO_3^–$ group of Remazol Yellow. Although the carboxyl groups can also undergo protonation under highly acidic conditions, their contribution is much smaller compared to $–NH_3^+$. As the pH increases, these functional groups progressively deprotonate ($–NH_3^+ \rightarrow NH_2$ and $–COOH \rightarrow COO^–$), reducing the net positive charge on the bio coagulant. This loss of charge weakens the electrostatic interaction with the anionic dye, thus decreasing the removal efficiency. Therefore, the optimum pH of 2 observed in this study can be attributed to the maximum protonation of the amine groups, which facilitates strong dye-coagulant interactions.

In this study, aloe vera gel acted as a flocculant to promote the formation of larger flocs. From the results of preliminary experiments, the removal efficiency of Remazol yellow dye using papaya seeds as a coagulant obtained % Removal of 83,69%, using aloe vera alone as a flocculant obtained % Removal of 72,22%. At optimum conditions obtained % Removal of 97,41% resulting from the modification of papaya seeds and aloe vera as natural coagulants-flocculants. The modification resulted in a higher removal efficiency, which confirms the synergistic effect where papaya seeds act as the main charge neutralizing coagulant while aloe vera increases the size and settling of flocs.

3.3 *Effect of the initial coagulation speed of the test solution on the percentage of Remazol yellow dye removal*

For the coagulant to be distributed uniformly throughout the solution, the coagulation rate is essential. By doing this, the repulsive interactions that exist between the Remazol yellow dye and the coagulant particles are lessened.

Table 3 Effect of the initial coagulation speed of the test solution on the percentage of Remazol yellow dye removal

Coagulation Rate	% Removal 1	2	3	Average	% Error	% Throughness
650	86.82	86.30	86.70	86.60 ± 0.27	0.31%	99.69%
750	88.58	88.99	88.94	88.84 ± 0.22	0.25%	99.75%
850	91.28	91.67	91.67	91.54 ± 0.22	0.24%	99.76%
950	**97.24**	**97.69**	**97.66**	**97.53 ± 0.25**	**0.26%**	**99.74%**
1050	90.61	90.65	90.23	90.50 ± 0.23	0.26%	99.74%

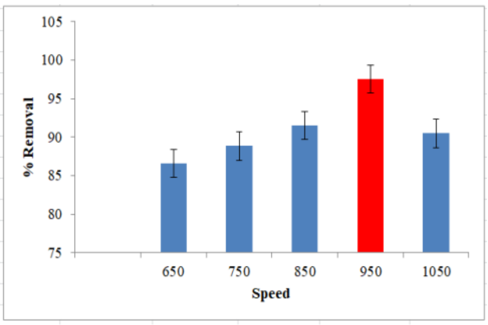

Figure 4 Effect of the initial coagulation speed of the test solution on the percentage of Remazol yellow dye removal

The removal efficiency of Remazol yellow dye was 86.61%, 88.84%, 91.54%, 97.53%, and 90.50% after five minutes of rapid stirring at an optimal speed of 650, 750, 850, 950, and 1050 rpm. **Fig. 4** shows the graph of the correlation between coagulation speed and the percentage of Remazol yellow dye elimination. According to the experimental findings, a higher coagulation speed is linked to a higher rate of dye removal, as shown by the % removal value over time in **Table 3**. A coagulation speed of 950 rpm produced the best removal percentage, 97.53%.

Previous research using synthetic coagulant APTES and alcohol modification to remove yellow Remazol dye resulted in a removal percentage of 58.2%, while the removal percentage produced in this study for removing yellow Remazol dye was 97.55%. This indicates that natural coagulants have the potential as an alternative to synthetic coagulants in removing yellow Remazol dye [36].

This rise in the percentage of elimination is crucial because it speeds up particle binding and creates bigger aggregates, which facilitates their separation from the dye. However, the removal % decreased when the stirring speed was increased to 1050 rpm after it had reached the optimal threshold. This happens because the floc breaks down into smaller particles due to coagulation that occurs too quickly beyond a specific limit. The dye clearance rate is adversely affected by these tiny particles because they are more challenging to settle.

3.4 Fourier Transform Infrared (FTIR) characterization

The functional groups in yellow and ideal Remazol were identified using an FTIR spectrophotometer.

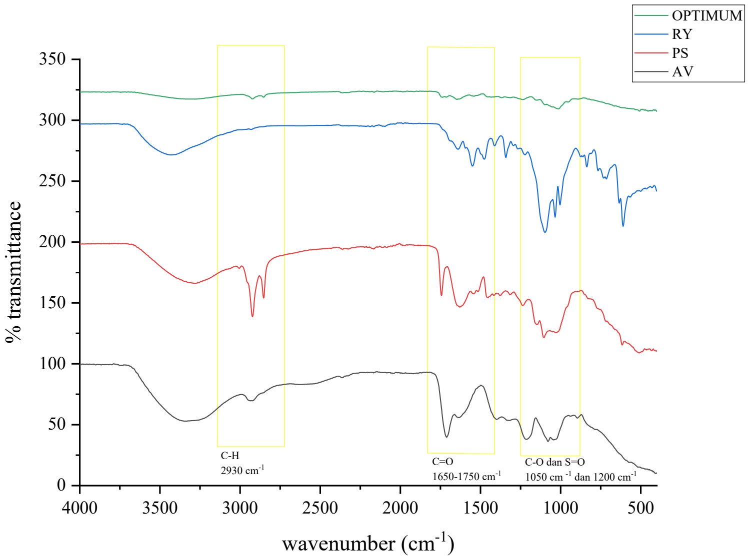

Figure 5 FTIR spectrum of Aloe Vera (AV), PS (Papaya Seeds), Remazol yellow (YR) dye and Optimum

To confirm that the Remazol yellow dye removal process had taken place, the FTIR characterization that was obtained was compared to the FTIR spectrum of the Remazol yellow dye test solution. FTIR characterization in the 4000-400 cm^{-1} wave number range.

Fig. 5 displays the FTIR spectra. The coagulant and flocculant characterization results indicate that before the coagulation-flocculation process, the FTIR spectrum of papaya seeds identified N-H vibrations. Meanwhile, in aloe vera, O-H bonds from hydroxyl groups, asymmetric stretching of the C=O double bond in the carboxylate group, and C-O bonds from alcohols and ethers were detected. After the coagulation-flocculation process in RK + BP + AV, absorption of the N-H group was still detected, there was a shift in absorption in the C=O and C-O groups, and the optimum sulfonate group detected in the Remazol yellow dye was no longer identified. Thus, the coagulation-flocculation procedure successfully removed the Remazol yellow dye.

4 Conclusion

Based on the investigation, the novel combination of papaya seeds and *aloe vera* gel proves to be a highly effective, dual-function agent for treating Remazol Yellow dye in wastewater. The study successfully identified the critical operational parameters for maximum dye removal. The most favorable conditions were achieved with a dye concentration of 30 mg/L at a pH of 2, coupled with a rapid mixing speed of 950 rpm during coagulation. Under this optimized regime, the process achieved a remarkable removal efficiency of 97.55%. The underlying mechanism is primarily driven by electrostatic interactions. In the acidic environment, the proteinaceous components within the papaya seeds become protonated, acquiring a positive charge. This allows them to attract and neutralize the negatively charged sulfonate groups ($-SO_3^-$) present on the Remazol Yellow dye molecules. The subsequent addition of aloe vera gel aids in forming larger, settleable flocs. FTIR analysis confirmed the success of this process, showing the effective removal of the dye's characteristic functional groups from the treated water. Consequently, this research demonstrates that a modified natural coagulant-flocculant system, utilizing agricultural resources, presents a viable, sustainable, and efficient alternative to synthetic chemicals for purifying textile wastewater containing azo dyes.

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