

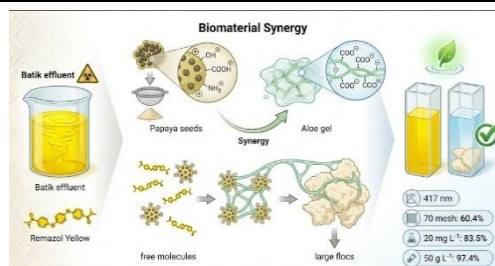
## The Effect of Coagulant Particle Size, Coagulant and Flocculant Dose on Remazol Yellow Dye Removal Effectiveness

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### ABSTRACT

Textile dye effluents are hazardous because many residual dyes are persistent, toxic, and potentially carcinogenic. This study assessed an environmentally friendly coagulation flocculation route to remove Remazol Yellow, an azo dye used in the batik industry in South Sulawesi, by substituting synthetic reagents with papaya (*Carica papaya*) seeds as a coagulant and Aloe vera gel as a flocculant. FTIR confirmed hydroxyl, carboxyl, and amine groups in papaya seeds, while Aloe vera gel was dominated by polysaccharide and carboxylate features. Jar tests were performed at room temperature using an initial dye concentration of 30 mg/L. Coagulation was conducted for 5 min at 950 rpm, followed by flocculation for 30 min at 125 rpm and sedimentation for 30 min. Residual dye was measured by UV Vis spectrophotometry at 417 nm. Operational variables were optimized, including coagulant particle size (30 to 100 mesh), papaya seed dose (10 to 50 mg/L), and Aloe vera gel dose (10 to 60 g/L). The optimum particle size was 70 mesh, giving 60.4 percent removal. The optimum papaya seed dose was 20 mg/L with 83.5 percent removal, and the optimum Aloe vera gel dose was 50 g/L, achieving 97.4 percent removal. These results demonstrate the potential of papaya seeds and Aloe vera gel as effective and sustainable materials for batik dye wastewater treatment.



**Keywords:** Adsorption; Montmorillonite; Dyes; Molecular Dynamics; GROMACS..

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## INTRODUCTION

The batik sector plays an important role in preserving Indonesian cultural heritage and supporting national economic growth. Expansion of the batik industry has occurred through both small scale enterprises and large scale production systems [1]. The rapid development of the batik industry has caused ecological degradation, especially due to the disposal of textile dyes which are the main component in

batik production and are the main source of non-biodegradable liquid waste [2]. According to the latest Minister of Environment and Forestry Regulation No. P.16 of 2019, states that dyestuff is a colorant that can be used to color textile materials and can be made from either natural or synthetic sources [3]. Textile waste is particularly hazardous due to its carcinogenic and toxic properties because it is difficult to degrade and typically originates from residual dyes, which are complex aromatic compounds in

the form of azo compounds and derivatives of the benzene group [4].

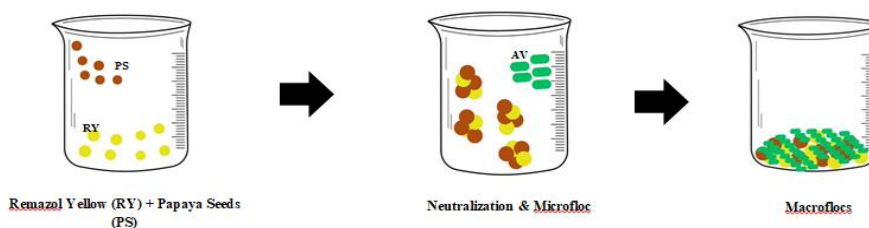
Remazol yellow is one kind of dye used in South Sulawesi's batik business. An azo dye called Remazol yellow is used to dye textiles since it is simple to apply, has a consistent hue, and doesn't wash off. Azo dyes are suitable as the primary dye in the textile dyeing and printing industry because they include a primary aromatic amine group [5]. The resulting wastewater is dark in color and, if improperly managed, will negatively impact the environment, particularly in water areas [6]. Because of their complicated structure and resilience, the majority of azo dyes are challenging to biodegrade [7].

Azo dyes pose serious risks to human health and aquatic ecosystems. Mutagenic and carcinogenic effects have been reported, alongside potential damage to organs such as the liver and kidneys [8]. Additionally, water-contaminating azo dyes decrease light penetration, which prevents photosynthesis and interferes with aquatic ecosystems natural growth [9]. Thus, to lessen adverse environmental effects, an efficient textile dye removal method is required [10].

Coagulation and flocculation represent one practical approach for reducing textile dye concentrations in wastewater [11]. In order to combine colloidal and suspended

materials to create flocs that can be deposited, coagulation-flocculation is the process of adding a chemical reagent to a liquid [12]. The mechanism by which the addition of coagulants and flocculants destabilizes colloidal particles is known as the coagulation-flocculation principle [13]. Among the benefits of the coagulation method are its ease of use, affordability, and capacity to absorb organic contaminants like colors [6].

Synthetic coagulants are associated with several drawbacks, including high production costs, non-biodegradability, and potential health risks such as neurotoxicity linked to aluminum-based coagulants, which have been implicated in neurodegenerative disorders like Alzheimer's disease [14]. To mitigate these environmental and health concerns while reducing treatment costs, natural coagulants derived from plant-based sources have gained increasing attention as sustainable alternatives [15]. Among various natural coagulants explored for wastewater treatment, papaya seeds (*Carica papaya* L.) have shown promising potential due to their high protein content, which includes cationic polyelectrolytes capable of neutralizing negatively charged dye molecules [16]. The interaction of coagulant-flocculants with the yellow dye Remazol is depicted in Figure 1.



**Figure 1.** Illustration of the interaction of coagulant-flocculant with the yellow dye Remazol

The concentration of yellow remazol in batik waste can be decreased by employing papaya seeds as a natural coagulant to remove yellow remazol from textile dyes. Papaya seeds' protein contains active groups called amines ( $\text{NH}_2$ ), which operate as cationic polyelectrolytes by protonating to  $\text{NH}_3^+$  at an acidic pH [17]. *Aloe vera* gel contains polysaccharides, mucilage, and complex carbohydrates that act as natural polymeric flocculants by bridging destabilized colloidal particles, thereby enhancing floc formation and sedimentation [18]. *Aloe vera* acts as a natural flocculants due to its high content of mucilage and polysaccharides such as acemannan which function as polymeric bridging agents that bind suspended particles in water, promoting floc formation and sedimentation [19].

Colloidal instability causes the Remazol yellow dye to coagulate. The sulfonate group ( $\text{SO}_3^-$ ) in the dye and the papaya seeds have different charges, which causes the release of a positively charged  $\text{H}^+$  ion and initiates the colloidal destabilization process. The particles are able to bond because of the charge neutralization brought about by this difference in charges. Microagglomerates consequently merge to create bigger macroagglomerates. Polysaccharides are the primary constituent of *Aloe vera* gel [19]. In the polysaccharide-particle complex, the *Aloe vera* polysaccharide main chain and side chain groups create bridges [20]. Colloidal particles are joined into bigger aggregates by polysaccharides, which

function as binders. Large flocs that form become heavier, which makes them simpler to filter or allows them to settle more quickly [21]. Because it contains complex sugars and carbohydrates that can bind particles in water and has a mucilage (gel) content similar to cacti, which can be utilized as a water purifier, *Aloe vera* was selected as a flocculants.

This study aims to evaluate the performance of a natural coagulant-flocculants system specifically, papaya seeds (*Carica papaya* L.) as coagulant and *Aloe vera* gel as flocculants for the removal of Remazol Yellow dye, with particular attention to the combined effects of coagulant particle size and coagulant-flocculant dosage. This approach addresses a research gap in optimizing fully natural, plant-based systems for azo dye removal.

## METHODS

### 1. Chemicals

Hydrochloric acid ( $\text{HCl}$  37%, Merck, Cat. No. 1.00317), sulfuric acid ( $\text{H}_2\text{SO}_4$ , 99%, Merck, Cat. No. 1.00731), and deionized water (resistivity:  $18.2 \text{ M}\Omega\cdot\text{cm}$ , produced using a Milli-Q® IQ 7000 system) were used as received. Filter paper Universal pH indicator paper (range: 1–14, Merck, Cat. No. 109535) was employed to estimate the pH of solutions prior to adjustment. Remazol Yellow dye ( $\text{C}_{18}\text{H}_{13}\text{N}_2\text{S}_4\text{O}_{12}\text{Na}_3$ , 99% purity) was used as the model pollutant. Papaya seed powder was prepared from *Carica papaya* L. seeds collected from local markets in Malang, East Java, Indonesia; seeds were dried, ground,

and sieved to the required particle sizes. Fresh gel was extracted from the parenchyma tissue of *Aloe barbadensis* Mill. (syn. *Aloe vera*) from Malang flower market.

## 2. Instruments

A UV-Vis Spectrophotometer (Genesys 10S UV-Vis), Fourier Transform Infra-Red (Shimadzu 8400s), an oven (Mettler UN30, Mettler GmbH, Germany), and a magnetic stirrer were among the tools utilized in this investigation.

## 3. Preparation of Natural Coagulants and Flocculants

Papaya seeds collected from local markets in Malang, East Java, Indonesia were washed with distilled water and dried at room temperature. Dried seeds were then oven heated at 110°C for 2 h to remove residual moisture and stabilize sample mass. Dried material was ground and sieved to obtain the required particle size fractions. Functional groups associated with coagulation activity, including hydroxyl (OH), carboxyl (COOH), and amine (NH<sub>2</sub>), were identified using FTIR (Shimadzu 8400s) to support mechanistic interpretation of charge neutralization and electrostatic interactions with anionic dye molecules.

Fresh gel was extracted from the parenchyma tissue of *Aloe barbadensis* Mill. (syn. *Aloe vera*) from Malang flower market. *Aloe vera* was cut into small square pieces, and the inner white part (parenchyma) was collected. The parenchyma layer was then crushed and centrifuged at 4000 rpm. The gel-like supernatant was collected and stored at 4°C. The *aloe vera* gel was

analyzed using an FTIR spectrophotometer (Shimadzu 8400s) to identify functional groups such as hydroxyl (–OH), carboxylate (–COO<sup>–</sup>), and polysaccharide moieties that are responsible for its flocculant activity through polymer bridging and interparticle aggregation.

## 4. Preparation of Dye Solutions

50 mg of Remazol yellow (C.I. Reactive Yellow 145; chemical formula: C<sub>18</sub>H<sub>13</sub>N<sub>2</sub>S<sub>4</sub>O<sub>12</sub>Na<sub>3</sub>; purity 99%) was dissolved in 500 mL of distilled water (approximately 25 °C) to create the Remazol yellow stock solution. For use as standard solutions, the stock solution was diluted to concentrations of 10, 20, 30, 40, and 50 ppm.

## 5. Determination of Maximum Wavelength

An aliquot of 3 mL from the 100 mg/L Remazol Yellow stock solution was pipetted into a 10 mL volumetric flask and diluted to the mark with distilled water to obtain a 30 mg/L solution. The diluted solution was scanned from 400 to 800 nm using a UV Vis spectrophotometer, and the wavelength at maximum absorbance was selected for subsequent measurements.

## 6. Standard Curve Preparation

Standard solutions of Remazol Yellow at 10, 20, 30, 40, and 50 ppm were measured at the selected maximum wavelength using a UV Vis spectrophotometer. A calibration curve was constructed by plotting concentration against absorbance, and the linear

regression equation and coefficient of determination ( $R^2$ ) were obtained using Microsoft Excel.

## 7. Coagulation-Flocculation

### Experiments

#### Effect of coagulant particle size on dye removal

Papaya seed powder was sieved to particle sizes of 30, 60, 70, 80, and 100 mesh. Papaya seed powder (0.1 mg) was added to a 30 mg/L Remazol Yellow test solution adjusted to pH 2. Mixing was performed in two stages using a magnetic stirrer: rapid mixing for 5 min at 950 rpm followed by slow mixing for 30 min at 125 rpm. Triplicate measurements were performed ( $n = 3$ ) at room temperature. Settling was carried out for 30 min, supernatant was filtered, and absorbance was measured at the maximum wavelength.

#### Effect of coagulant dosage on dye removal

Papaya seed powder at the optimal particle size identified previously was added to 30 mg/L Remazol Yellow solution at pH 2 using dosages of 10, 20, 30, 40, and 50 mg/L. Rapid mixing was conducted for 5 min at 950 rpm and slow mixing was conducted for 30 min at 125 rpm. Triplicate measurements were performed ( $n = 3$ ) at room temperature. Settling was conducted for 30 min, supernatant was filtered, and absorbance was measured at the maximum wavelength.

#### Effect of flocculant dosage on dye removal

Papaya seed powder at the optimal particle size and dosage was added to 30 mg/L Remazol Yellow solution at pH 2. Rapid mixing was performed for 5 min at 950 rpm. Slow mixing was continued for 30 min at 125 rpm, and Aloe gel was added at the start of the slow mixing stage using dosages of 10, 20, 30, 40, 50, and 60 g/L. Triplicate measurements were performed ( $n = 3$ ). Settling was carried out for 30 min, supernatant was filtered, and absorbance was measured at the maximum wavelength.

## 8. Data Analysis

Removal efficiency was calculated to quantify the percentage of Remazol Yellow azo dye eliminated during the coagulation flocculation process using the following equation:

$$\text{Removal Efficiency (\%)} = \frac{(C_0 - C)}{C_0} \times 100\%$$

$C_0$  represents the initial dye concentration measured in the absence of any coagulation treatment (mg/L), while  $C$  represents the final dye concentration measured after coagulation flocculation (mg/L). Final concentration values were obtained from UV Vis measurements by applying the calibration curve equation.

All results were reported as mean  $\pm$  standard deviation from three replicates under identical conditions ( $n = 3$ ). Normality and homogeneity of variance were confirmed using the Shapiro Wilk and Levene tests ( $p > 0.05$ ). Descriptive analysis and plotting were performed in Microsoft Excel, while one way ANOVA and LSD post hoc tests were conducted in IBM SPSS

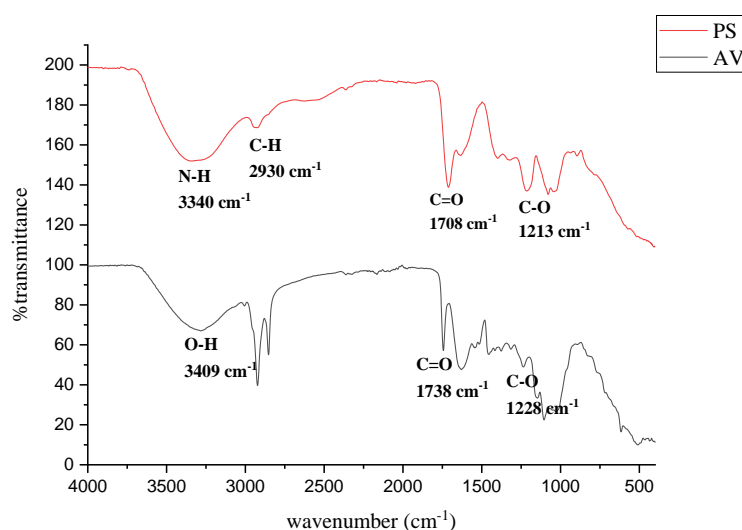
Statistics 22. Exact p values are provided in the Results section.

## RESULTS AND DISCUSSION

### 1. Functional Group Characterization of *Aloe Vera* and Papaya Seeds by FTIR

Functional groups in papaya and *Aloe vera* seeds were identified using FTIR spectroscopy. Figure 2 presents the FTIR spectra obtained in this study, showing the main absorption bands observed for both seed samples. A broad band at  $3340\text{ cm}^{-1}$  corresponds to O–H and N–H stretching vibrations, while the peak at  $1708\text{ cm}^{-1}$  is assigned to C=O stretching of carboxylic acid (–COOH) groups.

The presence of absorption –C–H is indicated by the appearance of an absorption band at the corresponding wavenumber  $2930\text{ cm}^{-1}$  and CO vibrations at peaks of  $1213\text{ cm}^{-1}$  is typically for C–O stretches. Our observed –OH and –NH stretching vibration at  $3340\text{ cm}^{-1}$  is comparable to the  $3438.8\text{ cm}^{-1}$  reported by [22] for papaya seeds. The identification of carboxyl, hydroxyl, and amide groups in our samples via FTIR, consistent with their established role as active components in coagulants [23], supports the potential of papaya and *Aloe vera* seeds for coagulation



**Figure 2.** FTIR spectra of *Aloe Vera* (AV) and Papaya Seeds (PS)

The presence of OH from the hydroxyl group from the water solvent is shown by a peak at wave number  $3409\text{ cm}^{-1}$  is an intrinsic component of *Aloe vera* in the characterization of natural *Aloe vera* flocculants. Asymmetric stretching and C=O double bonds are present in the carboxylate group, as indicated by the peak at wave

number  $1738\text{ cm}^{-1}$ . Indicates the peak at wave number  $1228\text{ cm}^{-1}$  is attributed to C–O stretching vibrations in polysaccharide structures, consistent with the presence of glycosidic linkages and sugar ring moieties. That both proteins and polysaccharides are known flocculant substances, and

polysaccharides were identified in the samples [24], [25].

## 2. Determination of Maximum Wavelength and Establishment of Standard Curve

Visible light spectrophotometry was performed over a wavelength range of 400 to 800 nm using distilled water as the blank. The Remazol Yellow solution showed a maximum absorbance of 0.404 at 417 nm.

## 3. Creating the yellow Remazol standard curve

A calibration curve was constructed using Remazol Yellow concentrations of 0, 10, 20, 30, 40, and 50 mg/L. The corresponding absorbance values were 0.000, 0.065, 0.137, 0.214, 0.289, and 0.358, respectively. Linear regression analysis produced the calibration equation  $y = 0.0071x$  with  $R^2 = 0.9991$ , indicating excellent linearity across the tested range. The resulting standard curve is presented in [Figure 3](#), and this equation was applied to determine Remazol Yellow concentrations in samples after the coagulation flocculation process.

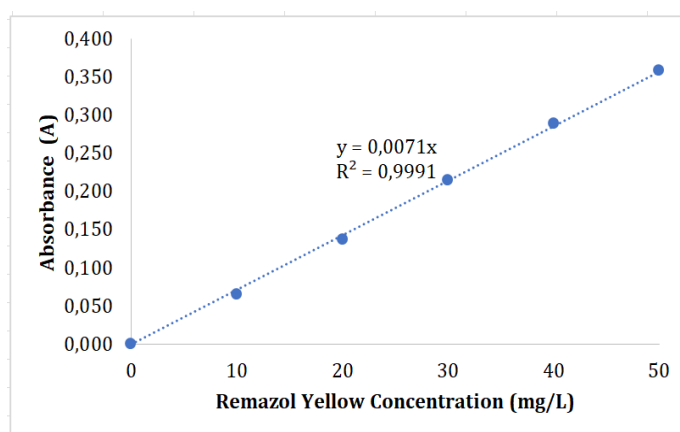


Figure 3 . Standard Remazol yellow dye curve

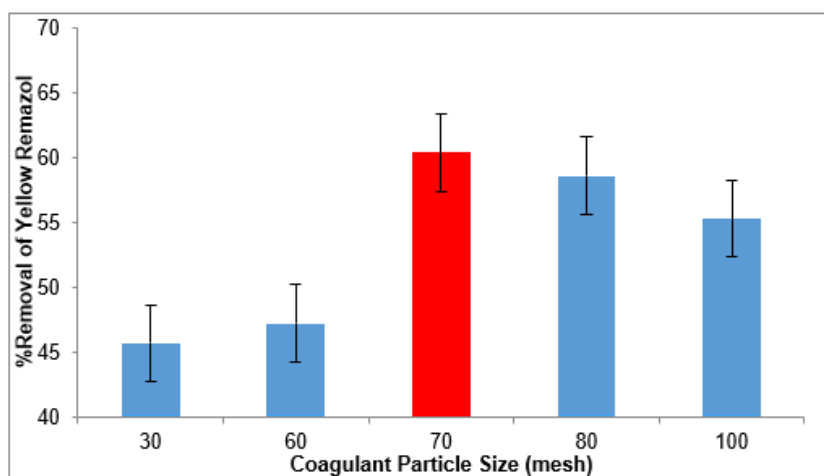
## 4. The impact of coagulant particle size on Remazol yellow dye removal effectiveness

Papaya seed powder was evaluated as a natural coagulant to determine the optimum coagulation condition for Remazol Yellow removal. Particle size was varied at 30, 60, 70, 80, and 100 mesh while other experimental conditions were kept constant. Differences in coagulant particle size influenced dye removal performance, indicating that physical characteristics of the

coagulant contribute to the efficiency of colloid destabilization and floc formation.

[Figure 4](#) presents the relationship between coagulant particle size and the percentage removal of Remazol Yellow. Removal increased from 45.66% at 30 mesh to 47.17% at 60 mesh, then reached the highest value at 70 mesh (60.34%). Removal subsequently decreased to 58.56% at 80 mesh and 55.27% at 100 mesh. The pattern indicates a size dependent optimum, where increasing fineness improves performance up to a

threshold, followed by reduced efficiency at higher mesh sizes.



**Figure 4.** Impact of coagulant particle size on Remazol yellow dye removal effectiveness

**Table 1.** Impact of coagulant particle size on Remazol yellow dye removal effectiveness

Particle Size (mesh)	% Removal	SD	Type Errors (%)	Calculation Method For Thoroughness (%)
30	45,66	0,40	0,87	99,13
60	47,17	0,27	0,58	99,42
70	60,34	0,33	0,54	99,46
80	58,56	0,44	0,75	99,25
100	55,27	0,23	0,42	99,58

The optimum performance at 70 mesh suggests that this particle size provides a favorable balance between accessible active sites and effective floc formation. Very coarse particles tend to provide fewer contact points and lower interaction probability with dye molecules, which can limit charge neutralization and aggregation. Very fine particles can aggregate among themselves or form dense suspensions that reduce the availability of active functional sites for interaction with dye molecules, which may hinder the development of large, settleable flocs and reduce apparent removal.

[Table 1](#) summarizes the numerical results in [Figure 4](#), including mean removal, standard deviation, type error, and the reported thoroughness calculation. The 70 mesh condition produced the highest average removal with low variability, supporting selection of this particle size for the next experimental phase. The subsequent coagulation flocculation optimization should therefore use 70 mesh as the fixed coagulant particle size to evaluate dosage effects and coagulant flocculant combinations.

#### **5. The impact of papaya seed coagulant dose on yellow Remazol dye removal effectiveness**

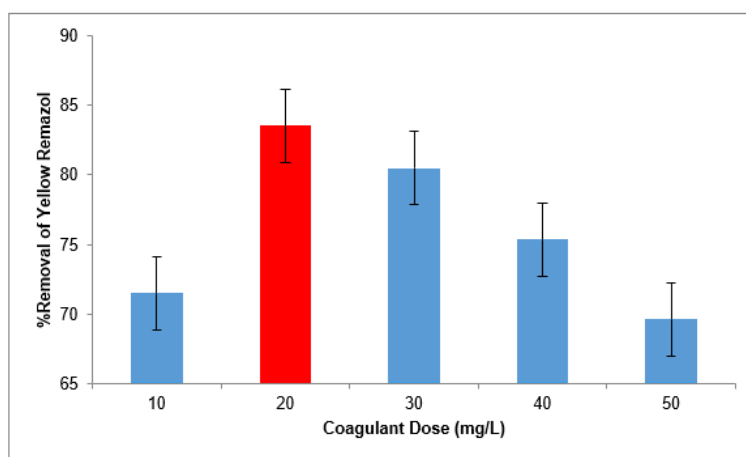


The most efficient amount of coagulant to treat wastewater and meet the required quality criteria is known as the optimal coagulant mass. The ideal circumstances for the coagulation-flocculation process depend heavily on the

coagulant mass. The coagulation-flocculation process, as well as later procedures like sedimentation and filtration, will be interfered with if the mass is not adequately managed.

**Table 2.** Impact of papaya seed coagulant dosage on yellow Remazol dye removal effectiveness

Dose (mg/L)	% Removal	SD	Type Errors (%)	Calculation Method For Thoroughness (%)
10	71,50	0,42	0,58	99,42
20	83,54	0,39	0,47	99,53
30	80,49	0,19	0,24	99,76
40	75,38	0,37	0,49	99,51
50	69,62	0,31	0,45	99,55



**Figure 5.** Impact of papaya seed coagulant dosage on yellow Remazol dye removal effectiveness

The results of this study are presented in [Figure 5](#), which shows that the percentage removal of Remazol Yellow increased as the initial dye concentration rose from 10 mg/L, reached a maximum at 20 mg/L, and then gradually decreased at higher concentrations. Consistent with this trend, [Table 2](#) indicates that the optimum performance occurred at the 20 mg/L condition, after which the removal efficiency declined.

Lower removal at dye concentrations below the optimum can be attributed to

suboptimal charge neutralization and particle bridging, meaning that floc formation was not yet effective. In other words, the coagulant dosage and collision frequency were insufficient to neutralize the negatively charged dye species and promote stable floc growth. Conversely, the decrease in removal at higher dye concentrations suggests that the coagulant became insufficient relative to the increased pollutant load, leaving more dye molecules unneutralized and reducing the formation of settleable flocs [26]. This drop happened because flocs were

prevented from forming because of the repulsive interaction between the papaya seed coagulant and the yellow Remazol dye, which caused the particles to stabilize once more. In the following trials, the ideal coagulant mass of 2 mg will be employed.

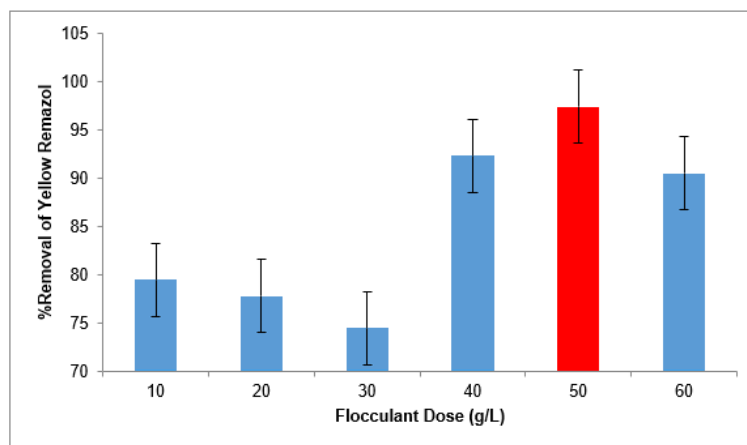
#### 6. The impact of *Aloe Vera* flocculant dosage on yellow Remazol dye removing effectiveness

*Aloe vera* gel dosage was varied from 10 to 60 g/L to evaluate its effect on Remazol

Yellow removal. [Figure 6](#) shows that removal efficiencies were 79.48%, 77.79%, 74.51%, 92.35%, 97.40%, and 90.49% at dosages of 10, 20, 30, 40, 50, and 60 g/L, respectively. The highest removal efficiency was achieved at 50 g/L, indicating this dosage as the optimum flocculant concentration under the tested conditions. [Table 3](#) summarizes the mean removal values along with standard deviation, type error, and calculation thoroughness for each dosage.

**Table 3.** Effect of dosage of *Aloe vera* flocculants on Remazol yellow dye removing effectiveness

Dose (g/L)	% Removal	SD	Type Errors (%)	Calculation Method For Thoroughness (%)
10	79,48	0,19	0,24	99,76
20	77,79	0,24	0,31	99,69
30	74,51	0,40	0,54	99,46
40	92,35	0,24	0,26	99,74
50	97,40	0,26	0,27	99,73
60	90,49	0,25	0,28	99,72



**Figure 6.** Effect of dosage of *Aloe vera* flocculants on Remazol yellow dye removing effectiveness

A decrease in removal efficiency was observed when the dosage increased from 50 to 60 g/L, as shown in Figure 6 and Table 3. Excess flocculant can cause

restabilization through repulsive interactions associated with excess surface charge, which disrupts floc formation and may trigger deflocculation. Increased viscosity at higher

dosages can also reduce particle collision frequency and limit polymer bridging, thereby lowering dye removal performance. Optimum performance is therefore associated with a dosage that supports charge neutralization and interparticle bridging without inducing repulsion or restabilization.

## CONCLUSION

This study successfully showed that *Aloe vera* and papaya seeds can act as effective natural agents in removing Remazol yellow through a coagulation-flocculation process. This study was conducted at pH 2 and room temperature. The high removal effectiveness of 97.405%, was obtained at a papaya seed coagulant particle size of 70 mesh with a dosage of 20 mg/L, and an *Aloe vera* flocculants dosage of 50 g/L. This natural coagulant-flocculants system is very promising for use in small and medium-scale batik businesses because it provides an economical, environmentally friendly, and sustainable alternative to traditional synthetic chemicals. Future research directions need to be carried out by scaling up the process, testing with different dyes, investigating long-term stability and assessing full-scale economic and environmental impacts.

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